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**Changes in surface ozone in South Korea on diurnal to decadal time scale  
for the period of 2001-2021**

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Short title: Ozone changes in South Korea

## Abstract

Several studies have reported an increasing trend of surface ozone in South Korea over the past few decades, using different measurement metrics. In this study, we examined the surface ozone trends in South Korea by analyzing the hourly or daily maximum 8-hour average ozone concentrations (MDA8) measured at the surface from 2001 to 2021. We studied the diurnal, seasonal, and multi-decadal variations of this parameter at city, province, and background sites.

We found that the 4th highest MDA8 values exhibited positive trends in 7 cities, 9 provinces, and 2 background sites from 2001 to 2021. For the majority of sites, there was an annual increase of approximately 1-2 ppb. After early 2010, all sites consistently recorded MDA8 values exceeding 70 ppb, despite reductions in precursor pollutants such as NO<sub>2</sub> and CO. The diurnal and seasonal characteristics of ozone exceedances, defined as the percentage of data points with hourly ozone concentrations exceeding 70 ppb, differed between the Seoul Metropolitan Area (SMA) and the background sites.

In the SMA, the exceedances were more prevalent during summer compared to spring, whereas the background sites experienced higher exceedances in spring than in summer. This indicates the efficient local production of ozone in the SMA during summer and the strong influence of long-range transport during spring. The rest of the sites showed similar exceedance patterns during both spring and summer. The peak exceedances occurred

1 around 4-5 PM in the SMA and most locations, while the background sites primarily  
2 recorded exceedances between 7-8 PM and throughout the night.

3 During the spring of the COVID-19 pandemic (2020-2021), ozone exceedances decreased  
4 at most locations due to significant reductions in NO<sub>x</sub> emissions in South Korea and China  
5 compared to the period of 2010-2019. The largest decreases in exceedances were observed  
6 at the background sites during spring. For instance, in Gosung, Gangwondo (approximately  
7 600 m above sea level), the exceedances dropped from 30% to around 5% during the  
8 COVID-19 pandemic.

9 Regional model simulations confirmed the concept of decreased ozone levels in the  
10 boundary layer in Seoul and Gangwon-do in response to emission reductions. However,  
11 these reductions in ozone exceedances were not observed in major cities and provinces  
12 during the summer of the COVID-19 pandemic, as the decreases in NO<sub>x</sub> emissions in South  
13 Korea and China were much smaller compared to spring. This study highlights the  
14 distinctions between spring and summer in the formation and transport of surface ozone in  
15 South Korea, emphasizing the importance of monitoring and modeling specific processes  
16 for each season or finer time scales.

17

1 **1. Introduction**

2 Ozone, a greenhouse gas and harmful air pollutant, can accumulate in the lower atmosphere  
3 through photochemical reactions involving nitrogen oxides and volatile organic  
4 compounds from both human activities and natural sources (National Research Council,  
5 1991; Monks et al., 2015). The increasing concentrations of ozone near the surface and in  
6 the troposphere are concerning. Gaudel et al. (2018) reported a significant increase in ozone  
7 levels in South Korea from 2000 to 2014, while North America and Europe experienced  
8 decreasing trends, using data from surface monitors, ozonesondes, and aircraft  
9 observations. Other studies have also observed rising ozone trends in South Korea between  
10 2001 and 2018 in their analysis of the long-term variations of multiple pollutants over Seoul  
11 (Kim and Lee, 2018) and South Korea (Kim et al., 2018) or in the reviews of current status  
12 and future directions of tropospheric ozone studies in South Korea (Lee et al., 2020) or in  
13 the trend estimates of the surface ozone observations (Yeo and Kim, 2021). Ozone in South  
14 Korea can be influenced by ozone and its precursor in China (Oh et al., 2010; Lee and Park,  
15 2022; Colombi et al., 2023). However, Gaudel et al. (2018) did not include Chinese data  
16 due to a lack of reported information. Recent studies have highlighted a rapid increase in  
17 ozone levels in China from 2004 to 2020, especially after 2013 (Li et al., 2019; Wang et  
18 al., 2020; Wang et al., 2022). Gaudel et al. (2020) also found that tropospheric ozone in

1 China and South Korea increased between 1996 and 2016. Considering the proximity of  
2 the two countries and their potential for ozone and precursor exchange, it is essential to  
3 study the ozone trends in South Korea in relation to those in China. Additionally, as spring  
4 and summer have distinct transport patterns and source-receptor relationships relevant to  
5 surface and tropospheric ozone (e.g., Cooper et al., 2010), it would be valuable to  
6 investigate ozone trends separately for these seasons.

7 The COVID-19 pandemic brought about significant changes in atmospheric  
8 composition (Bauwens et al., 2020; Koo et al., 2020; Seo et al., 2021). Analyzing deviations  
9 from long-term trends during the pandemic can provide valuable insights for future  
10 environmental policies aimed at mitigating ozone pollution. In this study, we examine  
11 ozone trends and exceedances in South Korea from 2001 to 2021, focusing on the warm  
12 seasons of spring and summer, including the COVID-19 period. In this study, we analyzed  
13 the 4<sup>th</sup> highest daily maximum 8 hours-average ozone concentrations (MDA8 O<sub>3</sub>) at  
14 various locations in South Korea for a global comparison because this is a metric used for  
15 the US Environmental Protection Agency National Ambient Air Quality Standard and the  
16 recent study by Wang et al. (2022) utilized the same metric for their study of Chinese ozone  
17 pollution. We also introduced a new metric of ozone exceedance, defined as the percentage  
18 of data points with hourly ozone concentrations exceeding 70 ppb. Previous published

1 works about surface ozone in South Korea have not focused on the two metrics used in our  
2 study. We analyze diurnal, seasonal, and decadal variations at 7 cities, 9 provinces, and 2  
3 background sites. Furthermore, we discuss the factors contributing to the observed  
4 temporal changes based on regional model results.

5 The manuscript is organized as follows. In section 2, the surface and satellite data,  
6 global and regional modeling, and other methods to utilize the data are explained. In section  
7 3, the results are summarized as long-term trends of ozone and its precursors,  
8 characteristics of diurnal variations, and spatiotemporal variations during the pandemic.  
9 The regional model results based on various emission scenarios are also shown to identify  
10 the source-receptor relationship. Finally, the results are summarized and future research  
11 directions are suggested in the conclusions.

12

## 13 **2. Data and Method**

### 14 **2.1. Long-term surface observational data**

15 The hourly surface air quality monitoring data are obtained from the Airkorea website  
16 (<https://www.airkorea.or.kr>), including ozone (O<sub>3</sub>), NO<sub>2</sub>, SO<sub>2</sub>, CO, PM<sub>10</sub>, and PM<sub>2.5</sub> (PM<sub>2.5</sub>  
17 data are provided since 2015). As of March 2020, there are about 500 monitoring stations  
18 over South Korea. These routine monitor data are available for many decades and can serve

1 as a main data set to examine long-term trends. We utilized hourly and daily maximum 8  
2 hour-average O<sub>3</sub> concentrations. The surface monitoring sites used in this study and the  
3 data availability are summarized in the Supporting Information 1 (SI1, Table S1) and  
4 Supporting Information 2 (SI2). O<sub>3</sub>, NO<sub>2</sub> and CO data are also averaged for spring and  
5 summer months. These surface monitoring data were used to investigate the impact of the  
6 COVID-19 pandemic in the Seoul Metropolitan Area.

7

## 8 **2.2. Highway toll number and mobile phone usage data**

9 To examine changes in mobility pattern during the COVID-19 pandemic, traffic counts  
10 from the Korea Expressway Corporation daily transit data were used  
11 (<http://data.ex.co.kr/portal/>). The expressway transit data covering 3 years (2019-2021) of  
12 traffic passing toll gates were quantified from Hi-Pass (electronic toll collection system)  
13 and cash toll collection. Vehicles passing toll gates were not classified in details.

14 To examine changes in mobility pattern during the COVID-19 pandemic, daily  
15 mobile phone movement provided by Android (Google COVID-19 Community Mobility  
16 Reports, 2020) and Apple (Apple COVID-19 Mobility Trends Report, 2020) are used.  
17 Android mobility data tracked movements of people using cell phones at the same spot,  
18 while Apple's mobility report collects personal vehicle routing requests from Apple Maps.

1 For Google and Apple mobility report, we used the Transit station Mobility metrics and  
2 driving mobility index in Seoul Metropolitan Area, respectively. The reports must be  
3 carefully used as it does not directly quantify on-road traffic.

4

### 5 **2.3. Satellite data: tropospheric NO<sub>2</sub> columns**

6 The TROPOspheric Monitoring Instrument (TROPOMI) on board of a low Earth polar  
7 orbiting satellite, European Space Agency (ESA) Sentinel-5 Precursor (S-5P) satellite with  
8 equator passing time 13:30 local time. The instrument provides measurements at  
9 unprecedently high spatial, temporal, and spectral resolutions (Veeffkind et al., 2012). In  
10 this study we utilized two available tropospheric NO<sub>2</sub> datasets from TROPOMI, NASA's  
11 standard product (SP) version 4.0 (Lamsal et al., 2021) and KNMI's (Royal Netherlands  
12 Meteorological Institute) product obtained from DOMINO v2.0 and QA4ECV v1.1  
13 (Derivation of TROPOMI tropospheric NO<sub>2</sub>) processing systems (Boersma et al., 2018).  
14 The spatial resolution of KNMI's tropospheric NO<sub>2</sub> retrieval product is 3.5 km x 7 km (3.5  
15 km x 5.5 km since 6 August 2019) and that of NASA's product is 3.5 km x 5.5 km. Level  
16 2 data with pixels passing quality assurance  $> 0.75$  and the cloud fraction  $< 0.5$  were  
17 selected for analysis following recommendations provided by Sentinel-5 precursor  
18 TROPOMI Level 2 product User Manual for nitrogen dioxide (Eskes et al., 2019).



1 TROPOMI data are regridded to a standard grid with a horizontal resolution of  $0.1^\circ$  latitude  
2  $\times 0.1^\circ$  longitude ( $11 \times 11$  km) and monthly averaged values were derived. As the random  
3 error in the TROPOMI single-pixel uncertainties influence 40 to 60% of the tropospheric  
4 column abundance, temporal and spatial averaging may remove the random errors  
5 (Bauwens et al., 2020).

6 We conducted the sensitivity test by applying different sampling conditions and  
7 found consistent results irrespective of quality control parameters: larger tropospheric  $\text{NO}_2$   
8 column reduction during spring than during summer between 2019 and 2020-2021  
9 (COVID-19 periods). Differences between KNMI and NASA retrievals are large when the  
10 the filtering condition of quality assurance  $> 0.5$  and cloud radiance fraction  $< 0.4$  is applied.  
11 When the stricter filter is applied, differences between KNMI and NASA retrievals are  
12 small. Therefore, the stricter filter (quality assurance  $> 0.75$  and cloud radiance fraction  $<$   
13  $0.5$ ) is selected. Since the NASA product released in November, 2022 were generated in a  
14 consistent manner for May 2018-December 2021, we mainly present the NASA MINDS  
15 product. We summarized the sensitivity tests in the Supporting Information 3 (SI3). The  
16 distribution of absolute tropospheric  $\text{NO}_2$  columns for different years are also shown in the  
17 SI3.

18

## 1 **2.4. CAM-Chem model simulations**

2 The atmospheric component of Community Earth System model (CESMv2.2), Community  
3 Atmosphere Model with Chemistry version 6 (CAM6-chem) is developed by National  
4 Center for Atmospheric Research (NCAR) (<https://www2.aocom.ucar.edu/gcm/cam-chem>).

5 The CAM-chem adapted MOZART-T1 as the tropospheric chemistry mechanism  
6 (Emmons et al., 2020). The simulation used in this study was configured with 1° horizontal  
7 resolution. The sea surface temperature was prescribed, and meteorological fields were  
8 nudged to Modern-Era Retrospective analysis for Research and Applications version 2  
9 (MERRA-2) instead of using self-produced meteorological field  
10 (<https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/>) (refer to SI1 Figure S1 for  
11 performance of the model wind). The simulation was performed from 2000 to 2020 and  
12 applied CMIP6 emission inventory (2000-2014) and SSP5-8-5 emission inventory (2015-  
13 2020). The first 3 years were regarded as a spin-up. In this study, we utilized the CAM-  
14 Chem results to estimate the impact of stratospheric ozone on the surface in each season.

15 CAM-Chem calculates the contribution of stratospheric ozone to tropospheric ozone,  $O_{3S}$   
16 as a three-dimensional variable in space. Originally,  $O_{3S}$  is ozone value above tropopause.  
17 Then  $O_{3S}$  is transported below tropopause and undergoes chemical losses in the model.

18 Evaluations of the CAM-Chem results against the data from the ozonesondes that were

1 launched in Pohang, South Korea are shown in the Supporting Information (SI1, Figure S2;  
2 Jeong et al., 2023). The model results and observations reasonably agree in terms of  
3 seasonal variability and absolute values. Especially, the CAM-Chem results agree with the  
4 observations at the 200 hPa level, close to tropopause.

5

## 6 **2.5. WRF-Chem model simulations**

7 The Weather Research and Forecasting (WRF) model coupled with Chemistry (WRF-  
8 Chem) is developed by National Oceanic and Atmospheric Administration (NOAA) and  
9 National Center for Atmospheric Research (NCAR) and collaborating institutes (Grell et  
10 al., 2005). We utilized WRF-Chem v4.4 to simulate regional meteorological fields and  
11 chemical compositions.

12 Our WRF-Chem set up utilizes the horizontal resolution of 28 x 28 km<sup>2</sup> and 60  
13 vertical levels. The simulation period is from 24th April 12 UTC to 11th June 12 UTC in  
14 2016. We restart the simulation at 12 UTC every day to reduce computing errors. The first  
15 7 days of model simulation is regarded as spin-up period. The analysis period is selected  
16 as 1st May to 10th June based on local time. The Global Forecast System (GFS) Final (FNL)  
17 analysis data are used for meteorological input and boundary conditions  
18 (<https://rda.ucar.edu/datasets/ds083.2/>). We used the Community Atmosphere Model with

1 Chemistry (CAM-Chem) output to the chemical boundary and first initial conditions  
2 (<https://www.acom.ucar.edu/cam-chem/cam-chem.shtml>) (Buchholz et al., 2019). The  
3 Model of Emissions of Gases and Aerosols from Nature (MEGAN) is used for biogenic  
4 emissions (Guenther et al., 2006).

5       There are 7 model sensitivity runs that adopt different emission scenarios. The  
6 control run is based on the standard EDGAR-HTAPv3 emission inventory representing  
7 2016 (Crippa et al., 2023). Park et al (2021) informed that biomass burning was not an  
8 important factor affecting air quality in South Korea during KORUS-AQ. Therefore,  
9 biomass burning emissions are omitted in this study. “No China” case removes all  
10 anthropogenic emissions in China. “No Seoul” case eliminates all anthropogenic emissions  
11 in Seoul. There is one case that decreased Chinese VOC emissions by 50%. There are two  
12 cases that reduced Chinese NO<sub>x</sub> emissions by 50%: the one case has the same VOC  
13 emissions as in the control case while the other case has the 50% reductions of VOC  
14 emissions as well. Lastly, there is one case that reduced Chinese NO<sub>x</sub> emissions by 75%.  
15 The WRF-Chem sensitivity runs are summarized and are discussed in Section 3. The  
16 extensive evaluations of the model results against the surface and airborne data from the  
17 KORUS-AQ field campaign in 2016 are shown in the Supporting Information (SI1 Table  
18 S2-S4 and Figure S3-S8) and Kim K.-M. et al (2023). More discussions about all bottom-

1 up emission inventories utilized for the WRF-Chem simulations and comparison of those  
2 with the emissions used for the CAM-Chem simulations are included in SI1 Table S5.

3

### 4 **3. Results**

#### 5 **3.1. Surface ozone trends**

6 In this study, ozone and its precursor concentrations in 7 cities, 9 provinces, and 2  
7 background sites in South Korea (Figure 1) are analyzed at diurnal, seasonal, and decadal  
8 time scales. Figure 2 and 3 shows the 4<sup>th</sup> highest daily maximum 8 hours-average ozone  
9 concentrations (MDA8 O<sub>3</sub>) for the cities and provinces for ozone season (May-September)  
10 from 2001 to 2021. The results from statistical analysis (slope, standard-deviation, p-value,  
11 signal-to-noise ratio) are summarized in Table 1. P-values were presented as suggested by  
12 Chang et al. (2021) and Wasserstein et al. (2019) for the purpose of estimating uncertainties  
13 in trends. The 4<sup>th</sup> highest MDA8 O<sub>3</sub> increases by 1.0-1.5 ppb yr<sup>-1</sup> with very high certainty  
14 for most of cities and provinces across South Korea in this period. In nearly all cities and  
15 provinces, the 4<sup>th</sup> highest MDA8 O<sub>3</sub> has been higher than 70 ppb since 2010 or earlier (see  
16 gray dashed line in Figures 2 and 3). The trend in Jeollanam-do (JLN) is small with very  
17 low certainty (p=0.67) partly because the MDA8 O<sub>3</sub> was high before 2010. The monitoring  
18 sites in Jeollanam-do include the Yeosu-Kwangyang region in which many petrochemical  
19 industries and iron steel complexes are located. This region experienced severe ozone  
20 problems in the 1990's to early 2000's (Ghim, Y. S. 2000). Because of discontinuity of data

1 records, the background sites are omitted in the trend analysis in Figure 2 and 3. Although  
2 there are missing data during the ozone season for the background sites, we estimated the  
3 trends of the 4<sup>th</sup> highest MDA8 O<sub>3</sub> for a reference (Table 1). The estimated trend for  
4 Ulleung Island is 0.42 ppb yr<sup>-1</sup> from 2001 to 2021 with low or very low certainty. The trend  
5 for Ulleung Island increases to 0.92 ppb yr<sup>-1</sup> for 2001-2019 with medium certainty. The  
6 estimated trend for Gosung is 0.73 ppb yr<sup>-1</sup> from 2001 to 2021 with medium to high  
7 certainty. The trend for Gosung increased to 1.45 ppb yr<sup>-1</sup> for 2001-2019 with very high  
8 certainty. Widely increasing long-term ozone trends in South Korea indicate a regional  
9 nature of this pollutant, potentially influenced by East Asian emissions, chemical  
10 transformations, and long-range transport (Colombi et al., 2023; Lee and Park, 2022).  
11 Therefore, it is imperative to understand the local and regional processes that enhance  
12 surface ozone. Ozone originated from Asia is known to be efficiently transported to North  
13 America during springtime (Jacob et al., 1999; Jaffe et al., 1999; Jaffe et al., 2003; Cooper  
14 et al., 2010; Lin et al., 2012; Langford et al., 2017; Jaffe et al., 2018) and summertime  
15 (Fiore et al., 2002; Liang et al., 2007) as well. Investigating seasonal differences in ozone  
16 in South Korea may provide insights on the relative importance of local and regional  
17 processes.

18

19 **3.2. Difference between spring and summer: background value, exceedance,**  
20 **stratospheric influence, and precursor concentrations**

1 3.2.1. Background values at the daytime and nighttime periods

2 Table 2 summarizes the abundances and differences between spring and summer ozone  
3 concentrations averaged for the daytime (10-20 Local Time (LT)) and the nighttime (01-06  
4 LT) periods. Standard deviations denoting year-to-year variability are also shown in the  
5 table. For the nighttime period, the averaged ozone concentration in spring is always higher  
6 than that in summer: differences between the two seasons range from 3.3 to 15.4 ppb and  
7 are generally larger than standard deviations. The differences for the background sites are  
8 approximately 13 ppb at this time. This clearly indicates the importance of large-scale  
9 influences in spring. The results are the same for the daytime period except for Seoul and  
10 Gyeonggi-do: the mean ozone concentrations in Seoul and Gyeonggi-do in summer are  
11 slightly higher than those in spring. The differences in the daytime period are small for  
12 Incheon, Daegu, and Chungcheongbuk-do, suggesting the importance of local chemistry  
13 in the areas during summer. Overall, standard deviations are larger than the differences in  
14 the daytime period except Busan, Jeollanam-do, Jeju Island, Gyeongsangbuk-do,  
15 Gangwon-do, and the background sites. The differences for the background sites in the  
16 daytime period are approximately 13-15 ppb, which are similar to the values in the  
17 nighttime period.

18 The surface ozone data for 01-06 LT over polluted regions are often omitted in the  
19 analysis because ozone loss reacting with NO is an important process to control ozone  
20 levels at nighttime. In this study, we utilized the ozone data at this time to find information

1 about background ozone because ozone is transported throughout a day and this process is  
2 essential in the studied region. WRF-Chem sensitivity runs demonstrated increase of ozone  
3 from upwind sources at this time (refer to SII Figure S9).

4 Additionally, we also calculated  $O_x (=O_3 + NO_2)$  that are not affected by the reaction of  
5 ozone with fresh local  $NO_x$  emissions during nighttime and compared this value at daytime  
6 and nighttime for each season (SII Table S6 and S7 for  $NO_2$  and  $O_x$ , respectively). Overall,  
7  $O_x$  during spring is higher than that during summer and nighttime differences are  
8 prominent, which is similar to the conclusions from the analysis of  $O_3$ .

9

### 10 3.2.2. Ozone exceedances

11 Figure 4 illustrates the ratio of summer ozone exceedances to spring ozone exceedances  
12 for the cities, provinces, and background sites. In Seoul, Incheon, and Gyeonggi-do, there  
13 are more exceedances in summer than in spring, indicating the significance of local ozone  
14 production during the summer season in these areas. Conversely, at the background sites  
15 such as Gosung and Ulleung Island, springtime exceedances dominate, highlighting the  
16 importance of high springtime ozone levels and their transport within and beyond Asia. For  
17 the remaining regions, springtime and summertime exceedances are comparable, or  
18 springtime exceedances are slightly higher than those in summer. Note that meteorological  
19 conditions in Seoul and Gyeonggi-do (differences between the two seasons) are similar to  
20 other cities and provinces (see SII Table S8). Therefore, the meteorological factors are not



1 main drivers of high summertime exceedances in Seoul and Gyeonggi-do region.

2           The diurnal variations of exceedances, as shown in Figure 5, confirm these  
3 findings. The summertime ozone exceedances are notably enhanced during the daytime,  
4 from 13 to 20 local time (LT), suggesting efficient photochemical ozone production during  
5 this season. The peak exceedances occur at 17 LT in Seoul and Gyeonggi-do, and one hour  
6 earlier at 16 LT in Incheon. Incheon, being situated adjacent to the West Sea (as depicted  
7 in Figure 1), experiences airflow from Incheon to Seoul under typical westerly or seabreeze  
8 conditions. The late-afternoon peaks (4-5 PM) in the region and the one-hour delay in peak  
9 exceedances in Seoul compared to the time of exceedances in Incheon imply that local  
10 circulation plays a significant role in the buildup and distribution of ozone within the  
11 Incheon, Seoul, and Gyeonggi-do region.

12           Springtime and summertime ozone exceedances predominantly occur during the  
13 daytime, with some extent of exceedances at night, in Daejeon, Busan, and Daegu (Figure  
14 5). Notably, the peaks in spring occur approximately two hours later than those in summer  
15 for the three cities, indicating a potential influence of transport during the spring season.  
16 Negligible exceedances are observed from midnight to 10 LT in the three cities due to high  
17 NO<sub>x</sub> pollution and the depletion of ozone associated with NO<sub>x</sub> during this time period.

18           At the background sites, springtime exceedances are much higher compared to  
19 summer, and nighttime exceedances are as frequent as daytime exceedances. In Gosung,  
20 springtime exceedances account for approximately 20% of the observations throughout the

1 day, whereas summertime exceedances are less than or equal to 10% (Figure 5). The  
2 observation site in Gosung is located at an altitude of approximately 600 meters above sea  
3 level, providing a unique opportunity to examine long-range transported plumes and  
4 background information at higher altitudes (refer to SI1 Table S9 and Figure S10). Diurnal  
5 variations of exceedances during spring and summer for all individual sites are illustrated  
6 in SI1 (Figure S11-S13).

7

### 8 3.2.3. Influence of stratospheric ozone

9 Stratospheric ozone can deeply intrude into the lower troposphere, leading to elevated  
10 surface ozone levels, particularly during the spring season (Lin et al., 2012; Lin et al., 2015).  
11 It is important to assess the contribution of stratospheric ozone to surface ozone in South  
12 Korea and understand its potential impact on surface ozone trends in the region using  
13 results from the CAM-Chem model. The derivation of the contribution of stratospheric  
14 ozone in the CAM-Chem is explained in the Supporting Information. Figure 6 presents the  
15 contribution of stratospheric ozone to surface ozone in South Korea for each season.  
16 According to our global chemistry-climate model simulations, stratospheric ozone has the  
17 greatest influence on surface ozone during winter and spring, increasing levels by 17-23  
18 ppb. The model suggests that approximately 37% and 76% of surface ozone in spring and  
19 winter, respectively, can be attributed to stratospheric ozone (refer to SI1 Table S10 for  
20 summary of the CAM-Chem results for all seasons at surface and 1 km above ground level).

1 However, during the summer season, the impact of stratospheric ozone on surface ozone is  
2 minimal, accounting for only around 4% of the surface ozone concentration. Therefore, it  
3 would be valuable to analyze ozone trends and exceedances separately for spring and  
4 summer. It is worth noting that the contribution of stratospheric ozone to surface ozone  
5 does not exhibit clear trends during the period from 2001 to 2021 (not shown). Note that  
6 the contribution of stratospheric ozone to tropospheric ozone at each altitude and time  
7 shown in this study should be a qualitative measure since the representation of this process  
8 has uncertainties and needs further assessment.

9

#### 10 3.2.4. Long-term trends of surface NO<sub>2</sub> and CO concentrations

11 In contrast to the trends of ozone, NO<sub>2</sub> and CO that are ozone precursors decreased both in  
12 spring and summer from 2001 to 2021 (Table 3 and 4). There are no systematic differences  
13 in the trends of NO<sub>2</sub> and CO between the two seasons. NO<sub>2</sub> has declined in Seoul, Busan,  
14 Daegu, Gwangju, Incheon, Gyeongsangbuk-do, and Gyeonggi-do with very high certainty.  
15 For the rest of sites, the declining NO<sub>2</sub> trends were found with medium-to-high certainty  
16 (refer to Chang et al., 2021 for assessment of uncertainty in the trend analysis). Seo et al.  
17 (2021) investigated the trend of NO<sub>2</sub> in the Seoul area utilizing satellite tropospheric NO<sub>2</sub>  
18 columns and surface NO<sub>2</sub> observations from 2005 to 2019 and found decrease of NO<sub>2</sub> only  
19 between 2015 and 2019. They did not find significant trends between 2005 and 2015.  
20 Therefore, the trends in our study are strongly influenced by recent NO<sub>2</sub> reductions prior  
21 to and during the COVID-19 pandemic. CO reductions are evident for a wider region with  
22 very high certainty. Only the CO trend in Jeollanam-do was estimated with high certainty

1 (instead of very high certainty). The decreasing trends of  $\text{NO}_2$  and CO were estimated with  
2 very low certainty in Ulsan throughout this period. Overall, signs of slopes agree between  
3 emission inventory and ambient concentrations at least for the cities, but site-to-site  
4 variations do not agree even for the cities. There are disagreements of signs of slopes  
5 between emission inventory and ambient concentrations for the provinces (refer to SI1  
6 Table S11 and S12). This can be attributed to the uncertainties in the bottom-up emission  
7 inventories of  $\text{NO}_x$  and CO in South Korea.

8 Ozone increases in South Korea despite reduction of main precursors at local scale  
9 can be attributed to the increase of long-range transport of ozone or potentially “VOC-  
10 limited” (or “ $\text{NO}_x$ -saturated”) local photochemical regime of South Korea. “VOC-limited”  
11 regime is the condition in which  $\text{NO}_x$  (sum of NO and  $\text{NO}_2$ ) concentration is high and VOC  
12 is a limiting factor to form ozone. In this case, VOC reduction would decrease ozone, while  
13  $\text{NO}_x$  reduction would nonlinearly increase ozone. Since long-range transport from China  
14 is frequent during spring, it is useful to identify characteristics of ozone exceedance in  
15 spring separate from summer.

16

### 17 **3.3. Changes detected during the COVID-19 pandemic (2020-2021) compared to** 18 **2002-2019**

19 Nationwide social distancing protocol enforced by Korean government started February 25  
20 of 2020 and lasted until April 18 of 2022, although levels of protocol differ. During spring  
21 in 2020 (until May 6, 2020), facilities for public use (libraries, swimming pools, museums,

1 and national parks) and religious, indoor sports, entertainment facilities were forced to  
2 close, and people were refrain from going out except for buying necessities, visiting a  
3 doctor, and commuting to/from work. Since May 6 of 2020, as number of new confirmed  
4 COVID-19 cases remain relatively steady, the guidelines have shifted from social  
5 distancing to distancing in daily life, no restrictions on people going out. Because a cluster  
6 of new COVID-19 cases emerged in mid-August, social distancing protocol (since August  
7 16 until early October) was again forced by the government, people were strongly  
8 recommended to stay indoors. After August 16 of 2020, there were well-defined  
9 government protocols as Level 1, 2, and 3: Level 1 is no restricted personal gathering and  
10 daily life, Level 2 allows personal gathering up to 8 people and discourage unnecessary  
11 and unurgent travel, and Level 3 allows personal gathering up to 3 people, requires remote  
12 work and online classes, and discourage travels. Most days in spring and summer in 2021  
13 were the period under the Level 2 protocol. In summary, most distinct changes in social-  
14 distancing protocols and traffic/mobile activities occurred between spring and summer in  
15 2020 in South Korea (refer to SI1 Figure S14-15).

16

### 17 3.3.1. Changes in ozone exceedances and local precursors during springtime

18 The frequency of springtime ozone exceedances increases from period P1 (2002-2010) to  
19 period P2 (2011-2019) across all observation sites in South Korea (Figure 7). However,  
20 during the COVID-19 period (P3: 2020-2021), the frequency of exceedances significantly

1 decreases at most sites. Notable reductions are observed in Daejeon, Daegu,  
2 Chungcheongbuk-do, Gyeongsangnam-do, Gyeongsangbuk-do, Gangwon-do, as well as  
3 the background sites Gosung (Gangwon-do) and Ulleung Island. In Gosung, the percentage  
4 of ozone exceedances drops from 30% during P2 to 5% during P3 in spring. Although  
5 Gosung is located close to the East Sea and is the region farthest from China within a  
6 similar latitude range, it is still susceptible to long-range transported ozone due to its high  
7 elevation (see SI1 Figure 10 for the elevation map and diagram of a possible ozone  
8 transport path).

9         Across all sites, the concentration of NO<sub>2</sub> shows little change from P1 to P2, with  
10 an average decrease of 5%. However, during the COVID-19 period (P2 to P3), there was  
11 an average reduction of 25% in NO<sub>2</sub> concentrations. CO concentrations also experienced a  
12 decrease of 22% from P1 to P2 and a further decrease of 14% from P2 to P3. However, the  
13 reductions in CO are relatively minor compared to the changes in NO<sub>2</sub> observed during the  
14 COVID-19 period. The decrease in ozone exceedances during COVID-19 may be  
15 associated with the reductions in NO<sub>2</sub> concentrations during this time.

16         A notable finding is the significant reduction in ozone levels at the background  
17 sites, such as Gosung and Ulleung Island, between P2 and P3. This suggests a cleaner  
18 background influenced by changes in emissions from sources in Asia and long-range  
19 transport. It is important to note that there were no significant changes in NO<sub>2</sub> and CO  
20 concentrations observed at the background sites from P2 to P3. There are several studies

1 reporting the increase of near-surface ozone after COVID lockdowns in the urban areas  
2 (e.g., Shi & Brasseur, 2020) because of expected non-linear relationship between ozone  
3 and NO<sub>x</sub> in the highly polluted regions. However, there are also studies reporting reductions  
4 of ozone concentrations from 1 to 8 km altitude in the northern extra-tropics during COVID  
5 (Steinbrecht et al., 2021). Parrish et al. (2020) reported zonal similarity of tropospheric  
6 ozone changes at northern mid-latitudes. Therefore, ozone reductions from P2 to P3 across  
7 the sites in South Korea may be associated with decreased background ozone at northern  
8 mid-latitudes to some extents. On top of this, local and regional emission changes during  
9 COVID may also play a role in reducing ozone exceedances in South Korea in this season.

10

### 11 3.3.2. Changes in ozone exceedances and local precursors during summertime

12 During summer, ozone exceedance frequencies also increase from P1 to P2 for all sites:  
13 Chungcheongnam-do has the largest increase from 3.2% to 11.3% and Gyeonggi-do,  
14 Daejeon, Jeollabuk-do, Gyeongsangnam-do and Gyeongsangbuk-do have similar increases  
15 (Figure 8). The ozone exceedances in the background sites Gosung, and Ulleung Island  
16 also increase in this period. NO<sub>2</sub> and CO concentrations decreased marginally from P1 to  
17 P2. During COVID-19, the ozone exceedance frequencies in summer increase in Seoul,  
18 Incheon, Gyeonggi-do, and Chungcheongnam-do, substantially decrease in Gangwon-do  
19 and the background sites, and does not show changes from P2 for the rest of sites. Because  
20 NO<sub>2</sub> concentrations decrease from P2 to P3 for Seoul, Incheon, Gyeonggi-do, and

1 Chungcheongnam-do contrasting with increases of ozone exceedance, chemical regime for  
2 these regions during summer is likely to be VOC-limited ( $\text{NO}_x$ -saturated) as mentioned  
3 above and as in previous studies (e.g., Kim et al., 2020). Ozone exceedance substantially  
4 decreases in the background sites from P2 to P3 during summer, indicating cleaner air at  
5 large-scale as shown in Steinbrecht et al. (2021).

6

7 3.3.3. Changes in precursor concentrations at a regional scale during spring and summer:

8 TROPOMI tropospheric  $\text{NO}_2$  columns

9 Figure 9 presents the spatial distributions of NASA TROPOMI tropospheric  $\text{NO}_2$  columns  
10 (Lamsal et al., 2022) in spring (MAM) and summer (JJA) across East Asia, along with their  
11 changes from 2019 to 2020 and from 2019 to 2021. The plot illustrates significant  
12 reductions in  $\text{NO}_2$  columns during the spring of COVID-19 in most areas of China, South  
13 Korea, and the surrounding seas. Changes in traffic activities in the Seoul Metropolitan  
14 Area were also detected between 2019 and 2020 (refer to SI1 Figure S14 and 15). The  
15 number of cars counted at the highway tolls in this region decreased by 6% in March, April,  
16 and May in 2020 compared to 2019, but this trend was reversed in June (SI1 Figure S14).  
17 Furthermore, observed concentrations of  $\text{NO}_2$ ,  $\text{SO}_2$ , CO,  $\text{PM}_{10}$ , and  $\text{PM}_{2.5}$  during the spring  
18 of 2020 showed reduction of 15-30% (SI1 Figure S15). Changes in traffic counts in the  
19 Seoul Metropolitan Area between 2019 and 2021 were small (SI1 Figure S14 and S15).  
20 But observed concentrations of  $\text{NO}_2$ ,  $\text{SO}_2$ , CO,  $\text{PM}_{10}$ , and  $\text{PM}_{2.5}$  were also reduced during



1 spring in 2021 compared to 2019 by 10-30% except for PM<sub>10</sub> that was enhanced due to  
2 Asian dust events in spring 2021 (SI1 Figure S15).

3 As depicted in Figure 9, TROPOMI tropospheric NO<sub>2</sub> columns also decreased  
4 during the summer in the same region, although in fewer locations and to a lesser extent  
5 compared to the spring, during the COVID-19 period. The observed NO<sub>2</sub>, SO<sub>2</sub>, CO, PM<sub>10</sub>,  
6 and PM<sub>2.5</sub> concentrations in the Seoul Metropolitan Area were also reduced during summer  
7 in 2020 or 2021 compared to 2019 by 2-20%. Surface NO<sub>2</sub> concentrations reduced by ~10%  
8 during summer, which is smaller than the reductions during spring (~20%; see SI1 Figure  
9 S14 and S15). Overall, the reductions in 2020/2021 from 2019 during summer are smaller  
10 than those during spring at the surface in the Seoul Metropolitan Area, which is similar to  
11 the seasonal changes detected from space.

12 The substantial decrease in NO<sub>2</sub> in China during spring, observed by satellite, is  
13 likely to contribute to significant reductions in ozone levels in South Korea due to long-  
14 range transport. Additionally, local reductions in NO<sub>x</sub> emissions in South Korea can lead  
15 to ozone decreases if the reductions are significant enough, especially in the "VOC-limited"  
16 chemical regime prevalent in this area. However, further investigation is required to  
17 understand the detailed source-receptor mechanism of ozone and its precursors in each  
18 season, which warrants long-term air quality model simulations in future studies. The next  
19 section of this study discusses the sensitivity of ozone concentrations in Seoul and  
20 Gangwon-do to various emission scenarios in China and South Korea, albeit within a

1 limited time period.

2

3 **3.4. Impacts of changes in East Asian emissions on surface/boundary layer ozone in**  
4 **South Korea: a modeling analysis**

5 3.4.1. Changes in surface/boundary layer ozone due to emission reductions: East Asian  
6 region

7 In this section, we will discuss WRF-Chem model simulations conducted during the  
8 KORUS-AQ 2016 field campaign (primarily in May; refer to Crawford et al., 2021 for  
9 detailed information) to gain insights into the impacts of emission changes on ozone  
10 concentrations in East Asia, including South Korea. We have extensively evaluated our  
11 model results with the airborne and surface observations acquired during the KORUS-AQ  
12 campaign and the routine surface monitors in China and South Korea. The model decently  
13 simulated boundary-layer ozone over South Korea (3% difference) for the cases that were  
14 strongly influenced by long-range transport. For local emission dominating cases, the  
15 model underestimated boundary-layer ozone over South Korea by 20%. The results are  
16 summarized in SI1 (Table S3 and Figure S8) and Kim K.-M. et al. (2023). This study  
17 considers two extreme cases: the "No China" case, where all anthropogenic emissions in  
18 China are removed, and the "No Seoul" case, where all anthropogenic emissions in Seoul  
19 are removed. Additionally, several other scenarios are examined, including a 50% reduction  
20 in Chinese NO<sub>x</sub> emissions only, a 50% reduction in Chinese VOC emissions only, a 50%

1 reduction in both Chinese NO<sub>x</sub> and VOC emissions, and a 75% reduction in Chinese NO<sub>x</sub>  
2 emissions only.

3 Our study reveals both increases and decreases in ozone concentrations due to  
4 emission changes resembling those during the COVID-19 period. Specifically, near-  
5 surface ozone concentrations in polluted regions increase, while ozone concentrations in  
6 the elevated layer show reductions (refer to Figures 10 and 11). A novel finding is the  
7 decrease in downwind ozone, from the near surface to the upper layer, resulting from  
8 reductions in NO<sub>x</sub>/VOC emissions in upwind pollution hotspots (refer to Figures 10 and  
9 11 for several sensitivity runs). For instance, a 50%-75% reduction in Chinese NO<sub>x</sub>  
10 emissions leads to decreased ozone concentrations in Korea, surrounding seas, and the  
11 Pacific Ocean, from the surface to the upper layers. However, near-surface ozone in  
12 Northeast China increases due to these emission changes.

13 Reductions in Chinese VOC emissions result in decreased ozone concentrations  
14 from the surface to the upper layer and from hotspots to downwind areas. Our study  
15 suggests potential changes in photochemical regimes with altitude over pollution hotspots,  
16 indicating NO<sub>x</sub>-saturated conditions near the surface and NO<sub>x</sub>-limited conditions in the  
17 elevated layer. Thus, the combined effects of vertical and horizontal ozone transport, as  
18 well as local production dependent on altitude, would determine the ultimate changes in  
19 ozone concentrations at specific locations and altitudes. It is important to note that the

1 accuracy of VOC emission estimates also influences the assessment, but this aspect is  
2 highly uncertain and requires further study.

3

#### 4 3.4.2. Vertical sensitivity of ozone changes in South Korea to East Asian emission 5 reductions

6 Figure 11 presents the vertical profiles of simulated ozone concentrations for different  
7 emission scenarios. In Seoul, the 50% reduction in Chinese NO<sub>x</sub> emissions only slightly  
8 decreases ozone concentrations near the surface but decreases them above 500 m AGL  
9 (above ground level) to a larger extent. The 50% reduction in Chinese VOC emissions  
10 causes a decrease in ozone concentrations from the surface to 2000 m AGL. In the elevated  
11 layer (> 1500 m AGL) in Seoul, the reduction in Chinese NO<sub>x</sub> emissions leads to a greater  
12 decrease in ozone concentrations compared to the reduction in Chinese VOC emissions.  
13 The scenario with a 50% reduction in both Chinese NO<sub>x</sub> and VOC emissions efficiently  
14 decreases ozone concentrations from the surface to 2000 m AGL, particularly above 1000  
15 m AGL. The scenario with a 75% reduction in NO<sub>x</sub> emissions decreases ozone  
16 concentrations near the surface similarly to the scenarios with a 50% reduction in NO<sub>x</sub> and  
17 VOC emissions, but it causes the largest ozone reductions above 1000 m AGL, except for  
18 the "No China" emission scenario. The "No China" emission scenario results in ozone  
19 concentrations 10-15 ppb lower than the control case at all altitudes. On the other hand, the  
20 "No Seoul" emission scenario leads to ozone concentrations about 20 ppb higher than the

1 control case near the surface, partly due to significantly reduced ozone depletion reactions  
2 with NO. The sensitivity test results for Seoul and Gosung, Gangwon-do are similar, except  
3 that all emission scenarios (including "No Seoul" and 50% reduction in Chinese NO<sub>X</sub>  
4 scenarios) cause a decrease in ozone concentrations in Gangwon-do. Both NO<sub>X</sub> and VOC  
5 emission reductions in China contribute to cleaner air in Gangwon-do, with the largest  
6 cleaning effect observed above 500 m AGL. This may explain the sharp decline in ozone  
7 exceedances observed in Gosung, located at an elevation of approximately 600 m AGL,  
8 during the COVID-19 pandemic (Figure 7). Refer to SI1 (Table S6 and Figure S10) about  
9 altitudes of monitoring sites in Gangwon-do including Gosung. The sensitivity runs clearly  
10 demonstrate the long-range transport of Chinese ozone or the influence of Chinese  
11 emissions on the eastern part of the Korean Peninsula, such as Gangwon-do, from May to  
12 the beginning of June 2016. Both reductions in Chinese VOC emissions and NO<sub>X</sub>  
13 emissions contribute to improving ozone pollution in the boundary layer (1-3 km) in South  
14 Korea.

15

#### 16 3.4.3. Comparisons with recent modeling research

17 Lee and Park (2022) investigated seasonal differences in ozone utilizing a chemical  
18 transport model. They reported the April mean ozone concentration of 39.3 ppb, which is  
19 slightly higher than the July counterpart (38.3 ppb) from their model simulations for the  
20 year 2016 and the selected surface monitor sites for 4 main regions (Seoul,

1 Chungcheongbuk-do, Gwangju, and Busan). Our study summarizes the differences  
2 between spring (March, April, May) and summer (June, July, August) for 21 years  
3 including 192 monitoring sites covering the whole of South Korea focusing on the analysis  
4 of long-term surface ozone observations. On average, the observed spring mean ozone is  
5 34.3 ppb and the summer mean ozone is 29.0 ppb over South Korea in our study. Lee and  
6 Park (2022) indicated that ozone air quality in South Korea is determined mainly by year-  
7 round regional background contributions (peak in spring). With some differences in details,  
8 the results from the two studies are qualitatively similar arguing high springtime  
9 background ozone value. One unique aspect of our modeling study is demonstrations of  
10 the impact of emissions in Seoul on Gangwon-do, causing slight ozone decrease in  
11 Gangwon-do with zero-Seoul emissions from surface to 2 km in May 2016. Our study  
12 highlights the diverse impacts of surface emission changes (over China or Seoul) on  
13 downwind ozone at different altitudes (Figure 11).

14 Colombi et al. (2023) performed an analysis on the effect of precursor changes on  
15 observed surface ozone increases in South Korea. A main difference between Colombi et  
16 al. (2023) and our study is the period of the study and whether it focuses on the surface  
17 ozone or vertical sensitivity explaining ozone variability at different locations in South  
18 Korea. Our study investigated surface ozone and ozone at various altitudes to consider the  
19 transport within and above the boundary layer between China and South Korea. Colombi  
20 et al. (2023) analyzed the surface ozone and NO<sub>2</sub> concentrations mainly over the Seoul

1 Metropolitan Area from 2015 to 2019. The increase of ozone was mostly attributed to  
2 decrease in NO<sub>2</sub> for the studied period.

3 Both Lee and Park (2022) and Colombi et al. (2023) indicated high background  
4 ozone concentration external to East Asia (or South Korea), suggesting difficulty of  
5 achieving ozone standards. Our study agrees to this point. Probably one different message  
6 is that reducing emissions of NO<sub>x</sub> and VOC here and there all together have positive  
7 impacts on reducing ozone downwind. For example, emission reductions associated with  
8 the COVID-19 would lead to decrease of ozone at most sites over South Korea in spring.  
9 However, our study also indicates that the ozone exceedances notably increased in SMA  
10 and Chungcheongnam-do (where large mobile, industrial, and power plant emission  
11 sources are located) during the summer of the COVID-19 pandemic.

12

#### 13 **4. Conclusions**

14 We conducted a study on the spatiotemporal variability of surface ozone in 7 cities, 9  
15 provinces, and 2 background sites in South Korea from 2001 to 2021. The 4<sup>th</sup> highest  
16 maximum daily 8-hour average (MDA8) ozone concentrations showed an increasing trend  
17 in all cities, most provinces, and background sites during this period (or 2001 to 2019),  
18 with a yearly increase of 1-2 ppb. After 2010, these concentrations reached approximately  
19 70 ppb or higher. If the US EPA National Ambient Air Quality Standards were applied,  
20 most of the monitoring sites in South Korea would have been considered nonattainment

1 areas for the past decade.

2 Ozone exceedances in this study were defined as the ratio of data with  
3 concentrations exceeding 70 ppb to the total data, which aligns with the US EPA standard.  
4 In Seoul, Incheon, and Gyeonggi-do, ozone exceedances were more frequent in summer  
5 than in spring. However, the opposite trend was observed in Daejeon, Gwangju, Jeollanam-  
6 do, Gyeongsangbuk-do, Gangwon-do, Jeju Island and the background site Gosung and  
7 Ulleung Island. In other areas, the frequencies of exceedances were similar between spring  
8 and summer. The majority of ozone exceedances occurred between 16-19 LT (4-7 PM).  
9 Interestingly, exceedances also occurred frequently at night in background sites such as  
10 Gosung and Ulleung Island, indicating a strong influence of long-range transport on surface  
11 ozone levels in these locations.

12 Ozone exceedances increased from period P1 (2002-2010) to period P2 (2011-2019)  
13 across all observation sites in South Korea during spring and summer. Overall, NO<sub>2</sub>  
14 concentrations showed declining trends from 2001 to 2021, but significant and relatively  
15 large decreases were only evident after the mid 2010s. NO<sub>2</sub> concentrations for P1 and P2  
16 were similar and increase of CO/VOC concentrations between the two periods were not  
17 detected or reported. Therefore, it is not clear what drove increase of ozone exceedances  
18 over South Korea from P1 to P2. The observed increase in ozone exceedances from P1 to  
19 P2 in South Korea may be partially attributed to the rise in anthropogenic emissions  
20 originating from China. More modeling experiments covering the P1 to P2 period would



1 help identify the main factors behind the ozone increases. It is important to investigate not  
2 only changes in anthropogenic emissions but also the impact of climate change on ozone  
3 variations during this period. We observed significant reductions in ozone exceedances  
4 across almost all monitoring sites in South Korea during the spring of the COVID-19  
5 pandemic (period P3, 2020-2021), which was attributed to decreased anthropogenic  
6 activities and subsequent lower emissions in both China and South Korea. It should be  
7 noted that ozone exceedances substantially increased in SMA and Chungcheongnam-do  
8 during the summer of the COVID-19 pandemic. We conducted sensitivity tests using a  
9 regional chemical model to investigate the impact of emission changes on ozone pollution  
10 in South Korea for a limited period in spring. The results suggest that reductions in Chinese  
11 NO<sub>x</sub> emissions as well as VOC emissions can contribute to the improvement of ozone  
12 pollution in South Korea. These findings provide valuable insights for future efforts to  
13 address ozone pollution in South Korea and emphasize the need for further research to  
14 project air quality and prioritize actions for the next decade or so.

15 In the future, employing multidecadal mathematical modeling on a local to global  
16 scale in both hindcast and forecast modes would be beneficial for better understanding  
17 ozone trends in South Korea. Additionally, reliable VOC observations and conducting  
18 intensive field campaigns, similar to the KORUS-AQ 2016, would provide crucial  
19 information to unravel the complexities of ozone chemistry in this region and facilitate the  
20 careful monitoring of changes in atmospheric composition relevant to ozone. Monitoring

1 ozone within the boundary layer and at higher altitudes is crucial for enhancing our  
2 understanding of ozone trends in South Korea. A network of ozonesondes at multiple sites  
3 with the capability of weekly launches would be a valuable complement to a large field  
4 campaign.

5

#### 6 **Code/Data availability**

7 • The surface monitor data for South Korea can be downloaded from

8 <https://www.airkorea.or.kr/web/>.

9 • Korea Expressway Corporation transit data: Daily traffic counts using highway,  
10 available at: <http://data.ex.co.kr/portal/>, last access: 31 December 2022.

11 • KORUS-AQ data: NASA/LARC/SD/ASDC. (2022). KORUS-AQ Aircraft Merge  
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14 [https://doi.org/10.5067/ASDC/SUBORBITAL/KORUSAQ\\_Merge\\_Data\\_1](https://doi.org/10.5067/ASDC/SUBORBITAL/KORUSAQ_Merge_Data_1)

15 • NASA TROPOMI NO<sub>2</sub> columns are available at

16 [https://disc.gsfc.nasa.gov/datasets/TROPOMI\\_MINDS\\_NO2\\_1.1/summary?keyw  
17 ords=tropomi%20no2](https://disc.gsfc.nasa.gov/datasets/TROPOMI_MINDS_NO2_1.1/summary?keywords=tropomi%20no2).

18 • KNMI TROPOMI NO<sub>2</sub> columns are available at

19 [https://disc.gsfc.nasa.gov/datasets/S5P\\_L2\\_NO2\\_HiR\\_2/summary?keywords  
20 =tropomi%20no2](https://disc.gsfc.nasa.gov/datasets/S5P_L2_NO2_HiR_2/summary?keywords=tropomi%20no2).

- 1       • CAM-Chem (CESM) code is available at  
2       [https://www.cesm.ucar.edu/models/cesm2/release\\_download.html](https://www.cesm.ucar.edu/models/cesm2/release_download.html).  
3       • WRF-Chem model can be downloaded from  
4       [https://www2.mmm.ucar.edu/wrf/users/download/get\\_sources.html](https://www2.mmm.ucar.edu/wrf/users/download/get_sources.html).

5

### 6   **Author contribution**

7   SWK initiates, designs, analyzes surface monitor data, and writes the manuscript, KMK,  
8   SHS, and SWK design and conduct WRF-Chem model runs, JYJ, JYJ, and SWK design  
9   and conduct CAM-Chem model runs, SHS processes the airkorea data, YSP and SHS  
10   process, analyze, and visualize TROPOMI data, and YSP and JYJ collect and analyze the  
11   highway traffic data. All authors edit the manuscript.

12

### 13   **Competing interests**

14   Authors declare no competing interests.

15

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3 extensive number of observations from ground to aircraft.

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12

13

1 **List of Tables**

2

3 Table 1. Trend estimates based on the 4th highest MDA8 O<sub>3</sub> values from 2001 to 2021.

4 The data were acquired from the surface monitoring network ([www.airkorea.or.kr](http://www.airkorea.or.kr)). Unit of  
5 slope and limit (2 sigma = 2 standard deviation) is ppb yr<sup>-1</sup>. SNR denotes signal-to-noise  
6 ratio defined as the ratio of absolute value of slope to standard deviation. For the use of P-  
7 value and SNR, refer to Chang et al. (2021). Data during typical ozone season (May-  
8 September) are analyzed. Some data for the background sites are missing (refer to SI2).

9

10 Table 2. Spring and summer ozone concentrations in Korean metropolitan cities and  
11 provinces. Both daytime (10-20 LT) and nighttime (01-06 LT) averages are shown.  
12 Standard deviations (S.D.) denoting year-to-year variability are shown in the parenthesis.  
13 Differences in concentrations between spring and summer (O<sub>3 spring</sub> - O<sub>3 summer</sub>) are in the  
14 bracket. The cities and provinces listed in the table are in counterclockwise order in regards  
15 to the South Korean map. The data from 2002 to 2019 are utilized.

16

17 Table 3. The observed trends of NO<sub>2</sub> concentrations in spring and summer from linear fits  
18 of the data covering 2001-2021. The data were acquired from the surface monitoring  
19 network ([www.airkorea.or.kr](http://www.airkorea.or.kr)). Unit of slope and limit (2 sigma = 2 standard deviation) is  
20 ppb yr<sup>-1</sup>. SNR denotes signal-to-noise ratio defined as the ratio of absolute value of slope  
21 to standard deviation. For the use of P-value and SNR, refer to Chang et al. (2021). Due to  
22 data limitations, the trend for the background sites could not be calculated.

23

24 Table 4. The observed trends of CO concentrations in spring and summer from linear fits  
25 of the data covering 2001-2021. The data were acquired from the surface monitoring

1 network ([www.airkorea.or.kr](http://www.airkorea.or.kr)). Unit of slope and limit (2 sigma = 2 standard deviation) is  
2 ppb yr<sup>-1</sup>. SNR denotes signal-to-noise ratio defined as the ratio of absolute value of slope  
3 to standard deviation. For the use of P-value and SNR, refer to Chang et al. (2021). Due to  
4 data limitations, the trend for the background sites could not be calculated.  
5



1 **Figure captions**

2

3 Figure 1. The locations of cities, provinces, and background sites in South Korea. The red,  
4 black, and blue color denote city, province, and background site, respectively: Cities – SUL  
5 (Seoul), INC (Incheon), DJN (Daejeon), GWJ (Gwangju), BSN (Busan), ULS (Ulsan),  
6 DGU (Daegu); Provinces – GGI (Gyeonggi-do), CCB (Chungcheongbuk-do), CCN  
7 (Chungcheongnam-do), JLB (Jeollabuk-do), JLN (Jeollanam-do), JEJ (Jeju Island), GSN  
8 (Gyeongsangnam-do), GSB (Gyeongsangbuk-do), GWO (Gangwon-do); Background  
9 sites – ULL (Ulleung Island), and GSU (Gosung, Gangwon-do).

10

11

12 Figure 2. The trend of the 4<sup>th</sup> highest daily maximum 8 hours average (MDA8) O<sub>3</sub>  
13 concentrations in the South Korean metropolitan cities from 2001 to 2021. Only the data  
14 for May-September (ozone season) are used. Bars denotes standard deviations among the  
15 sites within the city. The slopes (S) and correlation coefficients (r) from linear fits are  
16 shown in parentheses. Grey dashed line indicates 70 ppb that is the air quality standard  
17 defined by the US Environmental Protection Agency.

18

19 Figure 3. The same as in Figure 2 except for South Korean provinces.

20

21 Figure 4. Ratio of O<sub>3</sub> exceedances in summer to exceedances in spring. The exceedances  
22 are defined as the fraction of the data with hourly ozone concentration greater than 70 ppb  
23 among all available data. The red line indicates an one to one line. X-axis denotes names of  
24 cities, provinces, and background sites. Cities – SUL (Seoul), INC (Incheon), DJN

1 (Daejeon), GWJ (Gwangju), BSN (Busan), ULS (Ulsan), DGU (Daegu); Provinces - GGI  
2 (Gyeonggi-do), CCB (Chungcheongbuk-do), CCN (Chungcheongnam-do), JLB  
3 (Jeollabuk-do), JLN (Jeollanam-do), JEJ (Jeju Island), GSN (Gyeongsangnam-do), GSB  
4 (Gyeongsangbuk-do), GWO (Gangwon-do); Background sites - ULL (Ulleung Island),  
5 and GSU (Gosung, Gangwon-do). The data for 2002-2019 are utilized.

6

7 Figure 5. Diurnal O<sub>3</sub> exceedances over selected sites. (Top) cities and province in Seoul  
8 Metropolitan Area, (middle) other cities, (bottom) background sites. The data for 2002-  
9 2019 are utilized.

10

11 Figure 6. The contribution of stratospheric O<sub>3</sub> (O<sub>3s</sub>) to the O<sub>3</sub> concentrations in each season  
12 at surface and 1 km above ground level in South Korea. The plotted values are extracted  
13 from the CESMv2.2 results for the entire country.

14

15 Figure 7. (Top) O<sub>3</sub> exceedances (%), (middle) NO<sub>2</sub>, and (bottom) CO concentrations in  
16 South Korean cities, provinces, and background sites during spring for 2002-2010, 2011-  
17 2019, and 2020-2021 (COVID-19). X-axis denotes names of cities, provinces, and  
18 background sites. Cities - SUL (Seoul), INC (Incheon), DJN (Daejeon), GWJ (Gwangju),  
19 BSN (Busan), ULS (Ulsan), DGU (Daegu); Provinces - GGI (Gyeonggi-do), CCB  
20 (Chungcheongbuk-do), CCN (Chungcheongnam-do), JLB (Jeollabuk-do), JLN  
21 (Jeollanam-do), JEJ (Jeju Island), GSN (Gyeongsangnam-do), GSB (Gyeongsangbuk-do),  
22 GWO (Gangwon-do); Background sites - ULL (Ulleung Island), and GSU (Gosung,  
23 Gangwon-do).

24

1 Figure 8. The same as Figure 7 except for summer.

2

3 Figure 9. Differences in TROPOMI tropospheric NO<sub>2</sub> columns between 2019 and 2020 or  
4 between 2019 and 2021 (Difference = NO<sub>2</sub><sub>2020 or 20-1</sub> - NO<sub>2</sub><sub>2019</sub>). Unit: molecules cm<sup>-2</sup>. Wind  
5 vectors at 700 hPa from ERA-5 are shown for MAM and JJA, respectively. Here, ERA-5  
6 denotes the European Centre for Medium-range Weather Forecasts Reanalysis 5<sup>th</sup>  
7 generation (DOI: 10.24381/cds.6860a573).

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9 Figure 10. Differences in the WRF-Chem simulated ozone concentrations ( $\Delta O_3 =$   
10  $O_3_{\text{emission reduction case}} - O_3_{\text{control case}}$ ) at (top) surface and (bottom) 1000 m above  
11 ground level. Green to blue colors (yellow to red colors) denotes reduced (increased) ozone  
12 concentration due to the emission changes. All model simulation results are utilized.

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14 Figure 11. Vertical profiles of ozone from the WRF-Chem model simulations based on  
15 various emission scenarios: (top) Seoul, and (bottom) Gosung, Gangwon-do. The model  
16 results from 10 LT to 20 LT are averaged.

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1 Table 1. Trend estimates based on the 4<sup>th</sup> highest MDA8 O<sub>3</sub> values from 2001 to 2021. The  
 2 data were acquired from the surface monitoring network ([www.airkorea.or.kr](http://www.airkorea.or.kr)). Unit of  
 3 slope and limit (2 sigma = 2 standard deviation) is ppb yr<sup>-1</sup>. SNR denotes signal-to-noise  
 4 ratio defined as the ratio of absolute value of slope to standard deviation. For the use of P-  
 5 value and SNR, refer to Chang et al. (2021). Data during typical ozone season (May-  
 6 September) are analyzed. Some data for the background sites are missing (refer to SI2).

Location		Slope (ppb yr <sup>-1</sup> )	2-Sigma (ppb yr <sup>-1</sup> )	P value	SNR
City	Seoul (SUL)	1.19	0.38	< 0.01	6.23
	Incheon (INC)	1.07	0.37	< 0.01	5.72
	Daejeon (DJN)	1.22	0.49	< 0.01	4.96
	Gwangju (GWJ)	0.98	0.46	< 0.01	4.30
	Busan (BSN)	0.98	0.36	< 0.01	5.47
	Ulsan (ULS)	1.40	0.34	< 0.01	8.14
	Daegu (DGU)	1.12	0.46	< 0.01	4.89
Province	Gyeonggi-do (GGI)	1.26	0.27	< 0.01	9.33
	Chungcheongbuk-do (CCB)	0.79	0.51	< 0.01	3.09
	Chungcheongnam-do (CCN)	1.45	0.47	< 0.01	6.12
	Jeollabuk-do (JLB)	1.83	0.32	< 0.01	11.30
	Jeollanam-do (JLN)	0.08	0.39	0.67	0.41
	Jeju Island (JEJ)	0.74	0.50	< 0.01	2.96
	Gyeongsangnam-do (GSN)	0.83	0.52	< 0.01	3.18
	Gyeongsangbuk-do (GSB)	1.10	0.35	< 0.01	6.32
	Gangwon-do (GWO)	0.74	0.46	< 0.01	3.22
Background	Ulleung Island (ULL)	0.42	0.84	0.34	1.00
	(2001-2019)	(0.92)	(0.92)	(0.06)	(2.00)
	Gosung (GSU)	0.73	0.82	0.09	1.78
	(2001-2019)	(1.45)	(0.75)	(<0.01)	(3.88)

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1 Table 2. Spring and summer ozone concentrations in Korean metropolitan cities and  
 2 provinces. Both daytime (10-20 LT) and nighttime (01-06 LT) averages are shown.  
 3 Standard deviations (S.D.) denoting year-to-year variability are shown in the parenthesis.  
 4 Differences in concentrations between spring and summer ( $O_3$  spring -  $O_3$  summer) are in the  
 5 bracket. The cities and provinces listed in the table are in counterclockwise order in regards  
 6 to the South Korean map. The data from 2002 to 2019 are utilized.

Location		Daytime period	Nighttime period
		Spring / Summer (S.D.) [Difference]	Spring / Summer (S.D.) [Difference]
City	Seoul (SUL)	34.7(5.4) / 35.5(6.0) [-0.8]	20.5(3.8) / 17.0(4.3) [3.5]
	Incheon (INC)	34.7(4.0) / 32.7(4.8) [2.0]	25.1(2.9) / 19.7(3.8) [5.4]
	Daejeon (DJN)	41.7(6.4) / 36.7(7.1) [5.0]	22.9(3.3) / 18.7(3.8) [4.2]
	Gwangju (GWJ)	40.1(6.0) / 35.2(5.7) [4.9]	28.5(5.3) / 23.6(4.2) [4.9]
	Busan (BSN)	40.5(4.2) / 33.7(4.2) [6.8]	30.3(3.1) / 21.9(3.0) [8.4]
	Ulsan (ULS)	38.9(5.8) / 33.1(6.1) [5.8]	25.7(2.9) / 18.2(3.2) [7.5]
	Daegu (DGU)	40.0(5.8) / 37.3(5.9) [2.7]	24.0(3.8) / 19.1(3.5) [4.9]
Province	Gyeonggi-do (GGI)	37.8(4.6) / 38.1(6.0) [-0.3]	20.7(3.0) / 17.4(3.7) [3.3]
	Chungcheongbuk-do (CCB)	42.6(4.1) / 39.6 (4.6) [3.0]	24.9(3.6) / 20.5(3.2) [4.4]
	Chungcheongnam-do (CCN)	41.4(4.7) / 37.0(7.1) [4.4]	29.3(3.8) / 22.3(4.6) [7.0]
	Jeollabuk-do (JLB)	38.3(8.1) / 34.3(8.0) [4.0]	26.6(5.7) / 22.8(5.4) [3.8]
	Jeollanam-do (JLN)	43.1(4.2) / 34.9(4.0) [8.2]	33.4(3.4) / 23.9(2.7) [9.5]
	Jeju Island (JEJ)	49.3(6.0) / 34.2(4.3) [15.1]	44.0(5.7) / 28.6(3.9) [15.4]
	Gyeongsangnam-do (GSN)	44.7(4.9) / 39.8 (5.2) [4.9]	28.9(3.1) / 21.6(2.6) [7.3]
	Gyeongsangbuk-do (GSB)	45.5(5.3) / 37.5(5.4) [8.0]	28.5(4.2) / 20.1(3.3) [8.4]
Background	Gangwon-do (GWO)	44.5(4.0) / 39.1(4.7) [5.4]	27.9(2.9) / 20.5(3.0) [7.4]
	Ulleung Island (ULL)	56.6(7.2) / 43.9(6.1) [12.7]	55.9(6.5) / 43.1(6.0) [12.8]
	Gosung (GSU)	58.3(8.4) / 42.9(4.9) [15.4]	58.1(8.8) / 44.9(5.3) [13.2]

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2 Table 3. The observed trends of NO<sub>2</sub> concentrations in spring and summer from linear fits  
 3 of the data covering 2001-2021. The data were acquired from the surface monitoring  
 4 network ([www.airkorea.or.kr](http://www.airkorea.or.kr)). Unit of slope and limit (2 sigma = 2 standard deviation) is  
 5 ppb yr<sup>-1</sup>. SNR denotes signal-to-noise ratio defined as the ratio of absolute value of slope  
 6 to standard deviation. For the use of P-value and SNR, refer to Chang et al. (2021). Due to  
 7 data limitations, the trend for the background sites could not be calculated.

Stations		NO <sub>2</sub> Spring (Summer)			
		Slope (ppb yr <sup>-1</sup> )	2 Sigma (ppb yr <sup>-1</sup> )	P-value	SNR
City	Seoul (SUL)	-0.77 (-0.72)	0.22 (0.15)	< 0.01 (< 0.01)	6.94 (9.57)
	Incheon (INC)	-0.37 (-0.50)	0.22 (0.17)	< 0.01 (< 0.01)	3.36 (5.88)
	Daejeon (DJN)	-0.10 (-0.12)	0.14 (0.09)	0.21 (0.02)	1.43 (2.53)
	Gwangju (GWJ)	-0.51 (-0.35)	0.15 (0.09)	< 0.01 (< 0.01)	6.94 (7.74)
	Busan (BSN)	-0.64 (-0.49)	0.16 (0.11)	< 0.01 (< 0.01)	8.12 (8.93)
	Ulsan (ULS)	-0.04 (-0.06)	0.23 (0.19)	0.73 (0.51)	0.34 (0.63)
	Daegu (DGU)	-0.65 (-0.51)	0.18 (0.13)	< 0.01 (< 0.01)	7.21 (8.15)
	Province	Gyeonggi (GGI)	-0.41(-0.44)	0.22 (0.16)	< 0.01 (< 0.01)
Chungcheongbuk (CCB)		-0.18(-0.16)	0.20 (0.15)	0.09 (0.05)	1.82 (2.15)
Chungcheongnam (CCN)		-0.10(-0.12)	0.15 (0.12)	0.21 (0.08)	1.38 (1.97)
Jeollabuk (JLB)		-0.17(-0.25)	0.18 (0.14)	0.08 (< 0.01)	1.90 (3.61)
Jeollanam (JLN)		-0.21(-0.21)	0.16 (0.14)	0.02 (< 0.01)	2.56 (2.95)
Jeju Island (JEJ)		-0.18(-0.16)	0.20 (0.15)	0.10 (0.04)	1.76 (2.20)
Gyeongsangnam (GSN)		-0.12(-0.10)	0.17 (0.11)	0.18 (0.08)	1.42 (1.88)
Gyeongsangbuk (GSB)		-0.76(-0.49)	0.18 (0.13)	< 0.01 (< 0.01)	8.47 (7.74)
Gangwon (GWO)		-0.16(-0.20)	0.14 (0.10)	0.03 (< 0.01)	2.37 (4.18)

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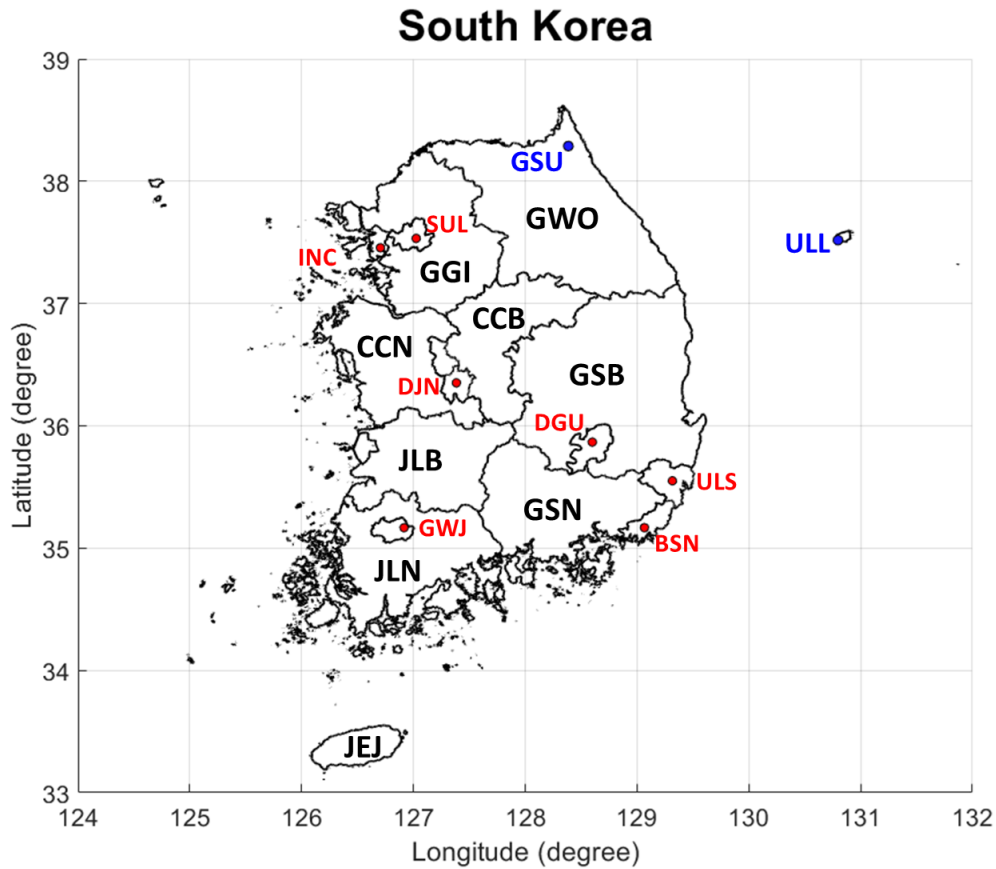
2 Table 4. The observed trends of CO concentrations in spring and summer from linear fits  
 3 of the data covering 2001-2021. The data were acquired from the surface monitoring  
 4 network ([www.airkorea.or.kr](http://www.airkorea.or.kr)). Unit of slope and limit (2 sigma = 2 standard deviation) is  
 5 ppb yr<sup>-1</sup>. SNR denotes signal-to-noise ratio defined as the ratio of absolute value of slope  
 6 to standard deviation. For the use of P-value and SNR, refer to Chang et al. (2021). Due to  
 7 data limitations, the trend for the background sites could not be calculated.

Stations		CO Spring (Summer)			
		Slope (ppb yr <sup>-1</sup> )	2 Sigma (ppb yr <sup>-1</sup> )	P-value	SNR
City	Seoul (SUL)	-7.56 (-5.34)	2.94 (1.66)	< 0.01 (< 0.01)	5.15 (6.44)
	Incheon (INC)	-7.65 (-4.64)	3.62 (2.46)	< 0.01 (< 0.01)	4.23 (3.77)
	Daejeon (DJN)	-15.53 (-9.71)	5.68 (5.56)	< 0.01 (< 0.01)	5.47 (3.49)
	Gwangju (GWJ)	-10.64 (-8.00)	3.60 (3.94)	< 0.01 (< 0.01)	5.91 (4.06)
	Busan (BSN)	-12.32 (-11.05)	3.90 (3.80)	< 0.01 (< 0.01)	6.32 (5.82)
	Ulsan (ULS)	-4.80 (0.75)	5.54 (5.28)	0.10 (0.78)	1.73 (0.28)
	Daegu (DGU)	-23.49 (-19.87)	5.50 (5.30)	< 0.01 (< 0.01)	8.54 (7.50)
Province	Gyeonggi (GGI)	-14.50 (-8.82)	2.18 (1.54)	< 0.01 (< 0.01)	13.30 (11.42)
	Chungcheongbuk (CCB)	-17.68 (-6.49)	6.70 (3.92)	< 0.01 (< 0.01)	5.28 (3.31)
	Chungcheongnam (CCN)	-20.95 (-9.33)	8.32 (4.62)	< 0.01 (< 0.01)	5.04 (4.04)
	Jeollabuk (JLB)	-21.33 (-15.07)	5.88 (4.34)	< 0.01 (< 0.01)	7.26 (6.95)
	Jeollanam (JLN)	-5.86 (-5.32)	4.40 (4.60)	0.02 (0.03)	2.66 (2.31)
	Jeju Island (JEJ)	-10.74 (-6.95)	5.00 (5.64)	< 0.01 (0.02)	4.30 (2.46)
	Gyeongsangnam (GSN)	-6.76 (-3.92)	4.44 (3.58)	< 0.01 (0.04)	3.04 (2.19)
	Gyeongsangbuk (GSB)	-27.54 (-17.48)	9.00 (6.64)	< 0.01 (< 0.01)	6.12 (5.27)
	Gangwon (GWO)	-15.31 (-9.03)	4.34 (4.16)	< 0.01 (< 0.01)	7.05 (4.34)

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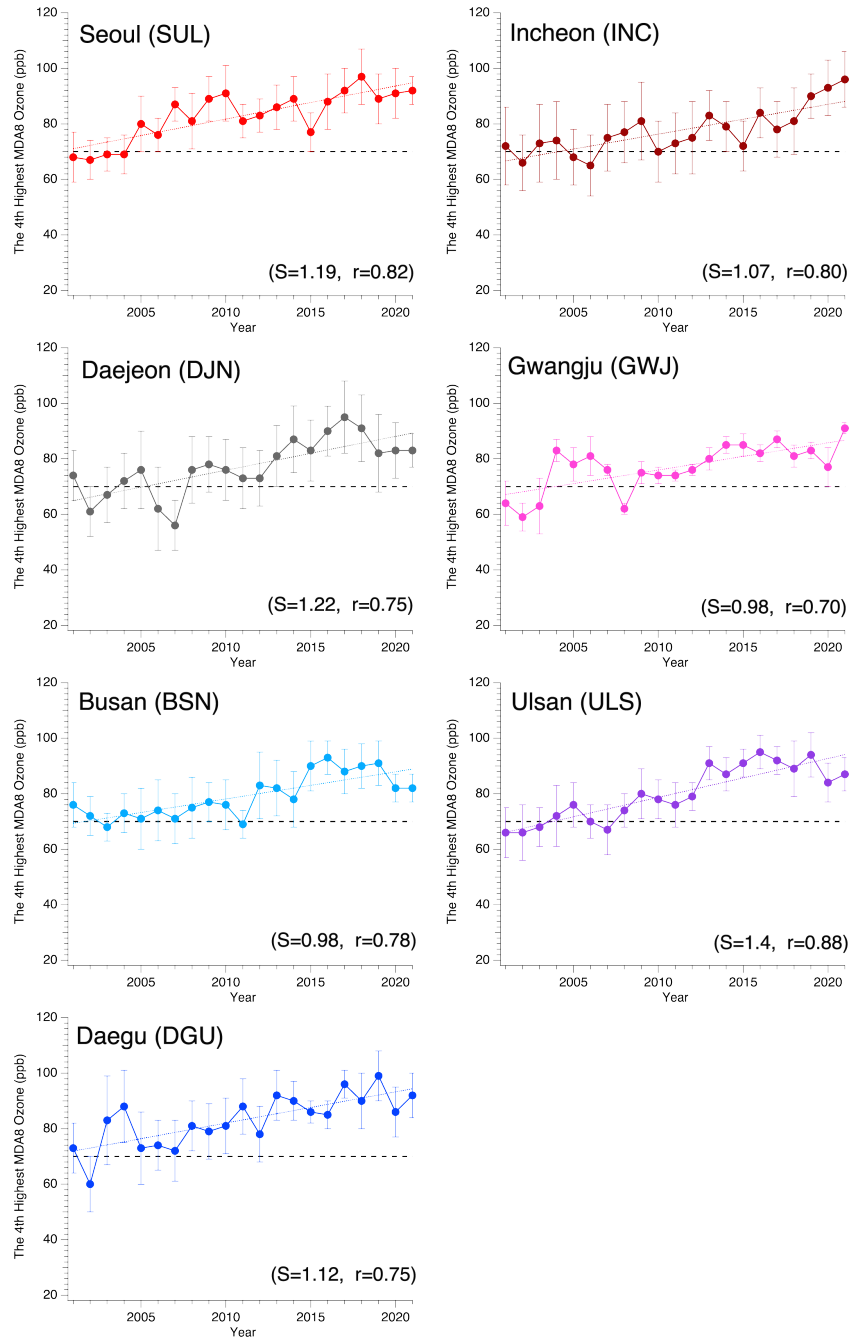
2 Figure 1. The locations of cities, provinces, and background sites in South Korea. The red,  
 3 black, and blue color denote city, province, and background site, respectively: Cities – SUL  
 4 (Seoul), INC (Incheon), DJN (Daejeon), GWJ (Gwangju), BSN (Busan), ULS (Ulsan),  
 5 DGU (Daegu); Provinces - GGI (Gyeonggi-do), CCB (Chungcheongbuk-do), CCN  
 6 (Chungcheongnam-do), JLB (Jeollabuk-do), JLN (Jeollanam-do), JEJ (Jeju Island), GSN  
 7 (Gyeongsangnam-do), GSB (Gyeongsangbuk-do), GWO (Gangwon-do); Background  
 8 sites - ULL (Ulleung Island), and GSU (Gosung, Gangwon-do).

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Figure 2. The trend of the 4<sup>th</sup> highest daily maximum 8 hours average (MDA8) O<sub>3</sub> concentrations in the South Korean metropolitan cities from 2001 to 2021. Only the data for May-September (ozone season) are used. Bars denote standard deviations among the sites within the city. The slopes (S) and correlation coefficient (r) from linear fits are shown in parentheses. Grey dashed line indicates 70 ppb that is the air quality standard defined by the US Environmental Protection Agency.

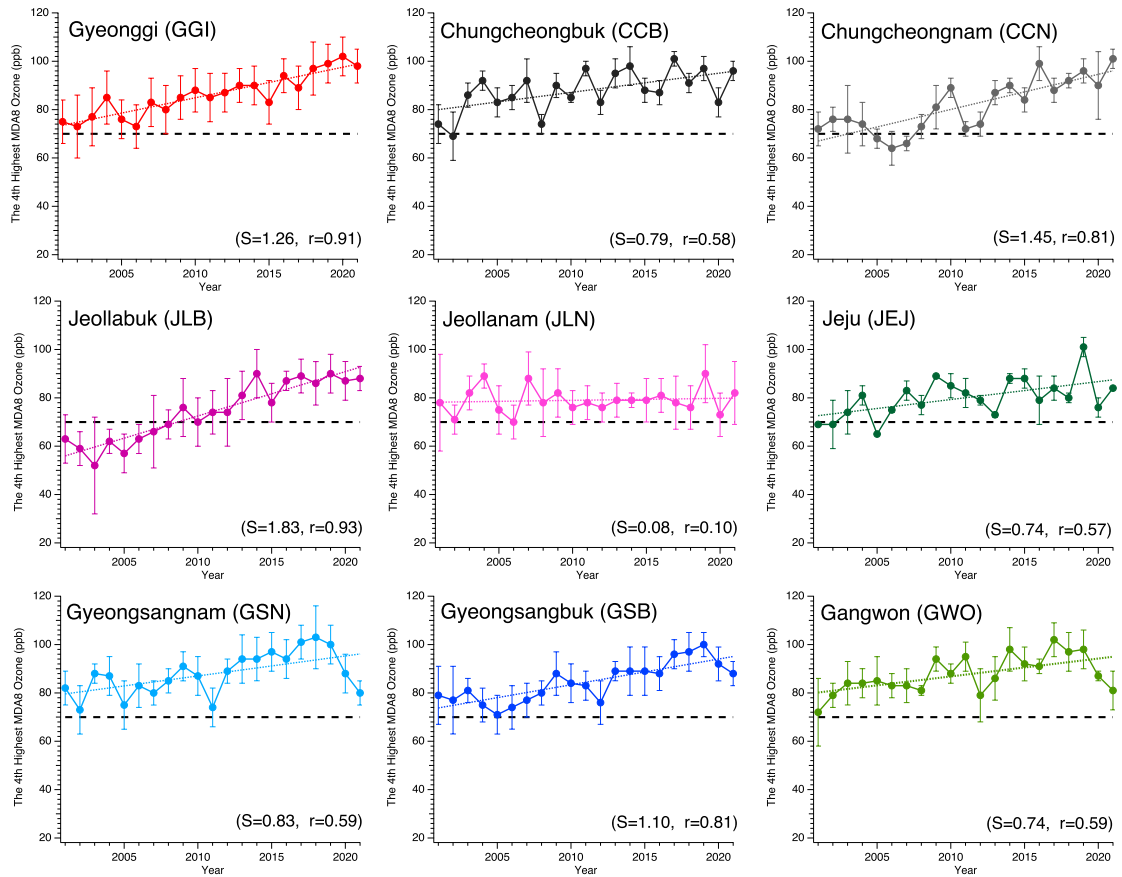
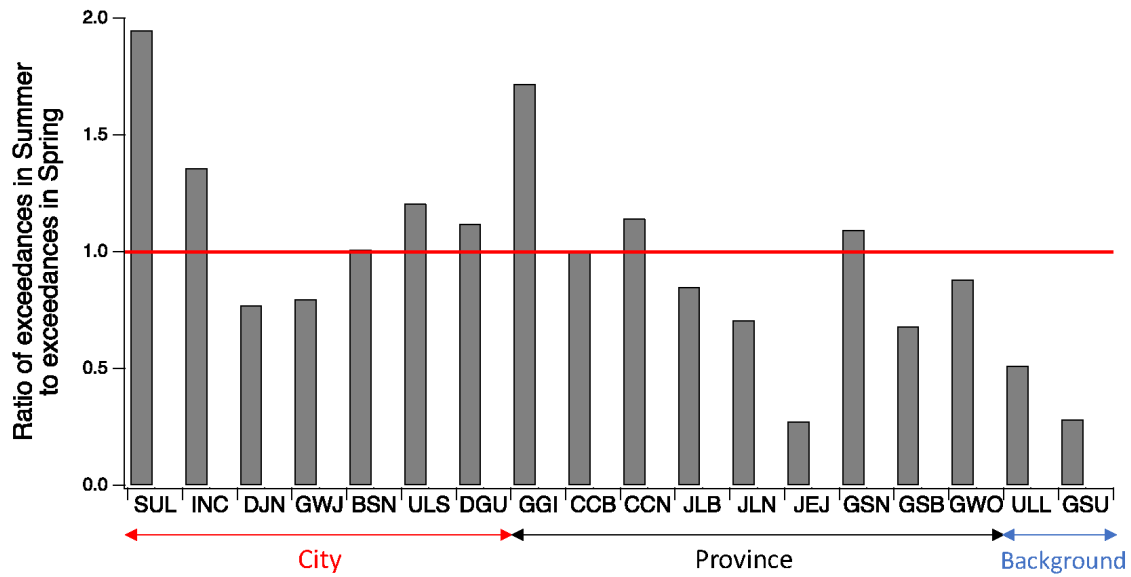


Figure 3. The same as in Figure 2 except for South Korean provinces.

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2 Figure 4. Ratio of O<sub>3</sub> exceedances in summer to exceedances in spring. The exceedances  
 3 are defined as the fraction of the data with hourly ozone concentration greater than 70 ppb  
 4 among all available data. The red line indicates an one to one line. X-axis denotes names  
 5 of cities, provinces, and background sites. Cities – SUL (Seoul), INC (Incheon), DJN  
 6 (Daejeon), GWJ (Gwangju), BSN (Busan), ULS (Ulsan), DGU (Daegu); Provinces - GGI  
 7 (Gyeonggi-do), CCB (Chungcheongbuk-do), CCN (Chungcheongnam-do), JLB  
 8 (Jeollabuk-do), JLN (Jeollanam-do), JEJ (Jeju Island), GSN (Gyeongsangnam-do), GSB  
 9 (Gyeongsangbuk-do), GWO (Gangwon-do); Background sites - ULL (Ulleung Island),  
 10 and GSU (Gosung, Gangwon-do). The data for 2002-2019 are utilized.

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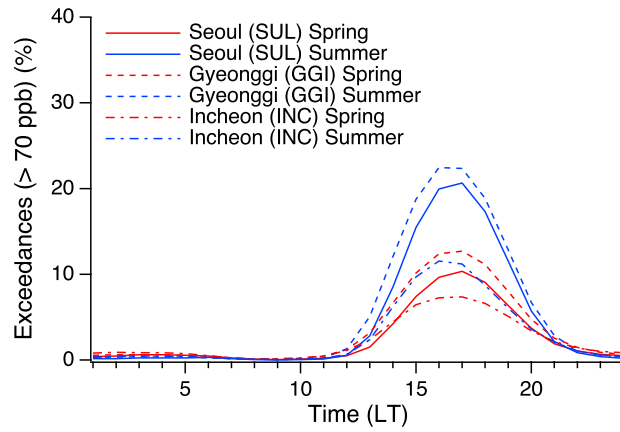
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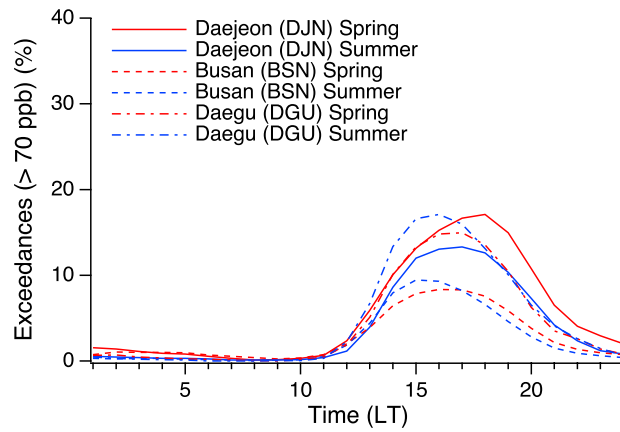
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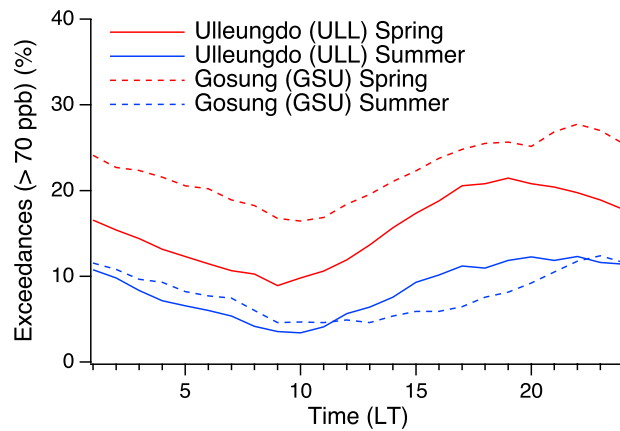
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5 Figure 5. Diurnal O<sub>3</sub> exceedances over selected sites. (Top) cities and province in Seoul  
6 Metropolitan Area, (middle) other cities, (bottom) background sites. The data for 2002-  
7 2019 are utilized.

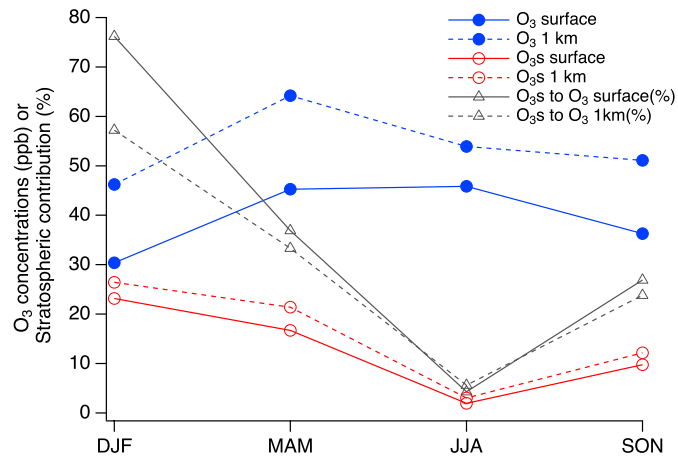
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2 Figure 6. The contribution of stratospheric O<sub>3</sub> (O<sub>3s</sub>) to the O<sub>3</sub> concentrations in each season  
 3 at surface and 1 km above ground level in South Korea. The plotted values are extracted  
 4 from the CESMv2.2 results for the entire country.

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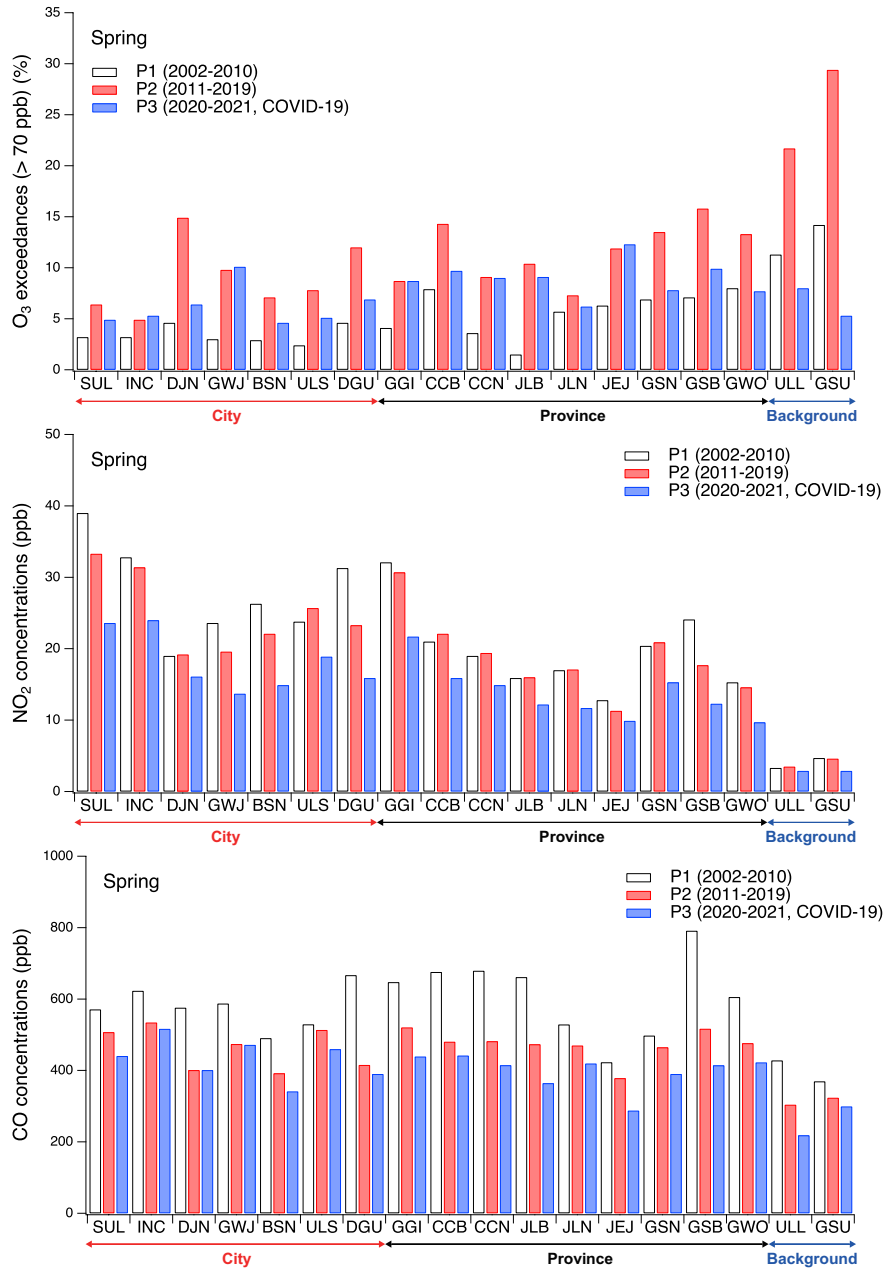
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 2 Figure 7. (Top) O<sub>3</sub> exceedances (%), (middle) NO<sub>2</sub>, and (bottom) CO concentrations in  
 3 South Korean cities, provinces, and background sites during spring for 2002-2010, 2011-  
 4 2019, and 2020-2021 (COVID-19). X-axis denotes names of cities, provinces, and  
 5 background sites. Cities - SUL (Seoul), INC (Incheon), DJN (Daejeon), GWJ (Gwangju),  
 6 BSN (Busan), ULS (Ulsan), DGU (Daegu); Provinces - GGI (Gyeonggi-do), CCB  
 7 (Chungcheongbuk-do), CCN (Chungcheongnam-do), JLB (Jeollabuk-do), JLN  
 8 (Jeollanam-do), JEJ (Jeju Island), GSN (Gyeongsangnam-do), GSB (Gyeongsangbuk-do),  
 9 GWO (Gangwon-do); Background sites - ULL (Ulleung Island), and GSU (Gosung,  
 10 Gangwon-do).

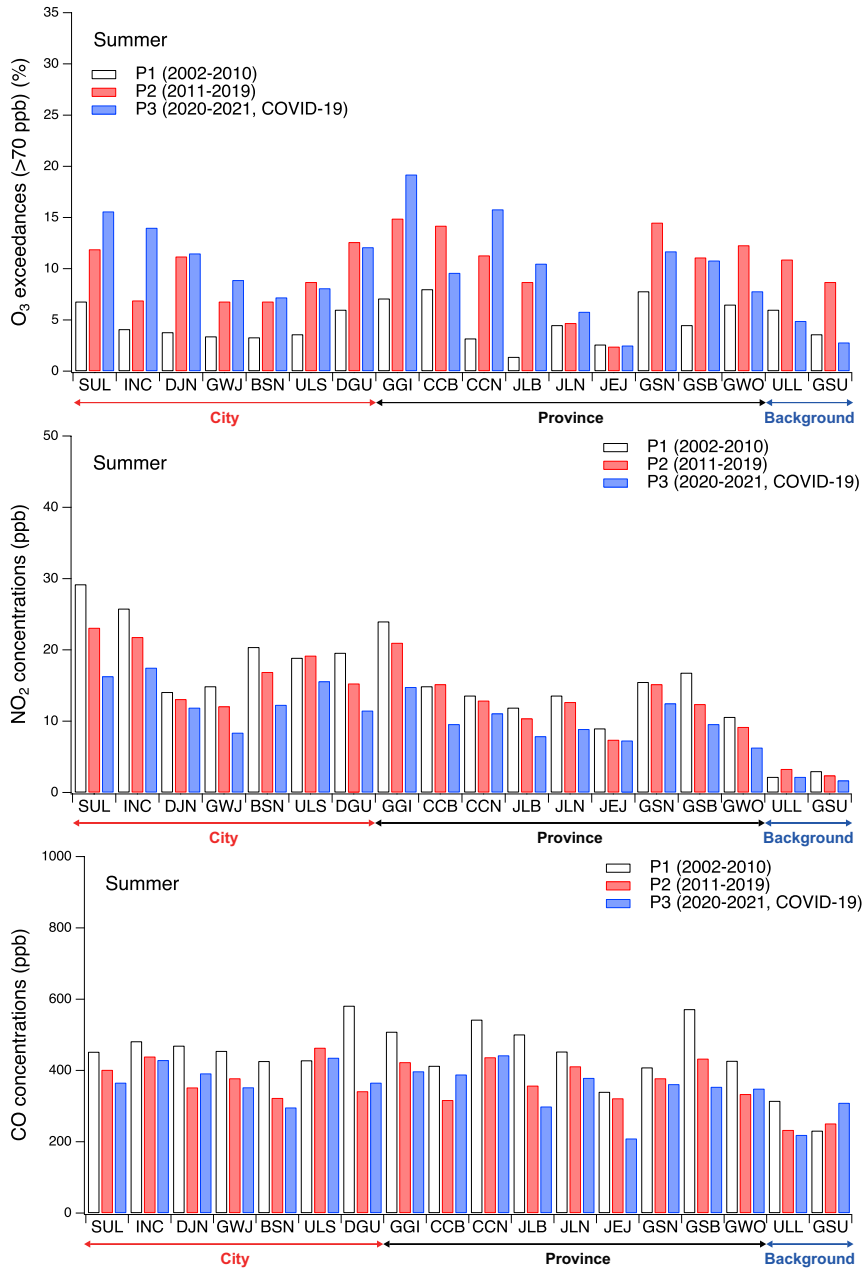
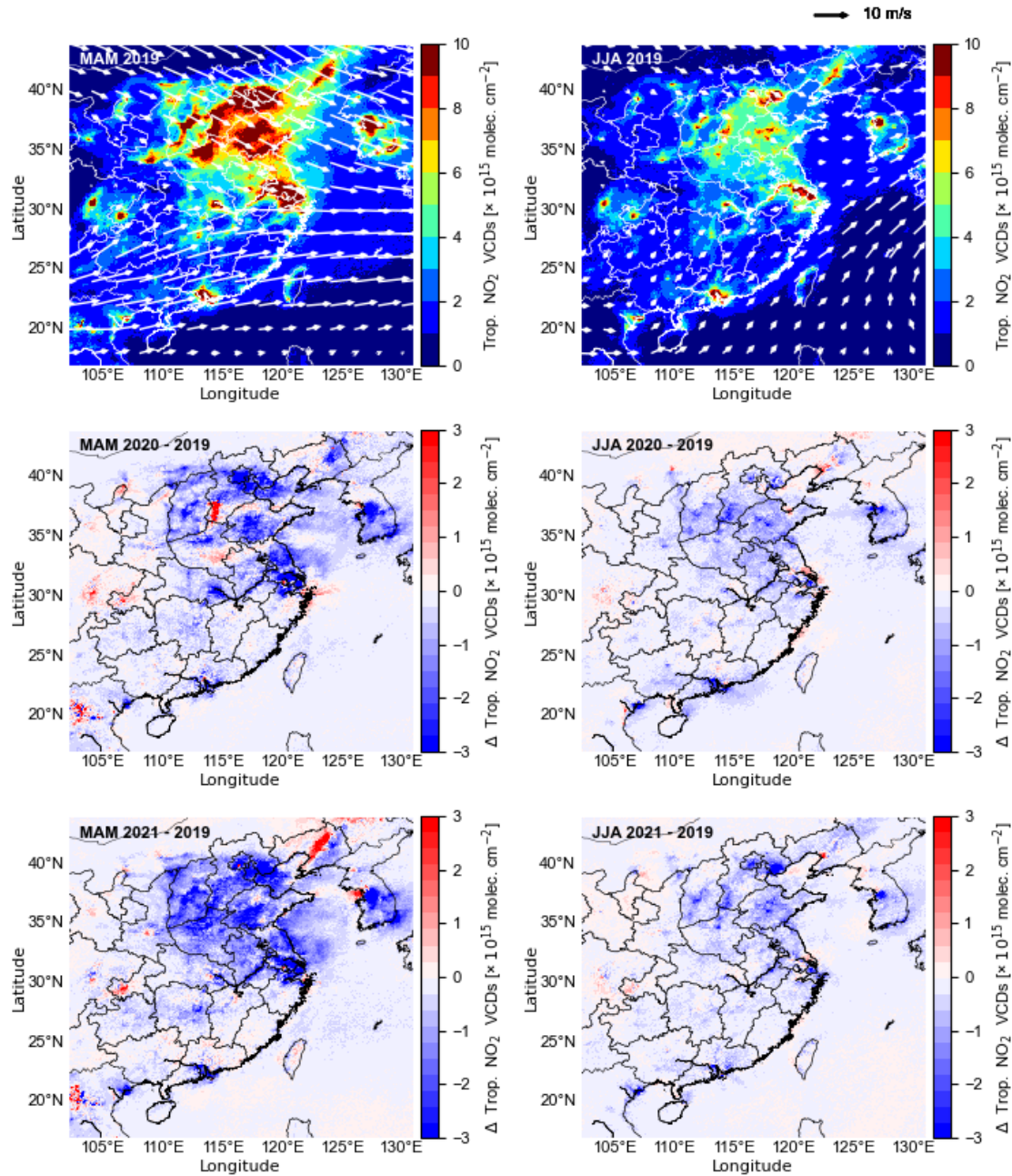


Figure 8. The same as Figure 7 except for summer.

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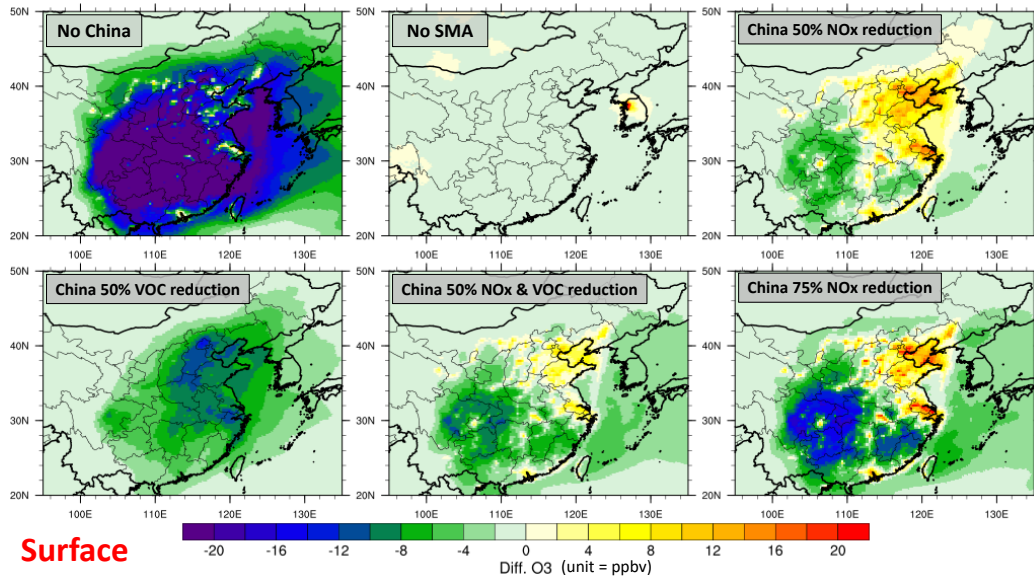
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2 Figure 9. Differences in TROPOMI tropospheric NO<sub>2</sub> columns between 2019 and 2020 or  
 3 between 2019 and 2021 (Difference = NO<sub>2</sub> 2020 or 2021 - NO<sub>2</sub> 2019). Unit: molecules cm<sup>-2</sup>. Wind  
 4 vectors at 700 hPa from ERA-5 are shown for MAM and JJA, respectively. Here, ERA-5  
 5 denotes the European Centre for Medium-range Weather Forecasts Reanalysis 5<sup>th</sup>  
 6 generation (DOI: 10.24381/cds.6860a573).

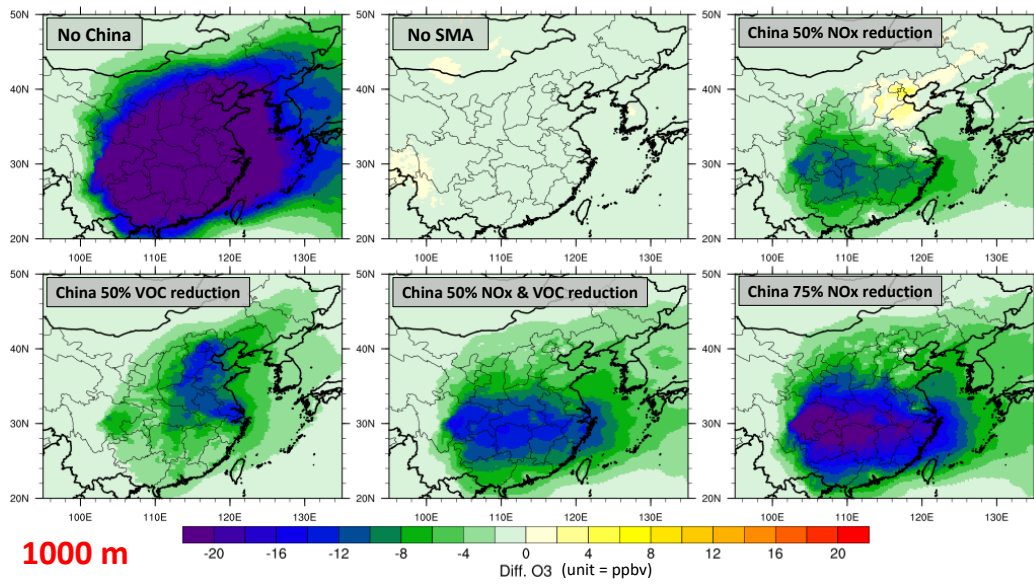
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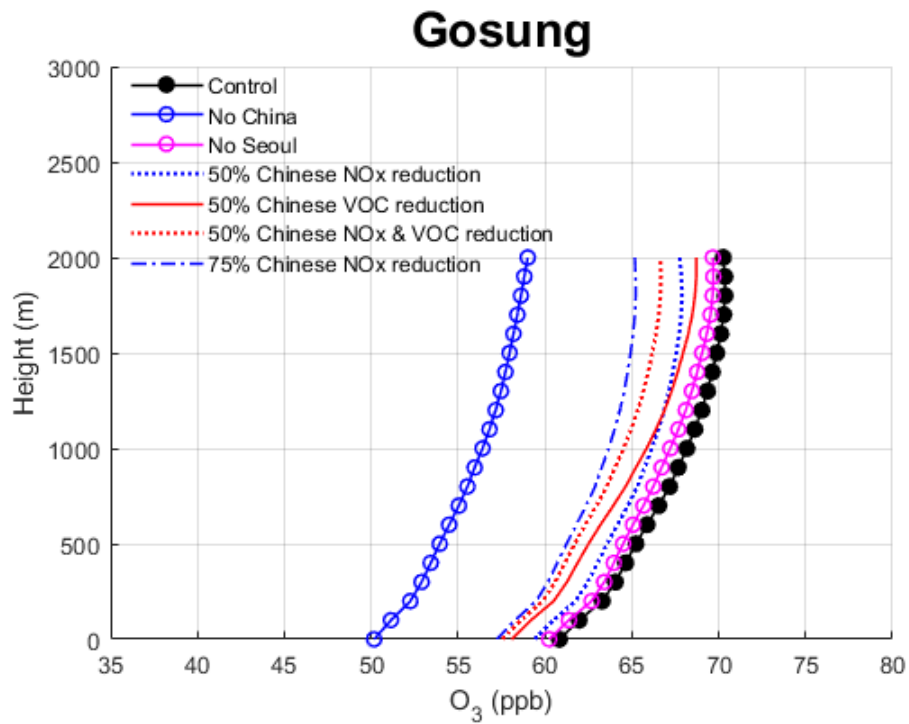
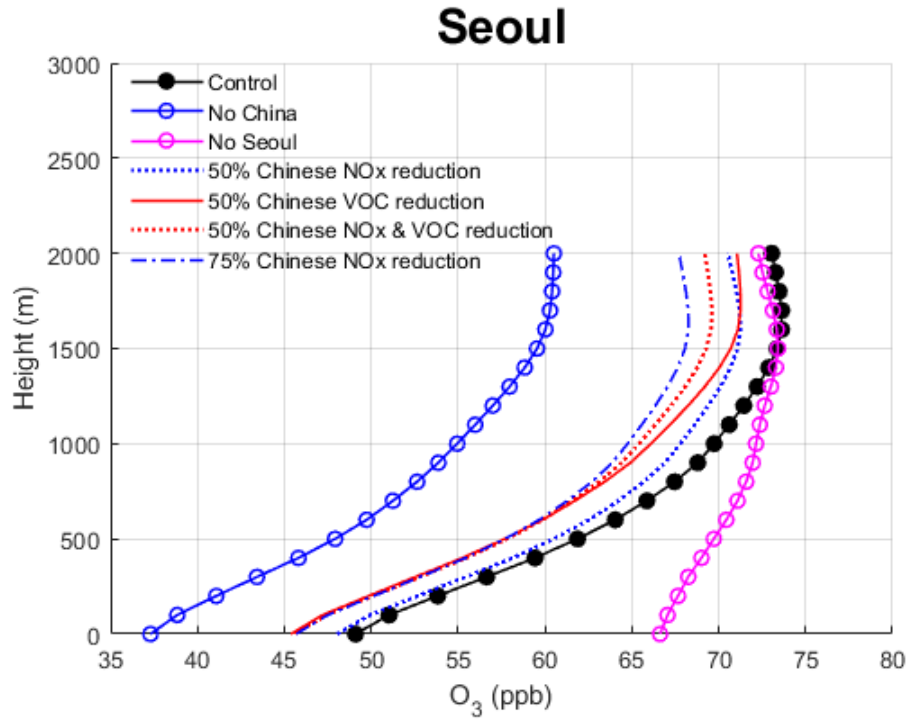


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4 Figure 10. Differences in the WRF-Chem simulated ozone concentrations ( $\Delta O_3 =$   
 5  $O_3_{\text{emission reduction case}} - O_3_{\text{control case}}$ ) at (top) surface and (bottom) 1000 m above  
 6 ground level. Green to blue colors (yellow to red colors) denotes reduced (increased) ozone  
 7 concentration due to the emission changes. All model simulation results are utilized.



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 2 Figure 11. Vertical profiles of ozone from the WRF-Chem model simulations based on  
 3 various emission scenarios: (top) Seoul, and (bottom) Gosung, Gangwon-do. The model  
 4 results from 10 LT to 20 LT are averaged.  
 5