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Large and increasing stratospheric contribution to tropospheric ozone over East Asia

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Abstract. Severe surface ozone pollution in South Korea and China in May-June is due in part to an elevated background subsiding from the free troposphere (750-350 hPa). Using IAGOS commercial aircraft observations and the GEOS-Chem model, we show that free tropospheric ozone over East Asia in May-June is the highest in the world and has increased from 68±3 ppb (mean and interannual standard deviation) in 2000-2004 to 78±4 ppb in 2015-2019. Free tropospheric ozone over East Asia is highest when carbon monoxide (CO) is low, both in the observations and GEOS-Chem, implying a large stratospheric influence on ozone. We find from GEOS-Chem that East Asia is a global hotspot for stratospheric downwelling of ozone and that this makes a major contribution to the free tropospheric ozone over the region in May-June. Stratospheric downwelling of ozone over East Asia in GEOS-Chem increased by 40% from 2000-2004 to 2015-2019, which can explain the observed free tropospheric ozone increase over this period. Increased stratospheric downwelling over East Asia appears to be driven by a strengthening of the jet stream. The large and increasing stratospheric contribution to the surface ozone background over East Asia is a major impediment to meeting ozone air quality standards.

1 Introduction

Surface ozone concentrations in China and South Korea are among the highest in the world and have risen over the past two decades (Fleming et al., 2018; Lu et al., 2018; Wang et al., 2022; Kim et al., 2023). Stringent emission control policies in both countries have succeeded in decreasing fine particulate matter concentrations ($PM_{2.5}$) (Bae et al., 2021; Pendergrass et al., 2022; Lee et al., 2024) but not surface ozone, which routinely exceeds national air

https://doi.org/10.5194/egusphere-2025-1799
Preprint. Discussion started: 7 May 2025

quality standards (80 ppb in China, 60 ppb in South Korea). Understanding the role of both domestic and background influences on surface ozone in East Asia is critical for developing adequate emission control strategies to improve air quality.

Background surface ozone for a given region is commonly defined as the ozone that would be present in the absence of anthropogenic emissions in that region. A major contribution is subsidence from the free troposphere, which caps the planetary boundary layer (PBL) at about 2 km altitude. Surface ozone in South Korea and much of China is highest in May-June (Lu et al., 2019; Li et al., 2021; Oak et al., 2025). The KORUS-AQ aircraft campaign in May-June 2016 found free tropospheric ozone concentrations averaging in excess of 80 ppb over and around the Korean peninsula (Miyazaki et al., 2018; Sullivan et al., 2019, Gaubert et al., 2020; Crawford et al., 2021; Park et al., 2021). A model study by Colombi et al (2023) found that subsidence of this elevated free tropospheric ozone drove surface background ozone in East Asia in May to exceed 50 ppb, considerably higher than the 20-40 ppb background values in North America and Europe (Fiore et al., 2003; Zhang et al., 2011; Emery et al., 2012; Jaffe et al., 2018). Ozonesondes and commercial aircraft observations also show a maximum of free tropospheric ozone over East Asia in late spring (Chen at al., 2023) that has been increasing in the last two decades (Wang et al., 2022). This elevated background could reflect influences from the stratosphere (Ma et al., 2024; Hong et al., 2024; Luo et al., 2024), lightning (Murray et al., 2016; Ye et al., 2024), and pollution originating outside East Asia (Han et al., 2019; Chen et al., 2023).

Here we aim to understand the sources driving the seasonality and multi-decadal (2000-2019) change in free tropospheric ozone over East Asia, and the implication for surface ozone concentrations. We examine seasonality and change over time of the ozone vertical distribution, putting it into context of other northern midlatitude regions. We use the relationship between ozone and carbon monoxide (CO) to separate anthropogenic and natural contributions to free tropospheric ozone. Finally, we use the GEOS-Chem global 3-D chemical transport model to attribute the sources of free tropospheric ozone and the increasing trend.

2 Observed free tropospheric ozone seasonality, trend, and relationship with CO

We use ozone profile data from the In-Service Aircraft for a Global Observing System (IAGOS, https://iagos.aeris-data.fr/) (Petzold et al., 2015) to compare the free tropospheric ozone seasonality and trends over three northern mid-latitude regions: East Asia (30.0-45.0 ° N, 109.0-145.0 ° E), Europe (35.0-54.0 ° N, -11.0-30.0 ° E), and the United States (28.0-49.0 ° N, -124.6.0- -66.8 ° E). The IAGOS measurements are made using a dual-beam ultraviolet absorption photometer, with an accuracy of ±2 ppbv (Blot et al., 2021; Nédélec et al., 2015). We average the ozone profiles vertically in 25 hPa bins for further analysis.

https://doi.org/10.5194/egusphere-2025-1799
Preprint. Discussion started: 7 May 2025

Figure 1 shows the mean ozone mixing ratio profiles for the three regions for 2000-2004 and 2015-2019 in March-April, May-June, and July-August. The numbers inset are mean column mixing ratios and standard deviations for the free tropospheric column, which we define here and elsewhere as the pressure range 750- 350 hPa. While Europe and the United States show negligible change in the free troposphere over the period, East Asia shows an increase peaking in May-June. Free tropospheric ozone in East Asia is higher than in Europe or the United States for all seasons, and by up to 15 ppb in May-June 2015-2019 when it averages 78 ppb.

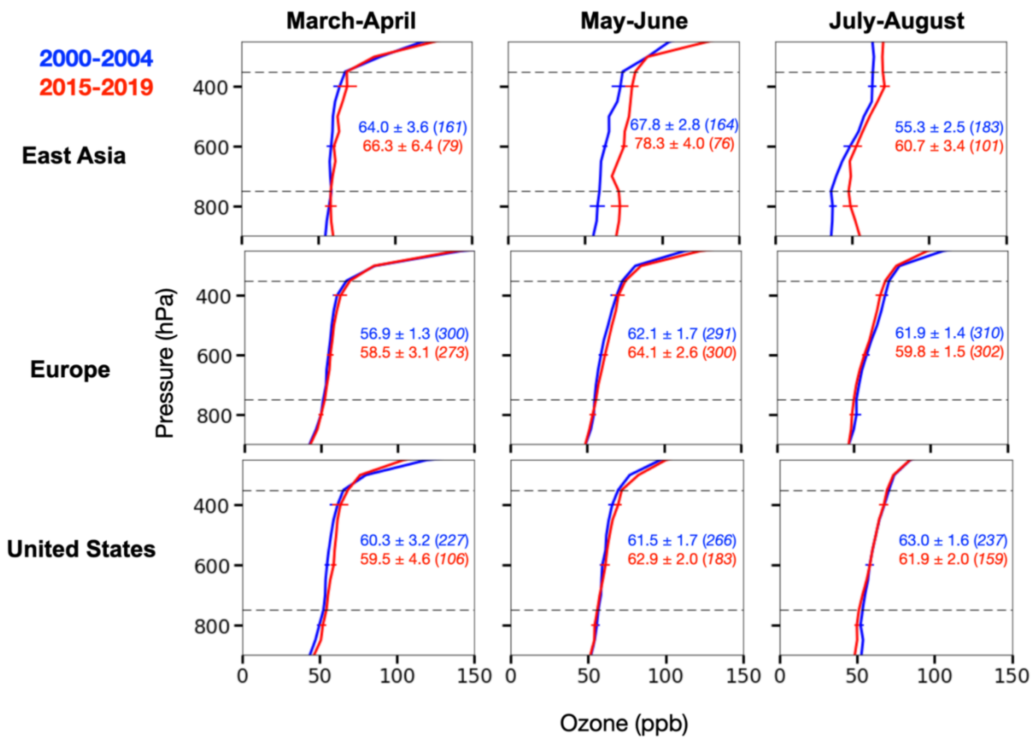

Figure 1 Mean IAGOS vertical profiles of ozone mixing ratios over East Asia (30.0-45.0 ° N, 109.0-145.0 ° E), Europe (35.0-54.0 ° N, -11.0-30.0 ° E), and the United States (28.0-49.0 ° N, -124.6.0- -66.8 ° E) for 2000-2004 and 2015-2019. Average and standard deviations of ozone column mixing ratios in the free troposphere (750-350 hPa) are shown with numbers of profiles in italics. Horizontal bars are standard deviations for selected 25 hPa vertical pressure levels. Dashed lines are the 750 hPa and 350 hPa pressure levels defining the free troposphere for the purpose of our work.

To investigate the specific conditions that contribute to elevated free tropospheric ozone over East Asia during May-June, we leverage the simultaneous measurements of CO from IAGOS. Collocated measurements of ozone and CO

https://doi.org/10.5194/egusphere-2025-1799
Preprint. Discussion started: 7 May 2025

have been used extensively as an indicator for stratospheric influence, since stratospheric air is rich in ozone and low in CO (Stoll et al., 2003; Knowland et al., 2017; Dreessen et al., 2019; Chen et al., 2022). Figure 2 shows mean ozone concentrations within 5 ppb CO increments in the IAGOS column data for 750-350 hPa. We find that ozone in the 2015-2019 data is highest under clean conditions (CO < 100 ppb), but not in the 2000-2004 data. This strongly suggests increasing stratospheric influence and a large contribution of the stratosphere in the 2015-2019 data. The ozone drop for the lowest CO values (< 70 ppb) likely reflects tropical air. We see no enhancement of ozone under polluted conditions with high CO, which would reflect PBL venting or fire influences, and this can be simply explained by the mean free tropospheric ozone being high relative to typical ozone levels in the polluted PBL or fire plumes. The association of high ozone with low CO is not seen in the free tropospheric data for Europe or the United States (SI Figure 1).

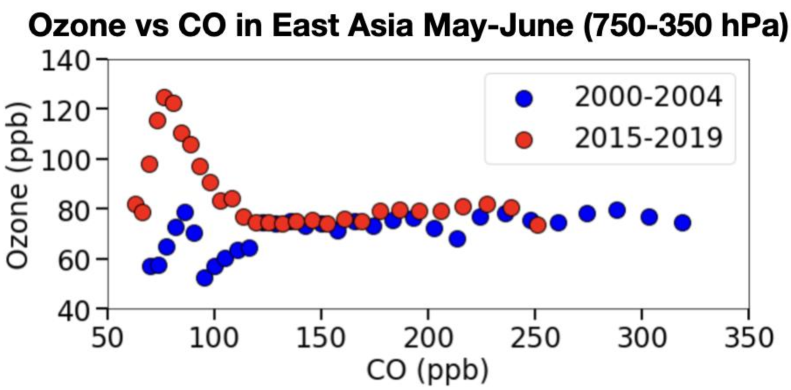

Figure 2 IAGOS ozone versus CO concentrations in the free troposphere (750-350 hPa) over East Asia during May-June 2000-2004 and 2015-2019. Ozone and CO column mixing ratios are from individual vertical profiles averaged over 25 hPa pressure levels. Ozone column mixing ratios are then averaged over bins of 5 ppb CO. Only bins with 50 or more observations are included.

3 GEOS-Chem simulation

We use the GEOS-Chem chemical transport model version 13.2.1 (http://geos-chem.org) to interpret the elevated free tropospheric ozone over East Asia and its trend. GEOS-Chem is driven by MERRA-2-assimilated (Modern-Era Retrospective analysis for Research and Applications) meteorological data with a horizontal resolution of 0.5° × 0.625°, degraded here to 2° × 2.5°, and 72 vertical levels from the surface to 0.1 hPa. Global anthropogenic emissions are from the Community Emissions Data System (CEDS) (Hoesly et al., 2018) and are superseded with the KORUSv5 inventory for East Asia (KORUSv5, http://aisl.konkuk.ac.kr) and the MEIC inventory for China (Zheng et al., 2018). Natural emissions include NO_x from lightning (Murray et al., 2012) and soil (Hudman et al., 2012), MEGANv2 biogenic volatile organic compounds (VOCs) (Guenther et al., 2012), dust (Meng et al., 2021),

https://doi.org/10.5194/egusphere-2025-1799
Preprint. Discussion started: 7 May 2025

and sea salt (Jaeglé et al., 2011). Open-fire emissions are from the Global Fire Emissions Database version 4 (GFED4; van der Werf et al., 2017).

Version 13.2.1 of GEOS-Chem includes a detailed oxidant-aerosol chemical mechanism for the troposphere and stratosphere (Wang et al., 2021). Here we add particulate nitrate photolysis, which was implemented in GEOS-Chem version 14.2.0 following Shah et al. (2023). Particulate nitrate photolysis has been found to be important for reproducing tropospheric ozone observations in the model, offsetting the effects of halogen-catalyzed ozone loss (Lin et al., 2024) and improving the simulation of NO_x in remote air (Shah et al., 2023). Including particulate nitrate photolysis, Colombi et al (2023) found that model was able to reproduce the observed seasonal cycle and spatial distribution of surface ozone in South Korea and China, as well as the observed decadal trend in South Korea. Shah et al (2024) and Lin et al. (2024) found that it enabled an unbiased simulation of tropospheric ozone compared to IAGOS and ozonesonde observations worldwide.

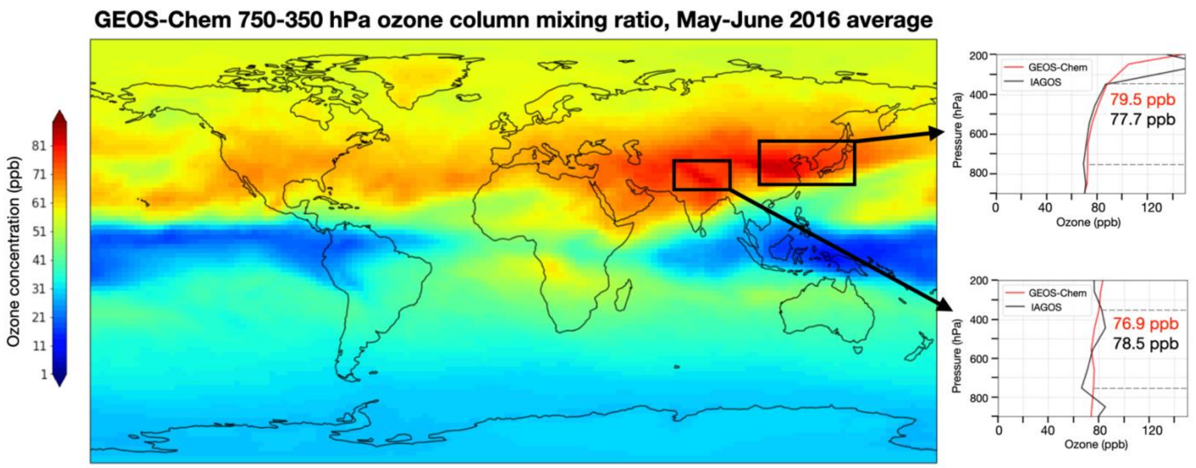

Figure 3 Mean free tropospheric (750-350 hPa) ozone column mixing ratios for May-June 2016 simulated by GEOS-Chem. Ozone vertical profiles over the East Asia and northern India maxima are also shown with comparison to IAGOS aircraft observations. Mean column mixing ratios for 750-350 hPa are shown in legend.

Figure 3 shows the global distribution of mean free tropospheric ozone simulated by GEOS-Chem in May-June 2016. Values are maximum over East Asia and northern India, consistent with IAGOS vertical profiles also shown in the Figure. The high pre-monsoon value over northern India has been reported before as due to high emissions and peak photochemical conditions (Lu et al., 2018), but we find in sensitivity simulations that it does not contribute significantly to the maximum over East Asia. We further analyzed the model maximum over East Asia using observed ozone-CO relationships from the KORUS-AQ aircraft campaign over and around the Korea peninsula up

https://doi.org/10.5194/egusphere-2025-1799
Preprint. Discussion started: 7 May 2025

to 300 hPa in May-June 2016 (Crawford et al., 2021), as shown in Figure 4. The PBL data show a generally positive correlation of ozone with CO as expected from pollution influence, though both model and observations show elevated ozone at the lowest CO concentrations consistent with subsidence from the free troposphere. The free tropospheric observations show a negative correlation of ozone with CO, consistent with Figure 2 and implying stratospheric origin.

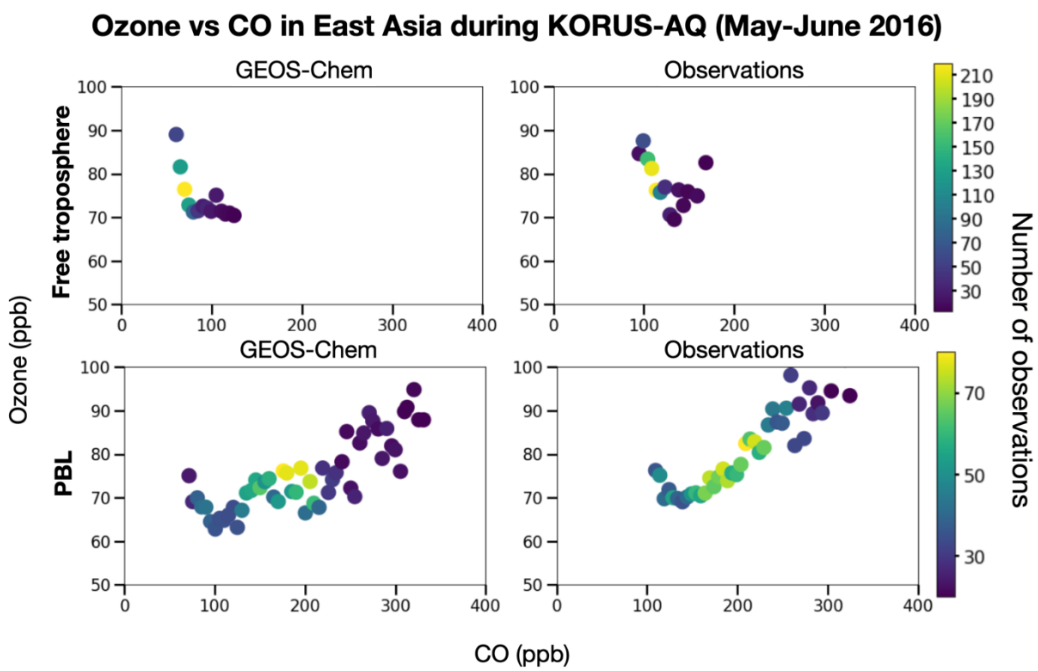

Figure 4 Ozone versus CO mixing ratios in the free troposphere (750-350 hPa) and PBL (surface -750 hPa) over and around the Korea peninsula during the KORUS-AQ aircraft campaign in May-June 2016. 1-minute observations are averaged over 5-ppb CO mixing ratio bins and compared to the GEOS-Chem simulation sampled along the aircraft tracks. Only bins with 20 or more observations are shown.

https://doi.org/10.5194/egusphere-2025-1799
Preprint. Discussion started: 7 May 2025

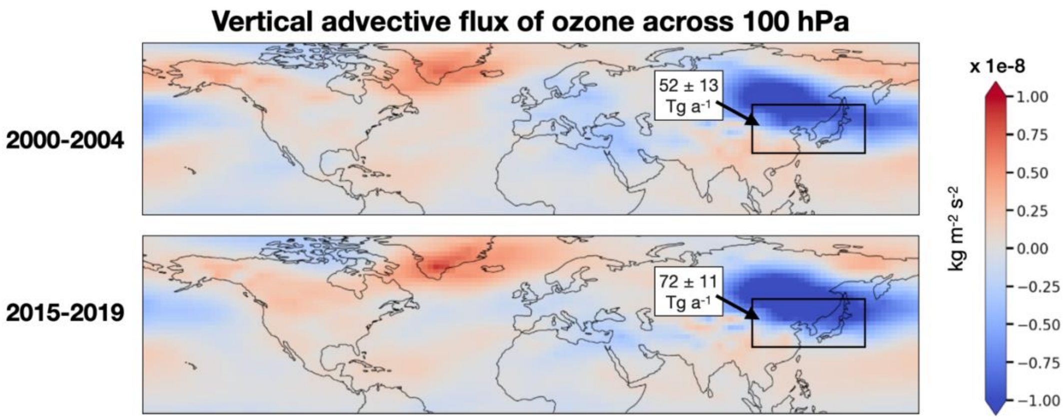

Figure 5 Annual mean vertical advective flux of ozone at 100 hPa, averaged for 2000-2004 and 2015-2019. Negative values (blue) indicate downward motion. Net downwelling rates over East Asia (30.0-45.0 ° N, 109.0-145.0 ° E) are shown inset with interannual standard deviations.

We can diagnose from GEOS-Chem the vertical advective flux of ozone at 100 hPa, a well-established indicator of stratospheric-tropospheric-exchange (Gettleman and Sobel, 2000). Fluxes in May-June are noisy and so we focus on annual means. Global annual mean ozone fluxes across 100 hPa and interannual standard deviations are 589 ± 17 Tg a^{-1} for 2000-2004 and 563 ± 37 Tg a^{-1} for 2015-2019, consistent with the observational estimate of 550 ± 140 Tg a^{-1} (Hu et al., 2017). Figure 5 shows the spatial distribution of the flux in the northern hemisphere, highlighting the intense downwelling over Northeast Asia. For the box in Figure 5 centered over Korea, the flux increases from 52±13 Tg a^{-1} in 2000-2004 to 72±11 Tg a^{-1} in 2015-2019, consistent with the ozone vs. CO relationships in Figure 2.

4 Attribution of the free tropospheric ozone maximum over East Asia

We use the tagged ozone capability in GEOS-Chem (Zhang et al., 2010; Ye et al., 2024) to further diagnose the contributions from the stratosphere and tropospheric source regions to the free tropospheric ozone over East Asia in May-June 2015-2019. The tagged ozone simulation uses archived odd oxygen production rates and loss frequencies from the full-chemistry GEOS-Chem simulation to replicate the ozone simulation in a way that can resolve the contributions from different ozone production regions (Wang et al., 1998). Odd oxygen removes from the accounting the fast reactions cycling ozone with short-lived reservoirs (Bates et al., 2020). Here we conduct the tagged ozone simulation at 0.5°×0.625° resolution over East Asia (10.0-45.0 ° N, 100.0-145.0 ° E), nested within the global 2°×2.5° simulation, to better capture stratospheric influence over the region.

Figure 6 shows the results of this source attribution for the mean May-June vertical ozone profile simulated by GEOS-Chem over East Asia for 2015-2019. We find that ozone produced in the stratosphere makes up 30% of total

https://doi.org/10.5194/egusphere-2025-1799
Preprint. Discussion started: 7 May 2025

ozone at 500 hPa, with a strong vertical gradient opposite to that from tropospheric production. Stratospheric production contributes 10 ppb of ozone in the PBL, which is higher than 2-5 ppb previously found over the United States in GEOS-Chem using the same tagging method (Fiore et al., 2003; Lin et al., 2015). GEOS-Chem tends to underestimate stratospheric influence and events in surface air due to stretched-flow numerical diffusion affecting the downward transport of lamina (Zhang et al., 2014). The stratospheric contribution to the PBL would also be larger if we expanded the odd oxygen family to include hydrogen oxide radicals produced from ozone photolysis as in Bates et al. (2020). GEOS-Chem results from Bates et al. (2020) show that expanding the odd oxygen family in this manner identifies an additional 10 ppb of PBL ozone over East Asia as originating from the stratosphere (SI Figure 2). From the expanded odd oxygen family perspective, much of the free tropospheric production over East Asia in Figure 6 could be of stratospheric origin.

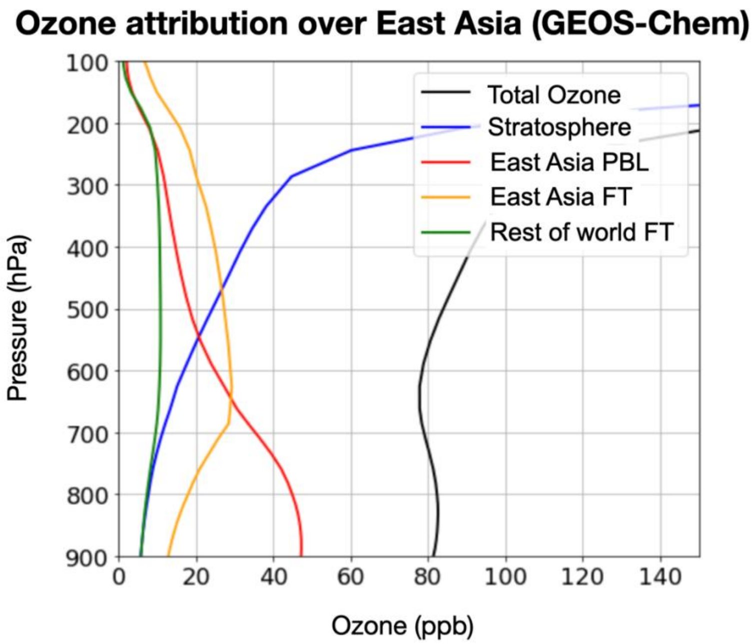

Figure 6 Source attribution of mean ozone over East Asia in May-June 2015-2019 as simulated by GEOS-Chem. Total odd oxygen is decomposed into the contributions from different source regions including the stratosphere, the free troposphere (FT, 750-350 hPa), and PBL (surface-750 hPa). PBL values are daily maximum 8-hour averages (MDA8).

We further investigate the contributions of stratospheric downwelling and emissions to the increase in May-June ozone over East Asia from 2000-2004 to 2015-2019 (Figure 1). Figure 7a shows tagged-ozone simulation results where production and loss frequencies are archived from a 2017 full-chemistry simulation, and meteorology is

https://doi.org/10.5194/egusphere-2025-1799
Preprint. Discussion started: 7 May 2025

simulated for individual years for the 2000-2004 and 2015-2019 periods. The resulting changes in ozone are solely from meteorology. We find no change below 600 hPa but a 5 ppb increase in the free troposphere above 600 hPa that is fully explained by increased stratospheric influence. Figure 7b shows full-chemistry simulation results for meteorological year 2017 but with either 2000 or 2019 emissions. We find an emission-driven increase of ozone of 8 ppb in the PBL decreasing to 2 ppb in the free troposphere. These model increases are consistent with the observed increases in Figure 1 and imply that the PBL increase of ozone is mainly driven by increasing emissions while the free tropospheric increase is mostly driven by increased stratospheric downwelling.

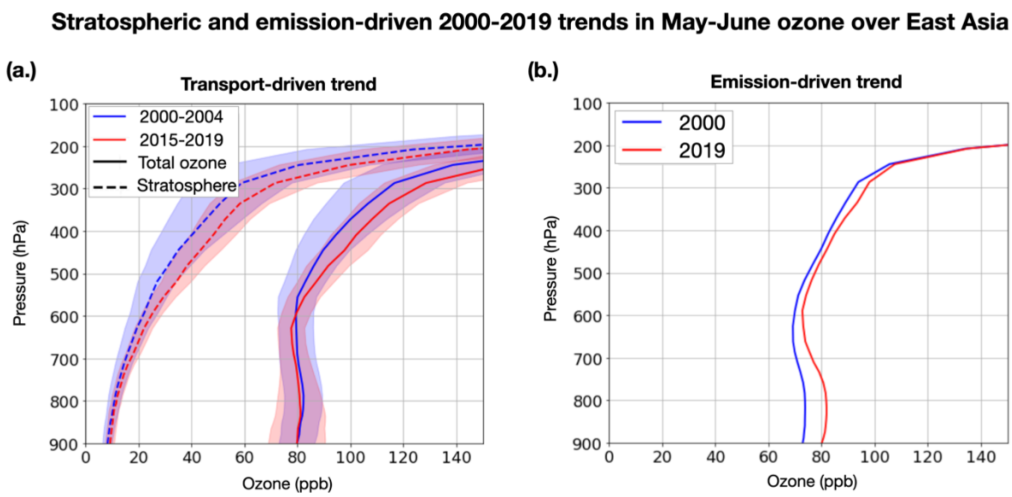

Figure 7 2000-2019 ozone trends simulated by GEOS-Chem over East Asia. Values are averages for May-June (MDA8 in the PBL below 750 hPa) over the domain (30.0-45.0 ° N, 109.0-145.0 ° E). The left panel shows total ozone and stratospherically produced ozone from tagged ozone simulations for meteorological years 2000-2004 and 2015-2019 all with the same ozone production rates and loss frequencies (2017 full-chemistry simulation). Shading indicates interannual standard deviations, The right panel shows full-chemistry simulations with emissions for year 2000 and 2019 and common 2017 meteorology.

Increasing stratospheric influence over East Asia from 2000-2004 to 2015-2019 can be related to strengthening of the jet stream. Figure 8 shows the 100 hPa and 200 hPa wind vectors from MERRA-2 for the two time periods. Jet stream activity is commonly associated with tropopause folds and stratospheric downwelling of ozone (Albers et al., 2018). We find an increase of high-altitude wind speeds over East Asia, reflecting a strengthening of the jet stream. A warming climate is expected to drive a strengthening and poleward-upward shift of the jet stream (Akritidis et al., 2019; Manney and Hegglin, 2018), increasing the frequency of tropopause folding events over East Asia (Ma et al., 2024). It is also expected to amplify zonal-mean temperature gradients and wave drag in the extratropical

https://doi.org/10.5194/egusphere-2025-1799
Preprint. Discussion started: 7 May 2025

stratosphere (Shepherd and McLandress, 2011; Neu et al 2014), increasing the ozone reservoir in the lowermost stratosphere (LMS) which can then be transported downward to the troposphere (Hegglin and Shepherd, 2009). We find evidence from ozondesonde data for increasing LMS ozone over East Asia (SI Figure 3).

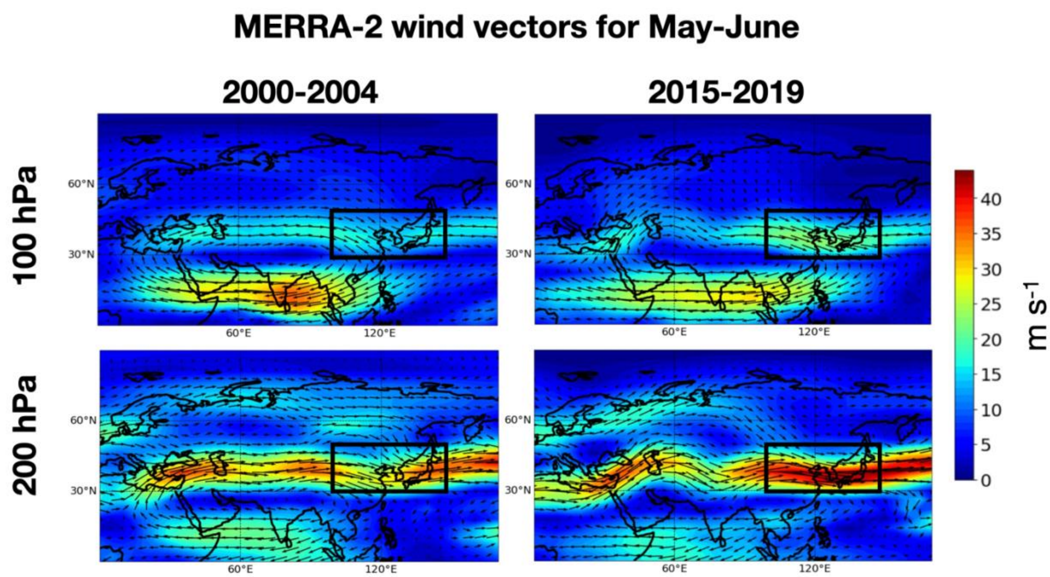

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Figure 8 Mean wind vectors at 100 hPa and 200 hPa for May-June of years 2000-2004 and 2015-2019 in the MERRA-2 assimilated meteorological data. East Asia (30.0-45.0 ° N, 109.0-145.0 ° E) is shown in the black box.

5 Conclusions

We examined the sources of elevated free tropospheric ozone (750-350 hPa) over East Asia in May-June to better understand the origin of this background contributing to severe and increasing surface ozone pollution in South Korea and China.

Aircraft observations and GEOS-Chem model simulations driven by MERRA-2 meteorology show that East Asia and northern India in May-June have the highest free tropospheric ozone concentrations in the world. Mean free tropospheric ozone observed over East Asia in May-June increased from 68±3 ppb in 2000-2004 to 78±4 ppb in 2015-2019, in contrast to Europe and North America where there was no significant trend. Free tropospheric ozone observed over East Asia in 2015-2019 was highest under low CO conditions (< 100 ppb) and this is reproduced by GEOS-Chem. The ozone vertical flux at 100 hPa in GEOS-Chem identifies Northeast Asia as a global hotspot for downwelling of stratospheric ozone, with a 40% increase in the downwelling flux from 2000-2004 to 2015-2019.

https://doi.org/10.5194/egusphere-2025-1799
Preprint. Discussion started: 7 May 2025

We used the GEOS-Chem tagged ozone (odd oxygen) simulation to identify the origin of free tropospheric ozone over East Asia. We find that stratospheric production accounts for 30% of free tropospheric ozone and 12% of planetary boundary layer (PBL) ozone over East Asia during May–June. Stratospheric influence diagnosed by the model would be higher with an expanded definition of odd oxygen to include hydrogen oxide radicals. The observed increase in free tropospheric ozone from 2000-2004 to 2015-2019 is reproduced by GEOS-Chem and is fully explained by the increased stratospheric downwelling, while the PBL ozone trend during the same period is mostly explained by changing anthropogenic emissions. The increased stratospheric downwelling is consistent with intensification of the jet stream over East Asia from 2000–2004 to 2015–2019. Intensification of the jet stream is expected in a warming climate.

We conclude that the unusually high free tropospheric ozone over East Asia in May-June (when surface ozone is maximum) is driven by intense and increasing stratospheric influence. This influence in turn elevates the background in surface air. Because of this elevated natural background, it appears unlikely that South Korea can meet its 60 ppb ozone air quality standard by controlling emissions. However, surface ozone concentrations over South Korea in May-June are frequently over 80 ppb, pointing to leverage for improving ozone air quality by decreasing NO_x and volatile organic compound (VOC) emissions (Oak et al., 2025).

Code Availability. The code used in this work is available upon request.

Data Availability. We use ozone profile data from the In-Service Aircraft for a Global Observing System (IAGOS, https://iagos.aeris-data.fr/).

Author Contribution. The original draft preparation was done by NKC, with review and editing by DJJ, XY, RMY, KHB, DCP, LHY, KL, and HL. DJJ contributed to the project conceptualization. Modeling was done by NKC, with additional support from XY, RMY, KHB, and LHY. The formal analysis was conducted by NKC with additional support from DJJ, XY, RMY, KHB, DCP, LHY, KL, and HL.

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