Foreign influences on tropospheric ozone over East Asia through global atmospheric transport

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Abstract

Tropospheric ozone in East Asia is influenced by the transport of ozone from foreign regions around the world. However, the magnitudes and variations of such influences remain unclear. This study was performed to investigate this influence using a global chemical transport model, GEOS-Chem, through the tagged ozone and emission perturbation simulations. The results show that foreign ozone is transported to East Asia (95 °E-150 °E, 20 °N-60 °N) mainly through the middle and upper troposphere. In East Asia, the influence of foreign ozone increases rapidly with altitude. In the middle and upper troposphere, the regional mean concentrations of foreign ozone range 32-65 ppbv, being 0.8-4.8 times higher than native ozone concentrations of 11-18 ppbv. Over the East Asian tropospheric column, foreign ozone appears most in spring and least in summer when the South Asian High constrains North American, European, and African ozone to north of 35 °N of East Asia. Annually, 40-45% of foreign ozone in the East Asian middle and upper troposphere is from North America (5-13 ppbv) and Europe (5-7 ppbv). At the East Asian surface, the annual mean of foreign ozone concentrations is ~22.2 ppbv,

which is comparable to its native counterpart of ~20.4 ppbv. The annual mean of anthropogenic ozone concentrations from foreign regions is ~4.7 ppbv at the East Asian surface, and half of it comes from North America (1.3 ppbv) and Europe (1.0 ppbv). Foreign ozone concentrations at the East Asian surface are highest in winter (27.1 ppbv) and lowest in summer (16.5 ppbv). This strong seasonality is largely modulated by the East Asian monsoon (EAM) via its influence on vertical motion. The large-scale subsidence prevailing during the East Asian winter monsoon (EAWM) favours the downdraft of foreign ozone to the surface, while widespread convection in the East Asian summer monsoon (EASM) blocks such transport. Interannully, the variation of foreign ozone at the East Asian surface is found to be closely related to the intensity of the EAM. Specifically, the stronger the EAWM is in a winter, the more ozone is from North America and Europe, because of the stronger subsidence behind the East Asian trough. In summer, ozone from South and Southeast Asia is reduced in strong EASM years due to weakened south-westerly monsoon winds. This study suggests substantial foreign influences on tropospheric ozone in East Asia and underscores the importance of the EAM in the seasonal and interannual variations of foreign influences on surface ozone in East Asia.

1 Introduction

Tropospheric ozone is a major pollutant, atmospheric oxidant, and greenhouse gas (Monks et al., 2015). Its sources include photochemical production in the troposphere and downward transport of ozone from the stratosphere (Lelieveld and Dentener, 2000; Gettelman et al., 2011). Having a lifetime of weeks to months in the free troposphere, ozone can be transported across regions and continents, driven by atmospheric circulation (HTAP, 2010). Therefore, tropospheric ozone in a region is affected by both native and foreign emissions and various physical and chemical processes at different temporal and spatial scales (Doherty et al., 2017; Huang et al., 2017; Han et al., 2018). Atmospheric transport makes ozone pollution a globalized issue related to health (Liang et al., 2018), ecosystems (Zhang et al., 2017),

and climate (B. Li et al., 2016). For example, the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) (Galmarini et al., 2017) has been established under the UNECE Convention on Long-Range

50 Transboundary Air Pollution, to improve the understanding of the intercontinental transport of air pollutants across the northern hemisphere. In recent years, East Asia has experienced severe ozone pollution, and surface ozone concentrations have increased (Gaudel et al., 2018; Lu et al., 2018). Through trans-Pacific and trans-Atlantic transport, ozone precursors emitted or ozone produced in East Asia can affect the ozone levels in North America (Verstraeten et al., 2015; Dunker et al., 2017;

55 Nopmongcol et al., 2017) and Europe (Karamchandani et al., 2017; Knowland et al., 2017; Jonson et al., 2018). Therefore, research on ozone outflow from East Asia to the rest of the world has been active, while relatively less attention has been paid to the ozone inflow from foreign regions to East Asia.

Two numerical simulations have been widely used for studying source-receptor relationships (Butler

60 et al., 2018): emission perturbation (sensitivity) (Fiore et al., 2009) and tagging tracer (Wang et al., 1998; Hou et al., 2014; Y. Zhu et al., 2017) simulations. The perturbation simulation examines how ozone within a receptor region responds to a perturbation of ozone precursor emissions in different foreign regions, while the tagged ozone simulation link ozone in each of model grids with its source regions. Concerning air quality, most previous studies focused on foreign influences on the surface-layer in East

65 Asia. These studies showed that the concentrations of ozone from foreign regions are larger at the East Asian surface in colder seasons (November-April) than in warmer seasons (May-October) (Fiore et al., 2009; Nagashima et al., 2010; Wang et al., 2011; Yoshitomi et al., 2011). They also found an uneven distribution of imported ozone from foreign regions at the East Asian surface (Hou et al., 2014; Y. Zhu et al., 2017; Han et al., 2018) and assessed the anthropogenic impacts from individual source regions

70 (Ni et al., 2018). For example, several studies suggested that 1-3 ppbv and ~1 ppbv of surface ozone in East Asia in spring can be respectively attributed to European (Holloway et al., 2008; X. Li et al., 2014)

and South Asian (Chakraborty et al., 2015) anthropogenic emissions. Moreover, the HTAP has made considerable progress in quantifying the contributions of anthropogenic emissions in foreign regions to surface ozone in East Asia (Turnock et al., 2018) and its health impact (Liang et al., 2018).

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Limited studies have suggested that foreign influences are much larger in the higher altitudes than at the surface in East Asia, such as Y. Zhu et al. (2017) and Han et al. (2018) on North American and African ozone, respectively. The contribution of anthropogenic emissions from foreign regions to ozone over China is also larger at high altitudes than at the surface in spring (Ni et al., 2018). However, how

80 imported ozone from foreign regions is distributed in the East Asian middle and upper troposphere, where ozone has larger radiative forcing than at the lower layers (Worden et al., 2008), remains unclear (Liu et al., 2002; Sudo and Akimoto, 2007). The influence in the East Asian middle and upper troposphere is important to climate change because of the considerable ozone radiative forcing over the area (Myhre et al., 2017). Such an impact has been rarely documented (Sudo and Akimoto, 2007). The

85 strongly latitude- and altitude-dependent radiative forcing of tropospheric ozone requires a further examination of the vertical variation of imported ozone in East Asia. Quantifying imported ozone sources at different altitudes also helps in understanding the transport mechanisms. Therefore, the foreign influence on ozone in East Asia throughout the entire tropospheric columns is desirable.

90 Previous studies suggested that the East Asian monsoon (EAM), a predominant climate feature in East Asia, could be a key player in modulating seasonal variation of foreign influences on East Asia (Wang et al., 2011; Chakraborty et al., 2015; B. Zhu et al., 2016; Y. Zhu et al., 2017; Han et al., 2018). Wang et al. (2011) suggested that the seasonal switch in wind patterns of the EAM can bring ozone from different foreign regions to China. Y. Zhu et al. (2017) and Han et al. (2018) demonstrated the

95 importance of the EAM to the vertical transport of imported ozone over East Asia. They found that the

East Asian winter monsoon (EAWM) can boost downdrafts of North American ozone to the East Asian surface (Y. Zhu et al., 2017), while the prevailing convections during the East Asian summer monsoon (EASM) block such transport of African ozone (Han et al., 2018). In addition, imported ozone can significantly drive the interannual variation of ozone over East Asian troposphere (Chatani and Sudo, 2011; Sekiya and Sudo, 2012; Nagashima et al., 2017), while Y. Zhu et al. (2017) reported that interannual variation in North American ozone over the lower troposphere of East Asian is closely related to the variation of the EAWM intensity. Therefore, for an enhanced understanding of the foreign influence on East Asia, the role of the EAM needs to be further investigated.

Since the 2000s, our understanding of the foreign influence on tropospheric ozone in East Asia has been advanced. However, previous studies individually focused on some specific aspects of this influence, such as that from one or a few source regions (X. Li et al., 2014; Y. Zhu et al., 2017; Han et al., 2018), that occurring during one or a few seasons (Ni et al., 2018), or that affecting surface ozone only in East Asia (Wang et al., 2011). The sources of ozone over the entire East Asian troposphere and the underlying transport mechanisms are inadequately documented. The anthropogenic and natural influences have not been separately assessed. The interannual variations of foreign influences, their sensitivity and the associated meteorology have been inadequately studied. Therefore, this study aims to address all of the research gaps and engage in a comprehensive assessment of foreign influences on tropospheric ozone in East Asia.

In this study, we use a global chemical transport model, GEOS-Chem, to quantify foreign influences on tropospheric ozone in East Asia from the perspectives of anthropogenic emissions and all emissions of ozone precursors. We characterize the seasonal, horizontal, and vertical variations of these influences and explore the potential mechanisms. We also search for a link between the interannual variations of

120   foreign influences and the EAM. In the following, section 2 describes the GEOS-Chem model and the

simulation experiments. The seasonal and interannual variations of the foreign influences are presented

in sections 3 and 4, respectively. A summary is provided in section 5.

2 Model description and simulation experiments

125   To clearly identify different sources of tropospheric ozone in East Asia, we define some terms used in

this paper. East Asia is defined as the receptor region (Figure 1), while the regions outside East Asia are

the foreign regions. Tropospheric ozone in this paper refers to ozone in the troposphere, also termed as

ozone (Table S1), unless stated specifically. Tropospheric ozone in East Asia consists of ozone

produced in the troposphere and downward ozone injected from the stratosphere (termed as

130   stratospheric ozone), i.e., tropospheric ozone = ozone produced in the troposphere + stratospheric ozone.

In this paper, we discuss the foreign influence on tropospheric ozone in East Asia from multiple

perspectives (Table S1). (1) The first is by regions inside and outside East Asia, in which the terms

"native ozone" and "foreign ozone" refer to ozone produced in the troposphere inside and outside East

Asia, respectively. "Foreign ozone" generally refers to ozone that is originally generated in a foreign

135   region's troposphere and is imported to the domain of East Asia. The ozone may distribute outside East

Asia, depending on the context. (2) The second is by foreign region, in which ozone produced in the

troposphere over a foreign region is named after that region, such as "North American ozone". Note that

stratospheric ozone injected to a foreign region is counted as stratospheric ozone because it is not

originated in the troposphere. (3) The third is by the sources of ozone precursors, in which the term

140   "anthropogenic ozone" refers to ozone produced from precursors with anthropogenic sources. "Non-

anthropogenic" is the sum of ozone produced in the troposphere from precursors with non-

anthropogenic sources, while "natural ozone" refers to non-anthropogenic ozone and stratospheric

ozone. (4) In the fourth perspective, the components of ozone are termed in more detail by further

divisions, such as by both sources of ozone precursors and foreign regions, as defined in Table S1. For

145 example, North American anthropogenic ozone refers to ozone produced from anthropogenic precursors

emitted from North America. The foreign influences are assessed in terms of the absolute contribution

with a unit of ppbv and the fractional contribution with a unit of percentage (%), which is the ratio of

foreign ozone to ozone in the same domain of interest, unless stated otherwise.

150 A global three-dimensional chemical transport model, GEOS-Chem (version v9-02) (Bey et al., 2001,

http://geos-chem.org), was used to simulate the global tropospheric ozone and the transport of foreign

ozone to East Asia from different source regions. GEOS-Chem includes detailed tropospheric O_3-NO_x-

hydrocarbon and aerosol chemistry. We ran GEOS-Chem simulations two modes in this study: the full

chemistry and tagged ozone modes, corresponding to the emission perturbation and tagged tracer

155 approaches respectively. The emission perturbation simulation quantifies anthropogenic foreign

influences on East Asia, while the tagged ozone simulation specifies overall foreign influences (natural

and anthropogenic), as well as stratospheric influence on East Asia. The simulations were driven by the

GEOS-4 meteorology from the Goddard Earth Observing System (GEOS) at the NASA Global

Modeling and Assimilation Office (GMAO), with 30 reduced vertical layers at the horizontal resolution

160 of 4 °latitude by 5 °longitude. To assess the sensitivity of the simulations to different meteorological

data and spatial resolutions, we also ran GEOS-Chem driven by GEOS-4 at 2 °by 2.5 °, GEOS-5 at 4 °

by 5 °, and GEOS-5 at 2 °by 2.5 °in the full chemistry mode.

Table 1 describes the experiments conducted in this study. We divided the world into eight regions

165 (Figure 1), including East Asia, North America, Europe, Africa, central Asia, South Asia, Southeast Asia,

and the rest of the world. East Asia, North America, South Asia, and Europe are also the study domains

in HTAP Phase 1, where the definitions were slightly different but with similarities. Ten simulations in

full chemistry mode were conducted from January 2004 to February 2006 (2004 for spin-up), including one control experiment (*CTRL*) and nine sensitivity experiments. In the *CTRL* experiment, all

170 anthropogenic and natural emissions were turned on, while in the sensitivity experiments, the anthropogenic emissions including nitrogen oxides (NO_x), carbon monoxide (CO), and non-methane volatile organic compounds (NMVOC) were turned off (100% perturbation) individually in each of the eight defined regions and in the whole world (*EAnth-GLO*). The anthropogenic ozone from a region X_i can be represented by the difference between *CTRL* and the sensitivity experiment for that region

175 (*EAnth-X_i*) (X. Li et al., 2014). Because ozone does not linearly respond to the reduction of its precursors (Fiore et al., 2009), the sum of the ozone response to the 100% perturbation for each region $\sum_{i=1}^{8}$ *(CTRL - EAnth-X_i)* is not equal to the ozone response to the 100% perturbation for the globe (*CTRL - EAnth-GLO*). Specifically in this study, ozone concentration from $\sum_{i=1}^{8}$ *(CTRL - EAnth-X_i)* is approximately 0-4 ppbv (0-20%) higher than that from (*CTRL - EAnth-GLO*) over East Asia depending

180 on altitude (Figure S1). Therefore, to fit the total anthropogenic ozone and isolate the relative contributions of anthropogenic emissions from different regions to the total anthropogenic ozone, a 'normalized marginal' linearization method (B. Li et al., 2016; Ni et al., 2018) was used to adjust the simulations:

185 $$CON\text{-}A = \frac{CTRL \text{ - } EAnth\text{-}A}{\sum_{i=1}^{8} (CTRL \text{ - } EAnth\text{-}X_i)} \times (CTRL \text{ - } EAnth\text{-}GLO) \qquad (1)$$

where *EAnth-A* indicates the sensitivity experiment for *A*. The calculated *CON-A* is the anthropogenic ozone from a specific source region *A* and is used in this study. The calculations were conducted at every model grid. Anthropogenic ozone from a specific source region is named after that region, such as

190 "North American anthropogenic ozone" (Table S1).

Meteorology can modulate foreign ozone over East Asia interannually through its influences on both transport and chemical processes (Liu et al., 2011; Sekiya and Sudo, 2012, 2014). In this study, we focus on its impact on the interannual variation of ozone transport. Therefore, we conducted a tagged ozone simulation from December 1985 to November 2006 (the first year was for spin-up). In the simulation, labelled as Tagged-Ozone (Table 1), daily ozone production and loss data in 2005 were produced from the full chemistry simulation (CTRL) beforehand and then were used in Tagged-Ozone for the years over 1985-2006. In this way, the seasonal variation in chemistry is considered in the simulation while there is no year-to-year variation in chemistry. In the meantime, both seasonal and interannual variations in meteorology are considered in the simulation. Tagged-Ozone included ten tracers, i.e., ozone, ozone from the stratosphere (STR), and ozone produced in the troposphere over East Asia and the seven foreign regions (Figure 1). Ozone produced in the troposphere over a region is named after that region (Table S1). For example, "European ozone" refers to ozone that is originally generated in the European troposphere. In most cases of this paper, "European ozone" is transported to the domain of East Asia. At some places in this paper, "European ozone" can be outside East Asia. Therefore, ozone originally produced in the stratosphere is tagged as a tracer of itself, termed "stratospheric ozone", no matter whether stratospheric ozone is directly transported to the East Asian troposphere or has passed through the troposphere over any of the foreign regions. Note that East Asian ozone is also termed native ozone. The tropopause pressure from the GEOS-4 meteorology was used at each dynamic timestep. The Linoz linearized ozone parameterization scheme (McLinden et al., 2000) was used in the calculation of stratospheric ozone. The calculated global cross-tropopause ozone varies interannually and is ~484 Tg in 2005. The natural and anthropogenic emissions of ozone precursors and the model configuration were the same as those in Han et al. (2018) and are described in detail there.

215   Figure 2 compares the ozone vertical profiles from the simulations with different meteorological data

and resolutions, averaged over East Asia by season. Among the simulations, ozone profiles over East

Asia were similar in shape and magnitude in each of the seasons, except near the surface, where the

simulated ozone concentrations obtained with GEOS-4 data were larger than those obtained with

GEOS-5. The differences between these simulations were smaller in summer and autumn than in winter

220   and spring. A difference within ±5% existed between the tagged ozone and full chemistry simulations

(Figures S2-S3), likely owing to the nonlinearity in chemistry. We further compared the GEOS-Chem

simulations with the ozone retrievals from to the Tropospheric Emission Spectrometer (TES) using the

monthly product TL2O3LN achieved from NASA Langley Atmospheric Science Data Center

(https://eosweb.larc.nasa.gov/project/tes/tes_table). The GEOS-Chem simulations smoothed with TES *a*

225   *priori* and the averaging kernels appeared lower than the TES measurements in the middle troposphere

by approximately 10 ppbv in spring and 5 ppbv in the other seasons (Figure 2). Note that TES

tropospheric ozone retrievals generally have a positive bias compared with ozonesonde measurements

(Nassar et al., 2008; Verstraeten et al., 2013). Verstraeten et al. (2013) identified that the bias is

approximately 2-7 ppbv and is different for the tropics (3 ppbv), sub-tropics (5 ppbv), and mid-latitudes

230   (7 ppbv). Our confidence in the GEOS-Chem performance is also based on extensive validation of

GEOS-Chem simulations of tropospheric ozone in East Asia (Wang et al., 2011; Jiang et al., 2015; J.

Zhu et al., 2017; Y. Zhu et al., 2017), North America (Zhang et al., 2008; Y. Zhu et al., 2017), Europe

(Liu et al., 2005; Kim et al., 2015), Africa (Han et al., 2018), and other regions (Liu et al., 2009; Jiang et

al., 2016).

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3 Seasonal variations of foreign ozone over East Asia

3.1 Native and foreign ozone over East Asia

Based on GEOS-Chem simulations in 2005, we show native and foreign ozone over East Asia at the

surface (Tables 2-3 and Figure 3) and in the East Asian troposphere (Figures 3-5). Figure 3 shows the horizontal variation of the partition of native and foreign ozone at the surface and 3 pressure levels in terms of the annual mean, while Figure 4 shows the vertical variation of the partition of native and foreign ozone over East Asia by season. Figure 5 is the same as Figure 4, but for anthropogenic ozone.

Through the tropospheric column in East Asia, the annual mean of foreign ozone partitions more in the upper layers than at the surface and reach a regional mean of 68% at 500 hPa (Figure 3f). At the surface, the fractional contribution of foreign ozone is lowest over South China, where it is lower than that of native ozone (Figure 3d and 3h). Vertically, foreign ozone constantly increases with altitude in all seasons (Figures 4a-4d). The concentrations of foreign ozone are larger than these of native ozone in spring, autumn, and winter throughout the troposphere, and in summer above ~650 hPa. Fractionally (Figures 4e-4h), foreign ozone accounts the most in the middle or upper troposphere, depending on seasons. In contrast, the fraction of native ozone is the largest near the surface and decreases with altitude. Specifically, between 700 and 200 hPa, the annual mean concentrations of foreign ozone averaged over East Asia range between 14-54 ppbv, which are 0.8-4.8 times higher than native ozone concentrations. Seasonally, the difference between foreign and native ozone between 700 and 200 hPa ranges 17-64 ppbv in spring (or foreign ozone is 1-7.8 times of its counterpart), -3-47 ppbv (-0.1-2.1 times) in summer, 16-53 ppbv in autumn (1-5.3 times), and 26-54 ppbv (2.9-13 times) in winter. In the middle troposphere at 500 hPa, foreign ozone (47.3 ppbv, 72% of ozone) and foreign anthropogenic ozone (9 ppbv, 15%) both peak in spring. In the upper troposphere at 300 hPa, foreign ozone is highest in spring (63.4 ppbv, 63% of ozone) and lowest in winter (51.1 ppbv, 57%).

At the East Asian surface, the annual mean foreign ozone concentrations are ~22.2 ppbv, which are comparable to that of its native counterpart (~20.4 ppbv) (Table 2). Seasonally, foreign ozone is the

largest in winter (27.1 ppbv), the second largest in spring (25.4 ppbv) and the smallest in summer (16.6 ppbv) (Table 2, Figure 4). Foreign ozone accounts for over 50% of ozone at the East Asian surface throughout the year, except in summer. This is in agreement with the estimate of 50-80% foreign contributions in spring made by Nagashima et al. (2010) and J. Li et al. (2016). Foreign anthropogenic ozone at the East Asian surface is 4.7 ppbv in the annual mean and peaks in spring (6.4 ppbv, 14.1% of ozone) when it is comparable to native anthropogenic ozone (6.9 ppbv, 15.1%) (Table 3, Figure 5). Wang et al. (2011) suggested a higher estimate of 12.6±2.3 ppbv in the annual mean foreign anthropogenic ozone in China. Ni et al. (2018) estimated that foreign and native anthropogenic ozone were both approximately 6 ppbv at the surface in China in the spring of 2008. In summer, foreign ozone (16.5 ppbv, 35.2% of ozone) and foreign anthropogenic ozone (3.7 ppbv, 8.2%) at the East Asian surface are both at seasonal minimums, whereas native ozone (30.1 ppbv, 64.1%) and native anthropogenic ozone (9.8 ppbv, 21.8%) both reach the seasonal maximum (Tables 2 and 3, Figures 4 and 5). Turnock et al. (2018) showed that in a future emission scenario with the reductions of 70%, 64%, and 86% in global anthropogenic emissions of CO, NMVOC, and NO_x from 2010 to 2050, foreign anthropogenic ozone at the East Asian surface would decrease by ~3.7 ppbv in the annual mean.

Examining anthropogenic ozone (Figure 5), vertically, foreign anthropogenic ozone is the largest in the upper troposphere in all seasons except in spring when it peaks in the middle troposphere (Figures 5a-5b). In contrast, native anthropogenic ozone is the largest at the surface and decrease with altitude in all seasons. Seasonally, in the middle troposphere at 500 hPa, foreign anthropogenic ozone peaks in spring (9 ppbv or 14% of overall anthropogenic ozone). In the upper troposphere at 300 hPa, foreign anthropogenic ozone is highest in summer (9.5 ppbv, 11%) and lowest in winter (6.3 ppbv, 7%).

The seasonal variation of foreign influences on tropospheric ozone over East Asia is modulated by

multiple factors. Ozone lifetime is one of the important factors, as a longer lifetime can lengthen the transport distance. Here, ozone lifetime in the boundary layer was calculated by the daily average dry deposition and chemical loss rate of ozone at each of the model grids. In the free troposphere, only the chemical loss rate of ozone is used for the calculation. As shown in Figure 6, ozone lifetime at the East Asian surface is longest in winter (11.3 days), shortest in summer (1.1 days), and intermediate in spring (4 days) and autumn (3.8 days). Ozone lifetime at the East Asian surface is approximately 9-28 days shorter than that in the middle troposphere (500 hPa) (Figure 6), due to the dry deposition and more active chemical reactions of ozone in the boundary layer (Fiore et al., 2002; Wang et al., 2011). From the meteorological perspective, subtropical westerlies are the major transport pathway for atmospheric pollutants moving from the west to East Asia (Wild et al., 2004; Y. Zhu et al., 2017; Han et al., 2018). The strength and location of the westerlies vary with season. The East Asian subtropical westerly jet is strongest in winter and weakest in summer (Figure 7, also see Zhang et al., 2006). Therefore, the combined effect of the ozone lifetime and the westerlies is more favourable to the transport of foreign ozone to East Asia in winter and spring than in summer. However, because the inflows to East Asia in winter have low ozone concentrations (Figures 7d and 7h), both foreign ozone and foreign anthropogenic ozone in the East Asian upper troposphere are at minimum in winter (Figures 4 and 5).

Because of the longer lifetime of ozone and the stronger westerly wind in the middle and upper troposphere (Figures 6 and 7), foreign ozone concentrations there are 1-2 times higher than at the surface (Figure 4). Figure 7 clearly shows that foreign ozone is transported to East Asia mostly through the middle and upper troposphere. It also demonstrates that the seasonality of foreign ozone in different tropospheric layers, particularly near the surface, is greatly impacted by vertical transport. In winter, the East Asian trough located around 130-140ºE in the middle troposphere is an important feature of the EAWM system. The downdrafts behind the East Asian trough (100ºE-140ºE) favour the descent of

foreign ozone from upper levels to the surface. Winter has the strongest downdrafts, followed by spring (Figure 7). Oppositely, in summer, the prevailing ascents in the EASM block foreign ozone from reaching the lower troposphere (Figures 7b and 7f, see also Y. Zhu et al., 2017; Han et al., 2018). Combined with the obstruction of the Tibetan Plateau, the blocking effect is obvious in summer (Figures 7b and 7f, see also Han et al., 2018). In addition, the downdrafts behind the European trough (~0-40 °E in Figures 7b and 7f) divert foreign ozone from reaching East Asia in summer (Y. Zhu et al., 2017; Han et al., 2018). Because of all of the above reasons, both foreign anthropogenic ozone and foreign ozone at the surface-layer in East Asia are higher in winter than in summer (Figures 4 and 5). Overall, the EAM is a dominant meteorological system influencing the seasonal variations of foreign ozone at the East Asian surface, primarily through the vertical motion of air masses.

3.2 Foreign ozone over East Asia by source region

3.2.1 Foreign ozone in the East Asian middle and upper troposphere by source region

Figure 8 shows how foreign ozone from different source regions distributes horizontally in the middle troposphere, illustrating significant foreign impacts on ozone over East Asia at this layer. The streamlines in Figure 8 roughly show the transport pathways, demonstrating the importance of the westerlies in driving the ozone transport from North America, Europe, Africa, and central Asia to East Asia. Foreign ozone over East Asia is shown vertically by region in Figure 9. In the East Asian middle and upper troposphere, foreign ozone accounts for 65-85% of all ozone produced in the troposphere (Figure 9). On average, ROW, North America, and Europe contributes 9-21 ppbv (10-19% of ozone), 5-13 ppbv (7-12%), and 5-7 ppbv (3-11%), respectively, to ozone in the East Asian middle and upper troposphere. The sum of ozone from these three regions accounts for ~60% of foreign ozone at these layers. The mean concentrations of North American ozone are higher than those of European ozone over East Asia at layers above 500 hPa, because of larger anthropogenic and natural emissions including

335   biogenic and lightning emissions in North America (Figure S4).

Taking 500 hPa as an example for the middle troposphere, the difference between foreign and native ozone concentrations at this level is highest in winter, when the difference is 37 ppbv, or foreign ozone is 6 times of native ozone (Figures 4 and 9). At the time, of all foreign regions, the ROW is the largest

340   contributor (13.5 ppbv, 31% of foreign ozone), followed by North America (9.1 ppbv, 21%), Africa (7.1 ppbv, 16%), Europe (4.9 ppbv, 11%), and South Asia (4.3 ppbv, 10%). In the upper troposphere at 300 hPa, the concentrations of foreign ozone from the ROW, North America and Africa in winter are 17.3, 10.9, and 8.0 ppbv, respectively, all higher than the concentrations of native ozone (5.6 ppbv).

345   In winter, the transport of African ozone to the East Asian middle and upper troposphere is mainly driven by the Hadley circulation and the subtropical westerlies (Han et al., 2018). Although South Asia and Southeast Asia are closer to East Asia, the atmospheric circulations are less favorable to the ozone transport from South and Southeast Asia to East Asia (Figure S5).

350   Vertically, foreign ozone from five of the source regions, North America, Africa, South Asia, Southeast Asia, and the ROW, increases obviously with altitude in the East Asian troposphere in all seasons (Figure 9). European ozone also increases with altitude in the East Asian troposphere except in winter. Central Asian ozone in the entire East Asian troposphere is lowest in winter and highest in summer when it peaks in the middle troposphere.

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Stratospheric ozone in the East Asian troposphere increases rapidly with altitude, being largest in winter and second largest in spring (Figure 9). For instance, stratospheric ozone in spring rises from 2.1 ppbv (4.3% of ozone) at the surface to 6.4 ppbv (9.6%) at 500 hPa and 27.1 ppbv (26.6%) at 300 hPa

(Figures 9a and 9i). The simulated springtime stratospheric ozone at the East Asian surface in this study is lower than those in some previous studies that used different global chemical transport models or different versions of GEOS-Chem, in which the stratospheric contribution was estimated as 10%-26% by Nagashima et al. (2010), 4-10 ppbv in China by Wang et al. (2011), and 11.2±2.5 ppbv (February-April) in Japan by Yoshitomi et al. (2011). The seasonality of the stratospheric influence in the middle troposphere are similar to those in B. Zhu et al. (2016), who showed that stratospheric ozone at some mountain sites (>1500 m) in China peaks in winter. By comparing GEOS-Chem simulations, MLS satellite observations, and MERRA and MERRA-2 reanalysis data, Jaeglé et al. (2017) suggested that GEOS-Chem underestimates the ozone enhancement from stratospheric intrusions in extratropical cyclones by a factor of 2, corresponding to a systematic underestimate of ozone in the lowermost extratropical stratosphere.

3.2.2 Foreign ozone at the East Asian surface by source region

Tables 2 and 3 show, respectively foreign ozone and foreign anthropogenic ozone at the East Asian surface by source region. On annual average (Table 2), foreign ozone plus stratospheric ozone at the East Asian surface is ~23.5 ppbv (54% of surface ozone). The annual mean ozone concentrations from each of the foreign regions range between 0.9-6.2 ppbv, which account for 2.0-14.2% of surface ozone. The largest contributing region is the ROW, followed by Europe, central Asia, North America, South Asia, Africa, and Southeast Asia. Seasonally, ozone from North America (5.7 ppbv, 14% of surface ozone), Europe (5.3 ppbv, 13%), Africa (2.4 ppbv, 6%), and the ROW (7.5 ppbv, 18%) peaks in winter, whereas ozone from South Asia (2.9 ppbv, 6%) peaks in spring and ozone from Central Asia (4.7 ppbv, 10%) and Southeast Asia (1.3 ppbv, 3%) peak in summer. The seasonal variations of North American, European, and African ozone are similar, i.e., decreasing from winter to summer and then increasing in autumn (Table 2), forming a unimodal distribution. South Asian ozone exhibits a unimodal seasonality,

as well (Chakraborty et al., 2015).

385   By ozone precursor, each of the foreign regions contributes less than 3 ppbv of anthropogenic ozone to the East Asian surface ozone in all seasons (Table 3). On average, the largest contributing regions are North America (27% of foreign anthropogenic ozone), followed by Europe (21%), South Asia (16%), central Asia (14%), and Southeast Asia (12%). Seasonally, a springtime maximum appears for anthropogenic ozone from Europe (1.8 ppbv, 4% of surface ozone), central Asia (1.0 ppbv, 2%), and

390   South Asia (0.9 ppbv, 2%), while anthropogenic ozone from North America is high in both winter (1.8 ppbv, 5%) and spring (1.7 ppbv, 4%). Anthropogenic ozone from Africa and the ROW is the smallest in total, less than 1 ppbv throughout the year. It is noteworthy that ROW ozone generated from anthropogenic precursors contributes little to the East Asian surface ozone (0.8% of surface ozone, Table 3), whereas the contribution of ROW ozone from both anthropogenic and non-anthropogenic

395   precursors is much more (14.2%, Table 2), implying the importance of natural emissions in the ROW to the East Asian surface ozone.

     The foreign anthropogenic influence on the East Asian surface ozone in springtime was previously studied. Holloway et al. (2008) reported that anthropogenic ozone from both North America and Europe

400   ranges between 1-3 ppbv in various regions in East Asia. Fiore et al. (2009) found that if anthropogenic emissions of ozone precursors were reduced by 20% in spring over North America, Europe, and South Asia, surface ozone in East Asia would decrease by 0.3-0.4, 0.2-0.3, and 0.1-0.2 ppbv, respectively. Chakraborty et al. (2015) suggested a decrease of 0.2 ppbv ozone at the East Asian surface in response to a 20% reduction of anthropogenic emissions in South Asia. The results of this study appear

405   comparable with the simulations in Fiore et al. (2009) and Chakraborty et al. (2015). X. Li et al. (2014) estimated that European and South Asian anthropogenic emissions can each contribute 2.4 and 1 ppbv

to surface ozone in western China. Ni et al. (2018) suggested that Europe and South Asia contributed 1.6 and 1.4 ppbv, respectively, to surface ozone in China in the spring of 2008. Wild et al. (2004) demonstrated that North American and European anthropogenic ozone in spring each range from 1.5 to 2.5 ppbv at the surface over Japan. Using the same model but with different emission data from those in Wild et al. (2004), Yoshitomi et al. (2011) simulated that North American and European anthropogenic ozone contribute 2.8±0.5 and 3.5±1.1 ppbv, respectively, to surface ozone in Japan during February-April. Overall, simulations from this study in spring are at the same magnitude as those in previous studies, and are slightly smaller than those in Yoshitomi et al. (2011) probably resulting from differences in the emission inventories and numerical models.

3.2.3 Latitudinal and longitudinal variations of foreign ozone in the East Asian troposphere by source region

Foreign ozone over East Asia vary largely with latitude (Figures 3 and 8), as illustrated in Figure 10 for different source regions. In nearly all the tropospheric layers (Figures 10a-10d), South and Southeast Asian ozone distributes mostly in the regions south of 35 °N in East Asia. In contrast, ozone from North America, Europe, and central Asia mainly appears north of 35 °N in East Asia. The fractional contributions of foreign ozone from South Asia, Southeast Asia, and Africa all decrease with latitude, whereas the fractional contributions of foreign ozone from North America, Europe, and central Asia all increase with latitude. The latitudinal variations of North American and European ozone are consistent with those in Hou et al. (2014) and Y. Zhu et al. (2017).

At the East Asian surface (Figure 10e-10h), the fractional contribution of foreign ozone peaks at the northern border of East Asia (60 °N) in the four seasons, ranging from 55% in summer to 85% in winter. In spring, native ozone at the East Asian surface is largest at approximately 30 °N (36.8 ppbv, 65.8% of

ozone). The latitude at which native ozone peaks shifts northward to 35 °N in summer (50 ppbv, 88% of ozone) and then southward to 25 °N in autumn (31 ppbv, 74%) and farther southward to 20 °N in winter (28 ppbv, 61%). This seasonal migration may be partially related to the variation of ozone production influenced by the EAM (Hou et al., 2015; S. Li et al., 2018). The concentrations of stratospheric ozone at the East Asian surface are largest between 42 °N and 46 °N in spring (2.4 ppbv) and winter (2.7 ppbv).

The latitudinal variations of foreign ozone from different source regions are likely due to the proximity of these foreign regions to East Asia, the topography in East Asia, and the meteorology along the transport pathways. In particular, in summer, atmospheric circulations in the upper troposphere over Eurasia are greatly influenced by the South Asian High (SAH, or Tibetan High, or Asian summer monsoon anticyclone), and its position and coverage are shown by the streamlines in Figure 11. SAH constrains foreign ozone from North America, Europe, Africa and the ROW to latitudes north of 35 °N in the East Asian upper troposphere (Figure 11). The SAH also blocks the northward transport of Southeast Asian ozone to East Asia. Furthermore, the SAH facilitates the build-up of South Asian ozone in the East Asian upper troposphere (Figure 11f) (Vogel et al., 2015), being a reason for the summer maximum of South Asian ozone over the region (Figure 9b).

The longitudinal variations of foreign ozone from different source regions over East Asia are shown in Figure S6. In the East Asian middle and upper troposphere, the variations with longitude are less obvious than those with latitude, especially in winter (Figure 10 vs. Figure S6). In the East Asian middle troposphere (500 hPa), central Asian and South Asian ozone decrease with longitude in summer but vary insignificantly with longitude in winter. At the East Asian surface, ozone from the two regions decreases with longitude. Longitudinal variations of North American, European, African, and Southeast Asian ozone are less obvious than those of central and South Asian ozone.

4 Interannual variations of foreign ozone at the East Asian surface

The Tagged-Ozone simulation (Table 1) was used to search for possible connections of the interannual variations between meteorology and foreign ozone in East Asia. The Tagged-Ozone simulation provided the means and year-to-year variations of native and foreign ozone from the different regions during the 20-year period (not shown). The mean native and foreign ozone at the East Asian surface were in close agreement with those in 2005 (Table 2) in the corresponding regions and seasons.

As a typical monsoon region, the climate in East Asia is largely influenced by the EAM. The monsoon circulation can impact ozone transport and distribution in East Asia (Y. Zhu et al., 2017; S. Li et al., 2018). To search for possible linkages between the EAM and the transport of foreign ozone to East Asia, we selected three EAWM indices and three EASM indices, respectively, for winter and summer. These monsoon indices were proposed to describe the features of the EAM from different perspectives (Q. Li et al., 2016; S. Li et al., 2018). The monsoon indices are each correlated with different types of foreign ozone at the East Asian surface. The linkages between the EAM and foreign ozone at the East Asian surface were assessed according to the mean of the correlation coefficients from the three indices in a season.

These six monsoon indices are widely used. The three EAWM indices were proposed by Sun and Li (1997), Jhun and Lee (2004), and Wang and Jiang (2004), respectively, corresponding to Equations (2)-(4). EAWMI1, defined by Sun and Li (1997), represents the EAWM strength by the averaged geopotential heights in the middle troposphere in the location of the East Asian trough, which is an important component of the EAWM. EAWMI2 (Jhun and Lee, 2004) reflects the meridional wind shear associated with the jet stream in the upper troposphere, mainly describing the variability of the EAWM

in the East Asian mid-latitudes. EAWMI3 (Wang and Jiang, 2004) uses the anomaly of the wind

480 velocity around the coast of East Asia in the lower troposphere. The three EASM indices were proposed

by Wang and Fan (1999), Li and Zeng (2002), and Zhang et al. (2003), respectively, corresponding to

Equations (5)-(8). EASMI1 (Wang and Fan, 1999) is defined from the shear vorticity in the lower

troposphere that reflects variations in both the monsoon trough and the subtropical high (Wang et al.,

2008). EASMI2 (Li and Zeng, 2002) is a unified dynamical index of the monsoon which characterizes

485 the seasonal and interannual variability of monsoons over different areas in the world. EASMI3 (Zhang

et al., 2003) is a vorticity index similar to that in Wang and Fan (1999) but in a slightly modified

domain.

$$EAWMI1 = -GPH_{500}(30\text{-}45\ {}^{\circ}N,\ 125\text{-}145\ {}^{\circ}E) \tag{2}$$

490
$$EAWMI2 = U_{300}(27.5\text{-}37.5\ {}^{\circ}N,\ 110\text{-}170\ {}^{\circ}E) - U_{300}(50\text{-}60\ {}^{\circ}N,\ 80\text{-}140\ {}^{\circ}E) \tag{3}$$

$$EAWMI3 = WS_{850}(25\text{-}50\ {}^{\circ}N,\ 115\text{-}145\ {}^{\circ}E) \tag{4}$$

$$EASMI1 = U_{850}(5\text{-}15\ {}^{\circ}N,\ 90\text{-}130\ {}^{\circ}E) - U_{850}(22.5\text{-}32.5\ {}^{\circ}N,\ 110\text{-}140\ {}^{\circ}E) \tag{5}$$

$$EASMI2 = \delta(10\text{-}40\ {}^{\circ}N,\ 110\text{-}140\ {}^{\circ}E,\ 850\ hPa) \tag{6}$$

$$\delta = \frac{\|\bar{V}_1 - V_i\|}{\|\bar{V}\|} - 2 \tag{7}$$

495
$$EASMI3 = U'_{850}(10\text{-}20\ {}^{\circ}N,\ 100\text{-}150\ {}^{\circ}E) - U'_{850}(25\text{-}35\ {}^{\circ}N,\ 100\text{-}150\ {}^{\circ}E) \tag{8}$$

In Equations (2)-(6), GPH_{500} is the geopotential height at 500 hPa, U_{300} is the zonal wind at 300 hPa,

WS_{850} is the wind speed at 850 hPa, U_{850} is the zonal wind at 850 hPa, and δ is a dynamical normalized

seasonality obtained from Equation (7). In Equation (7), $\bar{V}_1$ and V_i are the January climatological and

500 monthly wind vectors at a grid and $\bar{V}$ is the mean of the January and July climatological wind vectors at

the same grid. The norm $|| V ||$ is defined as $(\iint_s |V|^2 dS)^{1/2}$, where S denotes the domain of integration. U'_{850} in Equation (8) is the anomaly of U_{850}.

Figure 12 shows the interannual variations of foreign and native ozone at the East Asian surface driven by meteorology and the strength of the EAM. In winter (Figure 12a), the transport of both North American and European ozone is significantly related to the strength of the EAWM. The EAWM can explain more than 30% of the interannual variations of ozone at the East Asian surface from these two regions. A positive correlation between North American ozone and the EAWM was also found by Y. Zhu et al. (2017), who suggested that the increase of ozone transport under a strong EAWM condition is mainly caused by the enhanced downdraft from the Siberian High and the East Asian trough. This explanation can also be applied to European ozone. In contrast, native ozone is negatively correlated with the interannual variation of the EAWM strength. When the EAWM is strong in winter, the enhanced Siberian High strengthens the northerly wind in the East Asian lower troposphere, and, consequently, more native ozone is taken away (Q. Li et al., 2016). Among the three EAWM indices, foreign ozone from North America and Europe at the East Asian surface correlates the best with EAWM1 (Table S2), indicating the importance of the East Asian trough for the ozone transport. When the East Asian trough is deeper in a stronger EAWM, the enhanced downdrafts behind the trough can transport more foreign ozone from the upper levels to the East Asian surface.

In summer (Figure 12b), the interannual variations of the EASM strength were found to be positively correlated with those of ozone and native ozone but negatively correlated with those of South Asian and Southeast Asian ozone at the East Asian surface. The positive correlation between the EASM and surface ozone in East Asia was also reported in previous studies (Zhou et al., 2013; Yang et al., 2014; Hou et al., 2015; S. Li et al., 2018). Among the three EASM indices, the correlations coefficients are

525   higher for EASM1 and EASM3 than EASM2 (Table S2), indicating that these ozone anomalies

correlate closely with shear vorticity during the EASM. In a strong EASM year, a clear anomalous

cyclonic circulation appears over the area southeast of China in the lower troposphere, and the south-

westerly monsoon wind is weakened, as depicted by multiple studies (for example, Figure 5 in Yang et

al. (2014) and Figure 2 in S. Li et al. (2018)). The weakened south-westerly monsoon wind during a

530   strong EASM year enhances ozone and native ozone at the East Asian surface by reducing ozone export

(Yang et al., 2014). Meanwhile, the south-westerly wind brings less South Asian and Southeast Asian

ozone to the East Asian surface. The variation of the EASM intensity can approximately explain 32%,

31%, and 64% of the interannual variability in native, South Asian, and Southeast Asian ozone,

respectively.

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5 Discussion and conclusions

Using a global chemical transport model, GEOS-Chem, we investigated foreign influences on

tropospheric ozone over East Asia. We estimated these influences from the perspectives of the

anthropogenic and total emissions of ozone precursors, using the emission perturbation and tagged

540   ozone simulations, respectively. The distributions of foreign ozone in East Asia were characterized in

time (seasonally) and space (horizontally and vertically). Based on six EAM indices, links between the

EAM and interannual variations of foreign ozone at the East Asian surface were explored. Conclusions

were drawn as follows.

545   Foreign ozone is transported to East Asia mainly through the middle and upper troposphere because

of the longer lifetime of ozone and the stronger westerlies at these layers than in the lower troposphere.

In the East Asian middle and upper troposphere (700-200 hPa), the annual mean concentrations of

foreign ozone are 0.8-4.8 times higher than that of native ozone, as foreign ozone ranging in 32-65 ppbv

(65-85% of ozone produced in the troposphere) and native ozone ranging in 11-18 ppbv (Figures 4 and 9).

At the East Asian surface, the annual mean of native ozone (20.4 ppbv) is comparable to that of foreign ozone (22.2 ppbv) (Table 2), most of which is transported from North America and Europe. Regarding anthropogenic ozone only, the annual mean of foreign anthropogenic ozone (4.7 ppbv, 43% of total anthropogenic ozone) is slightly lower than its native counterpart (6.3 ppbv, 57%) (Table 3). Three-quarters of foreign anthropogenic ozone at the surface is from North America (27%), Europe (22%), South Asia (16%), and Southeast Asia (12%). Foreign ozone at the East Asian surface is greatly modulated by the downward transport in East Asia, and thus, its seasonality is dominated by the EAM system (Figure 7). The subsidence prevailing in the EAWM favours the downward transport of foreign ozone to the surface, while ascending flows in the EASM block such transport. Therefore, foreign ozone at the East Asian surface is highest in winter (27.1 ppbv, 66% of surface ozone) and lowest in summer (16.5 ppbv, 35%) (Table 2).

In the East Asian troposphere, foreign ozone from North America, Europe, and central Asia generally increases with latitude from 20 °N to 60 °N, whereas foreign ozone from South Asia and Southeast Asia decreases with latitude (Figures 8 and 10). In the upper troposphere, the SAH in summer blocks North America, European, and African ozone from transport to latitudes south of 35 °N in East Asia (Figure 11).

The interannual variations of foreign ozone at the East Asian surface is found to be closely related to the EAM strength (Figure 12). In winter, when the EAWM is strong in winter, more North American and European ozone tends to be transported to the East Asian surface because of the heavier downdrafts

behind the East Asian trough. Meanwhile, the strengthened north-westerly and north-easterly monsoon winds can enhance outflow of native ozone and thus reduce its concentrations in East Asia. In summer, if the EASM is strong, the weakened south-westerly monsoon wind enhances native ozone in East Asia by subsiding outflow of native ozone. In the meantime, the weakened south-westerlies reduce the transport of South Asian and Southeast Asian ozone to the East Asian surface.

This study revealed the significant foreign influences on tropospheric ozone over East Asia through global atmospheric transport. We provided a comprehensive assessment on this topic, which cover all seasons, all foreign regions, and all tropospheric layers in East Asia. We discussed this issue from multiple perspectives: by native and foreign ozone, by foreign region, by ozone precursor, and further by both source of ozone precursors and foreign region. In comparison, previous studies most focused on the influences from one or a few foreign regions (X. Li et al., 2014; Chakraborty et al., 2015; Zhu et al., 2017; Han et al., 2018), or during one or a few seasons (Ni et al., 2018), or on the foreign influence at the East Asian surface or boundary layer (Holloway et al., 2008; Wang et al., 2011; Hou et al., 2014). At the East Asia surface, this study compares reasonable with most of the previous studies, although there are some disagreements in various details.

We examined the foreign influence on ozone in East Asia throughout the entire tropospheric column. The simulations show that the concentrations of foreign ozone increase remarkably with altitude and is much higher than its native counterpart in the middle and upper troposphere. The influence in the East Asian middle and upper troposphere is important to climate change because of the considerable ozone radiative forcing in these altitudes (Myhre et al., 2017). Such an impact has been rarely documented (Sudo and Akimoto, 2007).

We found the influences of the EAM on the seasonal and interannual variations of foreign ozone distribution in East Asia, primarily through the vertical transport. Advancing from Zhu et al. (2017) for North American ozone, we found strong correlations between the strength of the EAM and the

600 interannual variation of foreign ozone from various regions including Europe, South Asia, and Southeast Asia. These findings highlight the importance of meteorology in the receptor region of East Asia, offering a new insight on the issue of foreign influence on tropospheric ozone in East Asia.

This study is subject to some uncertainties raising from the following sources. Firstly, GEOS-Chem,

605 as a numerical model, can be biased in simulating various processes in the atmosphere, which are common to all numerical models. For example, some studies suggested that the stratospheric influence may be underestimated in GEOS-Chem (Jaeglé et al., 2017). If this is the case, the absolute foreign influences assessed in this study would remain the same but the fractional contributions of foreign ozone would be overestimated. Secondly, we ran GEOS-Chem simulations with the GEOS-4

610 meteorology. Although GEOS-Chem driven by GEOS-4 performs strongly in simulating tropospheric ozone (Choi et al., 2017), the comparison between the GEOS-Chem and TES ozone data still indicates some biases in the simulation, especially in spring and winter (Figure 2).Thirdly, the anthropogenic emissions in 2005 were used in the simulation, which were scaled from the most recent inventories, such as the global inventory EDGAR v3.2 in 2000 (Olivier and Berdowski, 2001; Pulles et al., 2007)

615 and the INTEX-B Asia emissions in 2006 (Zhang et al., 2009). However, the anthropogenic emissions have significantly changed globally in the last decade (Jiang et al., 2018), especially in East Asia (Zheng et al., 2018). In the EDGAR emission inventories, global NO_x, CO, and NMVOCs from 2000 to 2010 changed by 9.5%, -1.2%, and 5.2%, respectively and the three species decreased across North America and Europe, while increased in East Asia (Turnock et al., 2018). Moreover, there have been some

620 changes in the global natural emissions, such as in biogenic NMVOC emissions (Chen et al., 2018).

Note that a change in anthropogenic emissions will affect the estimate of foreign anthropogenic ozone, which is approximately 20% of the total foreign ozone in East Asia (Figure 5 vs. Figure 4, Table 3 vs. Table 2). , Fourthly, methane from anthropogenic emissions were not addressed in this version of GEOS-Chem. Finally, ozone production and loss data generated from the full chemistry simulation for the tagged ozone simulation are archived at daily time step so that the chemical processes at sub-daily time step are not considered in the tagged ozone simulation. We assessed these uncertainties in Figures 2, S2 and S3. The results suggest that the main conclusions from this study are robust, including the orders of the magnitudes of foreign ozone in the East Asian troposphere, the variations of foreign ozone with time and space, and the importance of the EAM in modulating the variations.

Regarding the meteorological impacts on the interannual variation of foreign ozone in East Asia, this study underscores the importance of the EAM, representing an advancement from Y. Zhu et al. (2017) and Han et al. (2018). Future studies can examine the influences of other prominent climate systems, for instance, the North Atlantic Oscillation (NAO) (Bacer et al., 2016) and the El Niño Southern Oscillation (ENSO) (Sekiya and Sudo, 2012, 2014; Hou et al., 2016).

Data availability

The GEOS-Chem model is available at http://acmg.seas.harvard.edu/geos/. The TES ozone data can be downloaded from https://eosweb.larc.nasa.gov/project/tes/tes_table.

Author contributions

Han and Liu designed and conducted the simulations, analysed the results, and wrote the manuscript. Yuan, Wang, Zhuang, and Zhang contributed to the data analysis and result interpretation. Liu conceived the research problems and supervised the study.

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Competing interests

The authors declare that they have no conflict of interest.

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940   Table 1. GEOS-Chem simulations and experiments in this study[1].

Simulation Type	Period	Experiment	Description
Full chemistry simulation	2005 (2004 for spin-up)	1. CTRL	Including all emissions
		2. EAnth-GLO	Excluding global anthropogenic emissions
		3. EAnth-EAS	Excluding anthropogenic emissions in EAS
		4. EAnth-NAM	Excluding anthropogenic emissions in NAM
		5. EAnth-EUR	Excluding anthropogenic emissions in EUR
		6. EAnth-AFR	Excluding anthropogenic emissions in AFR
		7. EAnth-CAS	Excluding anthropogenic emissions in CAS
		8. EAnth-SAS	Excluding anthropogenic emissions in SAS
		9. EAnth-SEAS	Excluding anthropogenic emissions in SEAS
		10. EAnth-ROW	Excluding anthropogenic emissions in ROW
Tagged ozone simulation	1987-2006 (1986 for spin-up)	11. Tagged-Ozone	Tagging ozone produced in the troposphere over different regions[2]

[1]The abbreviations stand for different regions, including East Asia (EAS), North America (NAM), Europe (EUR), Africa (AFR), central Asia (CAS), South Asia (SAS), Southeast Asia (SEAS), and the rest of the world (ROW) (see Figure 1). "CTRL" represents "a controlled run". "EAnth" means "Excluding anthropogenic emissions".

945   [2]The daily ozone production and loss data in 2005 was generated in experiment 1 (CTRL) and archived for the tagged ozone simulation. Therefore, the tagged ozone simulation includes the seasonal variation in chemistry but without interannual variation in chemistry.

Table 2. The contributions of foreign ozone from different regions to surface ozone in East Asia in 2005. The fractional contributions are indicated in brackets, taking surface ozone in East Asia as 100%[1].

		Annual	Spring	Summer	Autumn	Winter
Native ozone		20.4 (46.4%)	20.7 (43.1%)	30.1 (64.2%)	19.2 (49.1%)	11.5 (28.1%)
Foreign ozone	Total	22.2 (50.5%)	25.4 (52.9%)	16.5 (35.3%)	19.7 (50.4%)	27.1 (66.4%)
	NAM	3.4 (7.8%)	3.7 (7.7%)	1.2 (2.6%)	3.3 (8.4%)	5.7 (14.0%)
	EUR	4.3 (9.7%)	5.1 (10.6%)	2.3 (4.9%)	4.2 (10.8%)	5.3 (13.0%)
	AFR	1.2 (2.8%)	1.2 (2.6%)	0.3 (0.7%)	0.8 (2.1%)	2.4 (5.9%)
	CAS	4.0 (9.1%)	4.1 (8.5%)	4.7 (10.1%)	3.8 (9.8%)	3.3 (8.0%)
	SAS	2.1 (4.8%)	2.9 (6.1%)	2.0 (4.2%)	1.6 (4.0%)	2.2 (5.5%)
	SEAS	0.9 (2.0%)	1.0 (2.1%)	1.3 (2.8%)	0.7 (1.7%)	0.7 (1.7%)
	ROW	6.2 (14.2%)	7.4 (15.4%)	4.7 (9.9%)	5.3 (13.7%)	7.5 (18.3%)
Stratospheric ozone		1.3 (3.0%)	2.1 (4.3%)	0.3 (0.7%)	0.4 (1.0%)	2.5 (6.2%)
Surface ozone		43.9 (100%)	48.0 (100%)	46.9 (100%)	39.1 (100%)	40.8 (100%)

950   [1]All values are the means over East Asia. The abbreviations stand for different regions (Figure 1), including East Asia (EAS), North America (NAM), Europe (EUR), Africa (AFR), central Asia (CAS), South Asia (SAS), Southeast Asia (SEAS), and the rest of the world (ROW).

Table 3. The contributions of anthropogenic ozone (in ppbv) from different regions to surface ozone in East Asia in 2005. The fractional contributions are indicated in brackets, taking surface ozone in East Asia as 100%[1].

		Annual	Spring	Summer	Autumn	Winter
Native anthropogenic ozone		6.3 (15.2%)	6.9 (15.1%)	9.8 (21.8%)	6.2 (16.6%)	2.2 (5.9%)
Foreign anthropogenic ozone	Total	4.7 (11.3%)	6.4 (14.1%)	3.7 (8.2%)	4.1 (10.8%)	4.6 (12.1%)
	NAM	1.3 (3.1%)	1.7 (3.7%)	0.6 (1.4%)	1.1 (2.9%)	1.8 (4.6%)
	EUR	1.0 (2.4%)	1.8 (3.9%)	0.7 (1.6%)	1.0 (2.6%)	0.6 (1.6%)
	AFR	0.2 (0.4%)	0.2 (0.4%)	0.2 (0.4%)	0.1 (0.4%)	0.2 (0.5%)
	CAS	0.6 (1.5%)	1.0 (2.2%)	0.5 (1.2%)	0.7 (1.8%)	0.3 (0.9%)
	SAS	0.7 (1.8%)	0.9 (2.0%)	0.7 (1.6%)	0.5 (1.4%)	0.8 (2.1%)
	SEAS	0.5 (1.3%)	0.5 (1.1%)	0.7 (1.6%)	0.5 (1.2%)	0.5 (1.3%)
	ROW	0.3 (0.8%)	0.4 (0.8%)	0.2 (0.5%)	0.2 (0.6%)	0.4 (1.2%)
Natural ozone		30.5 (73.6%)	32.0 (70.8%)	31.4 (70.0%)	27.2 (72.5%)	31.2 (82.0%)
Surface ozone		41.5 (100%)	45.4 (100%)	44.9 (100%)	37.5 (100%)	38.1 (100%)

[1]All values are the means over East Asia. The abbreviations stand for different regions (Figure 1), including East Asia (EAS), North America (NAM), Europe (EUR), Africa (AFR), central Asia (CAS), South Asia (SAS), Southeast Asia (SEAS), and the rest of the world (ROW).

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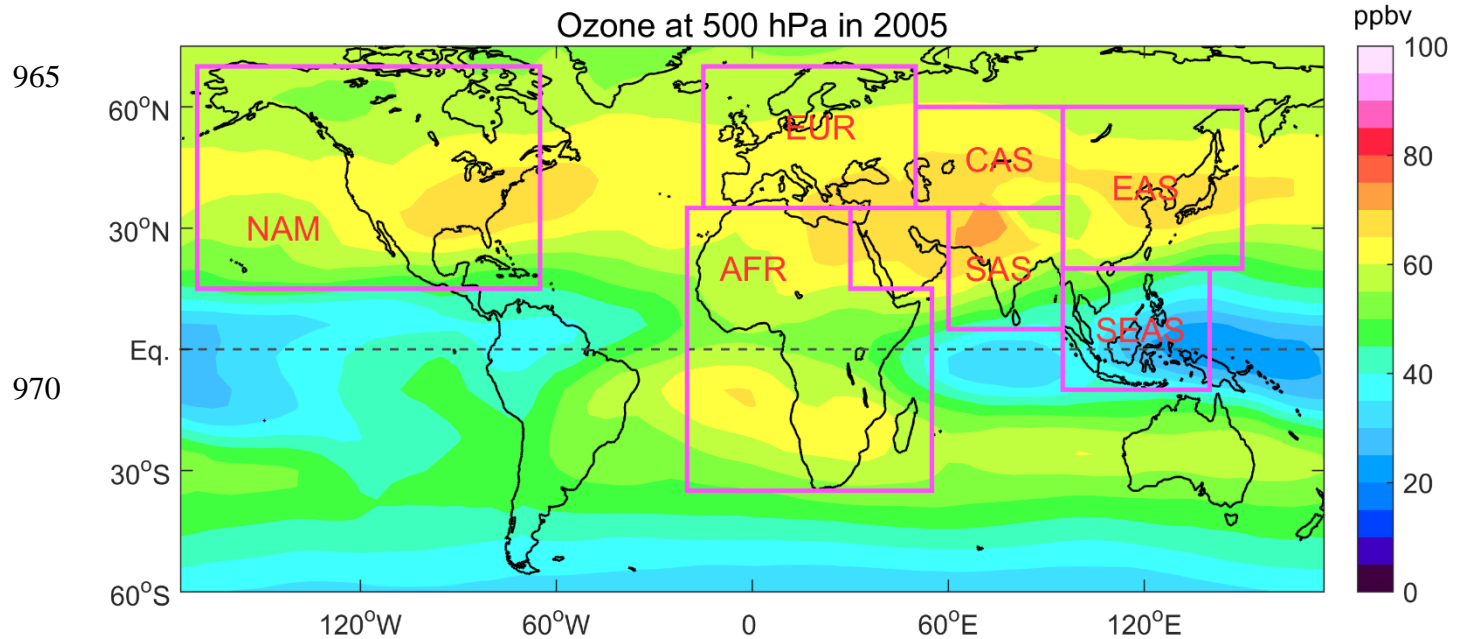
975 Figure 1. Annual mean ozone concentrations in the middle troposphere (500 hPa) in 2005 from GEOS-

Chem simulations. The purple boxed areas define seven regions, plus the rest of the world (ROW, all

regions except the boxed regions). The seven regions include East Asia (EAS, 95 ̊E-150 ̊E, 20 ̊N-60 ̊N),

North America (NAM, 170 ̊W-65 ̊W, 15 ̊N-70 ̊N), Europe (EUR, 15 ̊W-50 ̊E, 35 ̊N-70 ̊N), Africa

(AFR, 20 ̊W-30 ̊E, 0-35 ̊N and 20 ̊W-55 ̊E, 35 ̊S-0), central Asia (CAS, 50 ̊E-95 ̊E, 35 ̊N-60 ̊N),

980 South Asia (SAS, 60 ̊E-95 ̊E, 5 ̊N-35 ̊N), and Southeast Asia (SEAS, 95 ̊E-140 ̊E, 10 ̊S-20 ̊N).
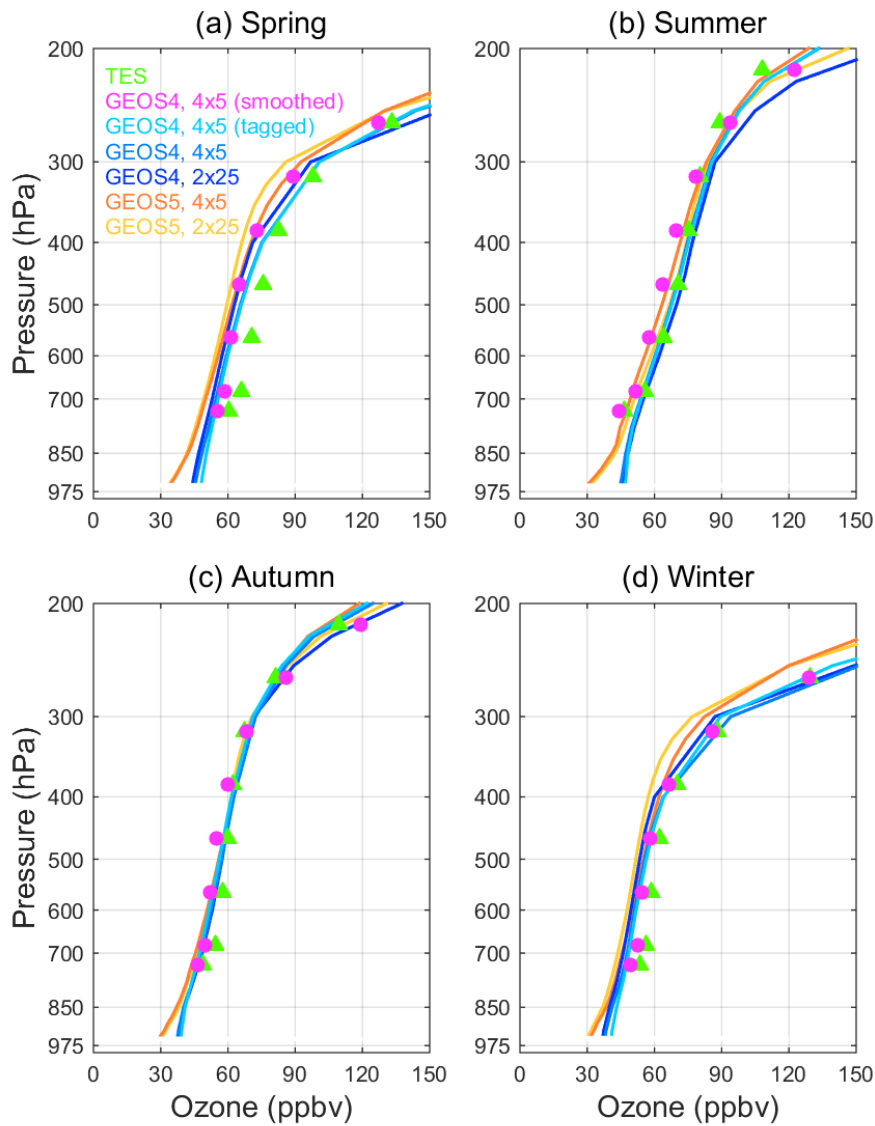
Figure 2. Vertical ozone profiles averaged over East Asia in 2005 from GEOS-Chem simulations driven by GEOS-4 and GEOS-5 meteorological data with different horizontal resolutions (4 °by 5 °and 2 °by 2.5 °in latitude and longitude, indicated in different colours). TES retrievals (in green) and GEOS-Chem simulations smoothed with the TES *a priori* and averaging kernels (in purple) are also shown.

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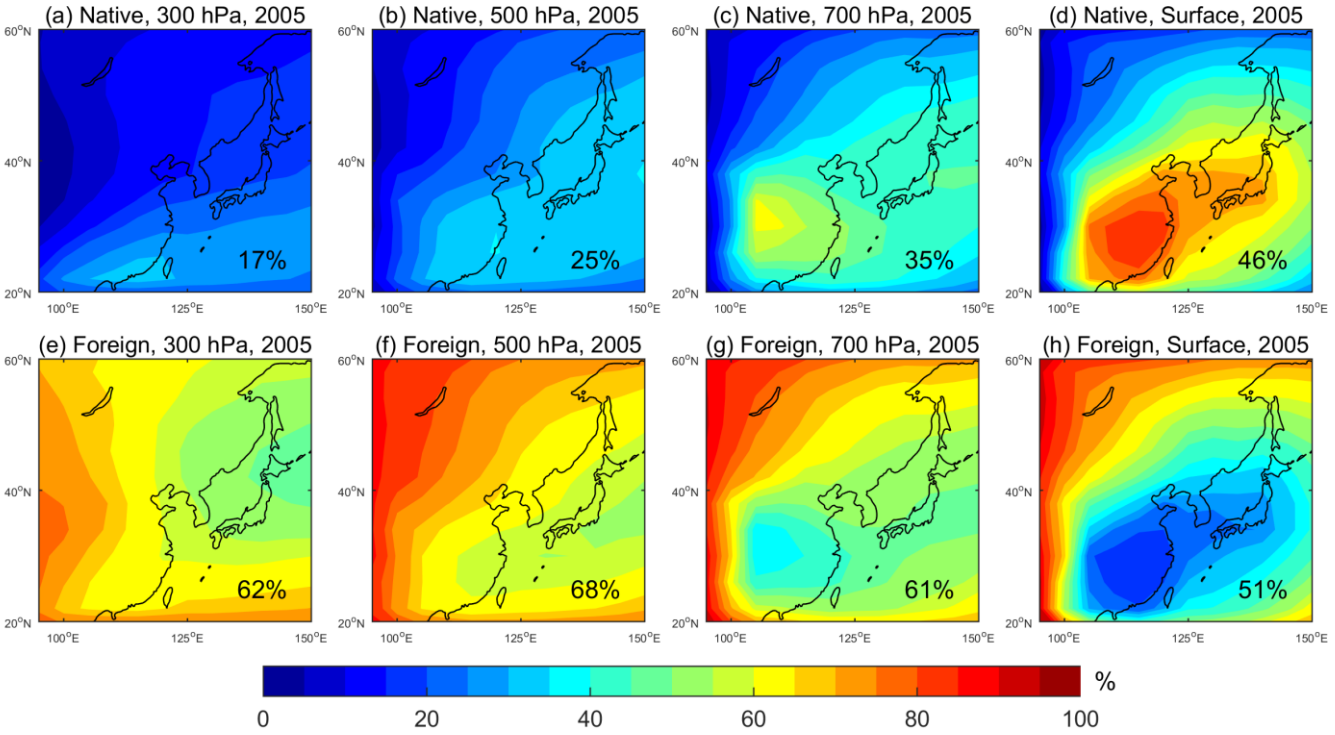
Figure 3. Fractional contributions (in %) of native (a-d) and foreign (e-h) ozone to ozone at different layers over East Asia in 2005. The regional mean is shown in the bottom right corner of each panel.

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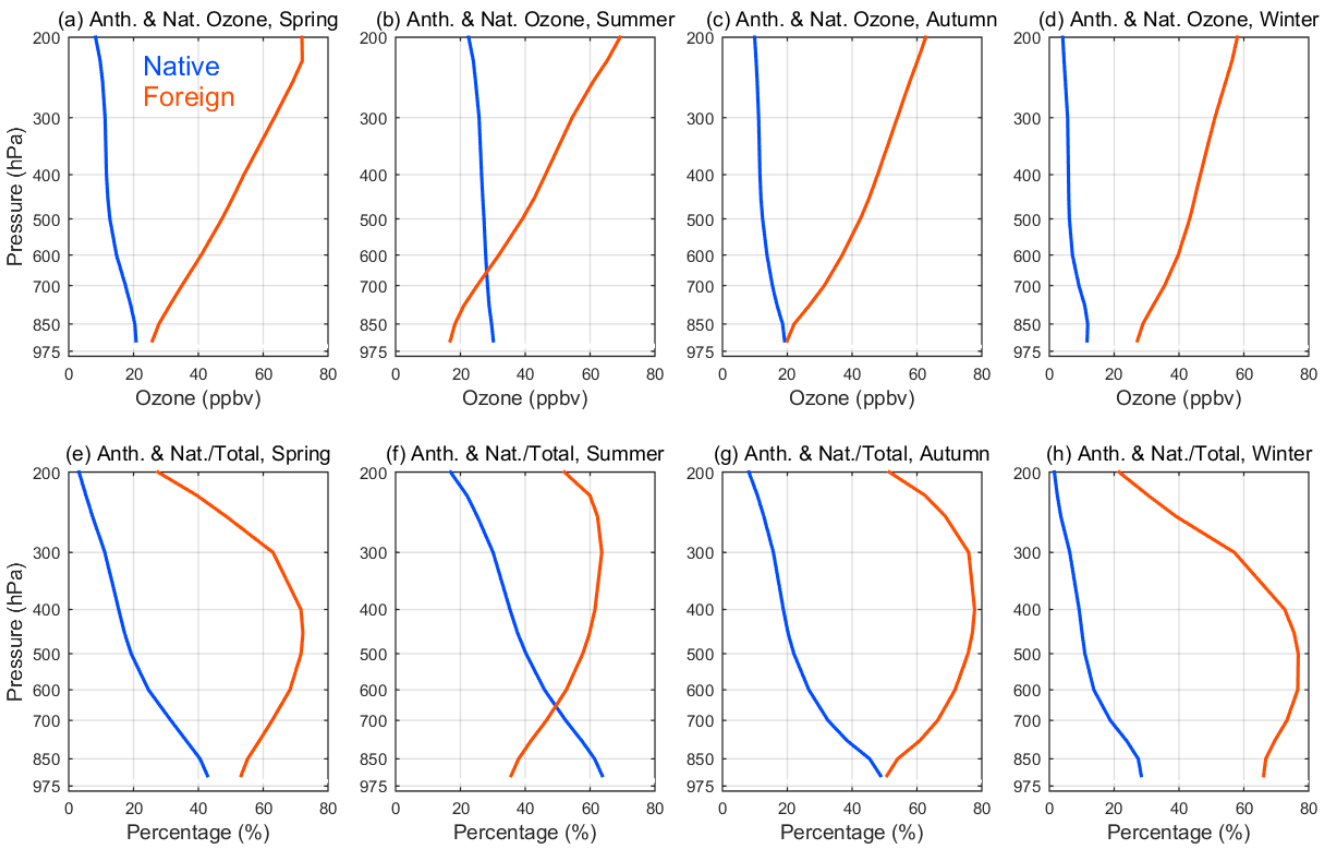
Figure 4. (a-d) Mean vertical profiles of native ozone (in blue) and foreign ozone (in red) (in ppbv) over East Asia by season in 2005. (e-h) The same as (a-d), but for the fractional contribution (in %) of each component to ozone in the East Asian troposphere at each altitude.

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1065 Figure 5. (a-d) Mean vertical profiles of native anthropogenic ozone (in blue) and foreign anthropogenic

ozone (in red) (in ppbv) over East Asia by season in 2005. (e-h) The same as (a-d), but for the fractional

contribution (in %) of each component to ozone in the East Asian troposphere at each altitude. Note the

difference in the scale of the x-axis between Figure 3 and Figure 4.

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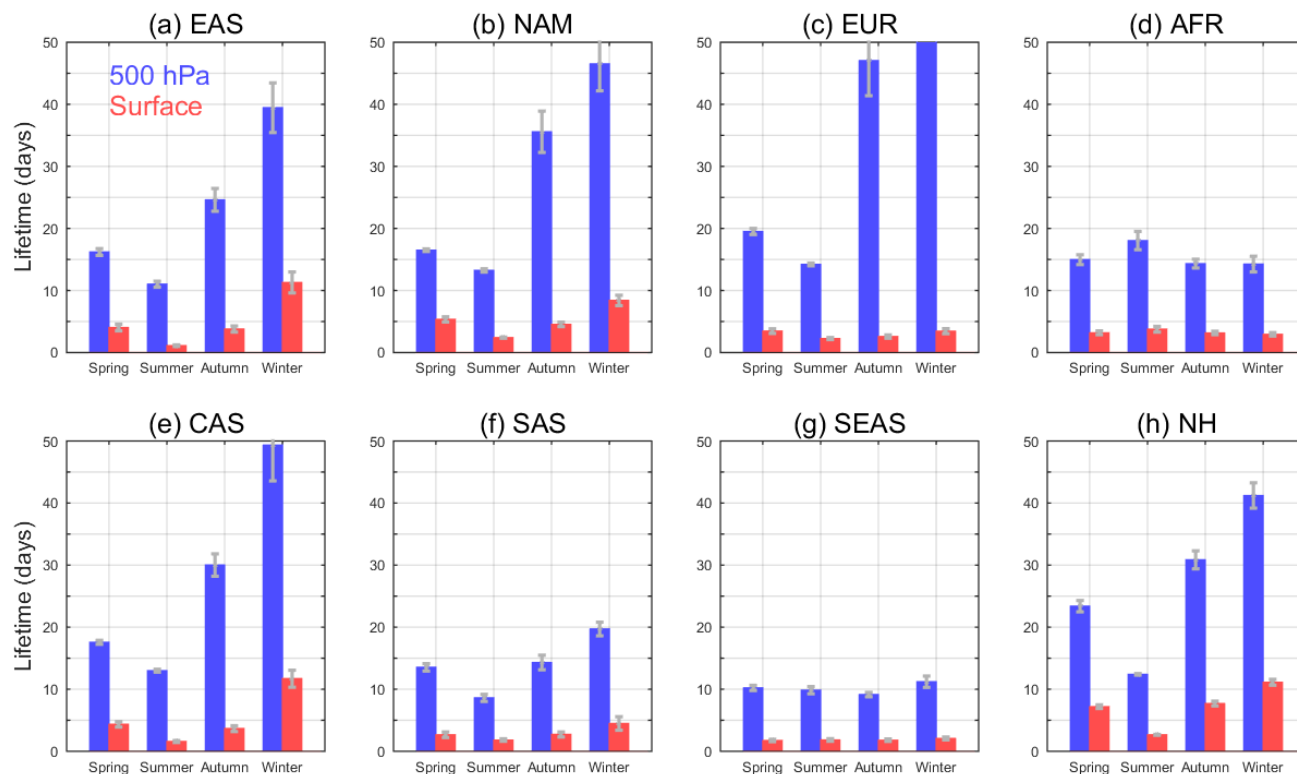
Figure 6. Ozone lifetime averaged in various regions in the middle troposphere (MT, 500 hPa) and at the surface in 2005, calculated from the ozone dry deposition and chemical loss rate in GEOS-Chem simulations. The error bar is 4 times the standard error of the mean (equivalent to the 95% confidence limit of the mean). The regions include (a) East Asia (EAS), (b) North America (NAM), (c) Europe (EUR), (d) Africa (AFR), (e) central Asia (CAS), (f) South Asia (SAS), (g) Southeast Asia (SEAS), and (h) the Northern Hemisphere (NH).

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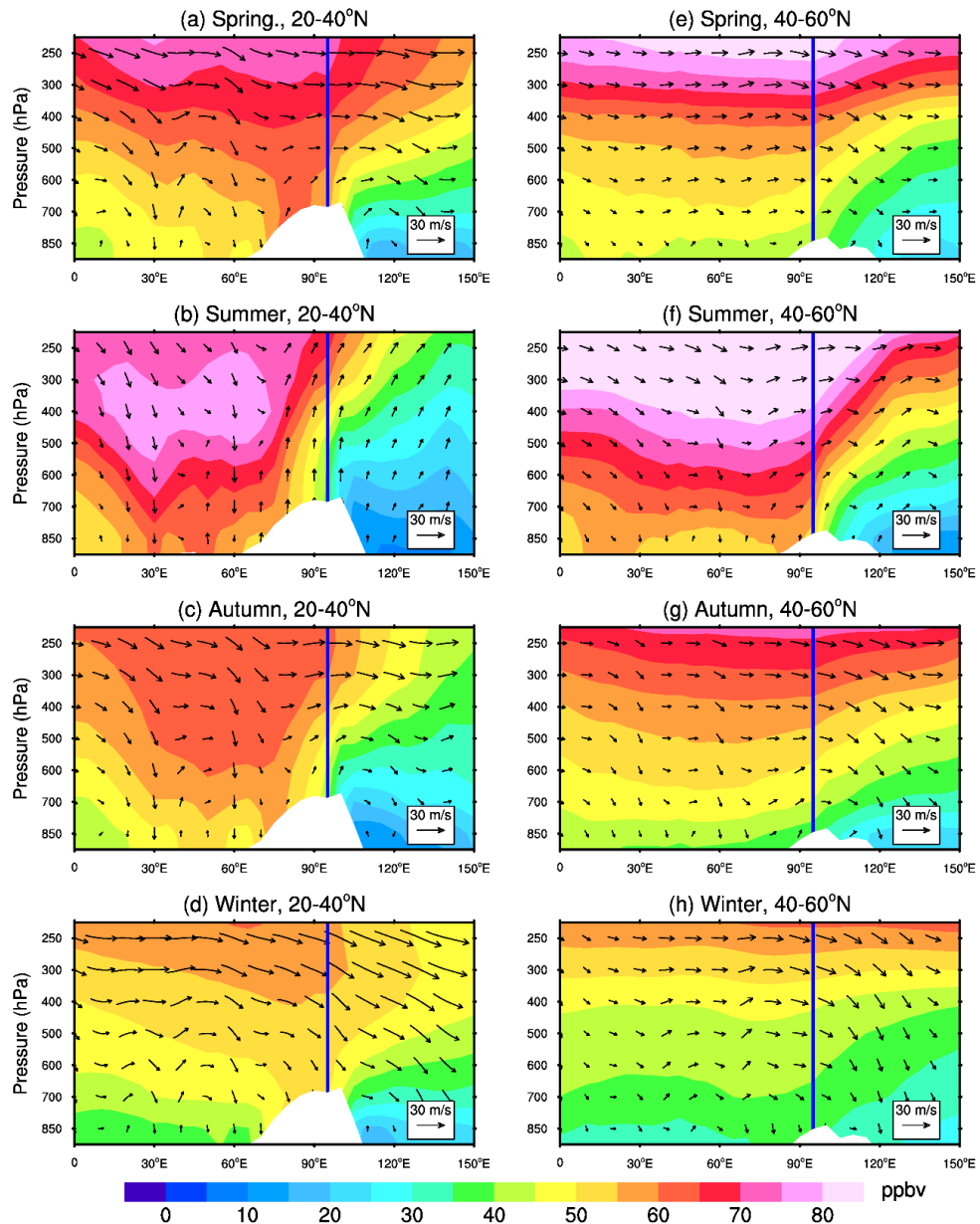
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1115 Figure 7. Longitude-pressure cross-sections of foreign ozone (in colour) and wind field (in arrows)

averaged between 20-40 N (a-d) and 40-60 N (e-h) in 2005. The white areas indicate topography. The

blue lines indicate the western border of East Asia. The vertical velocities in the p coordinates are

enlarged 1000 times for illustration purposes.
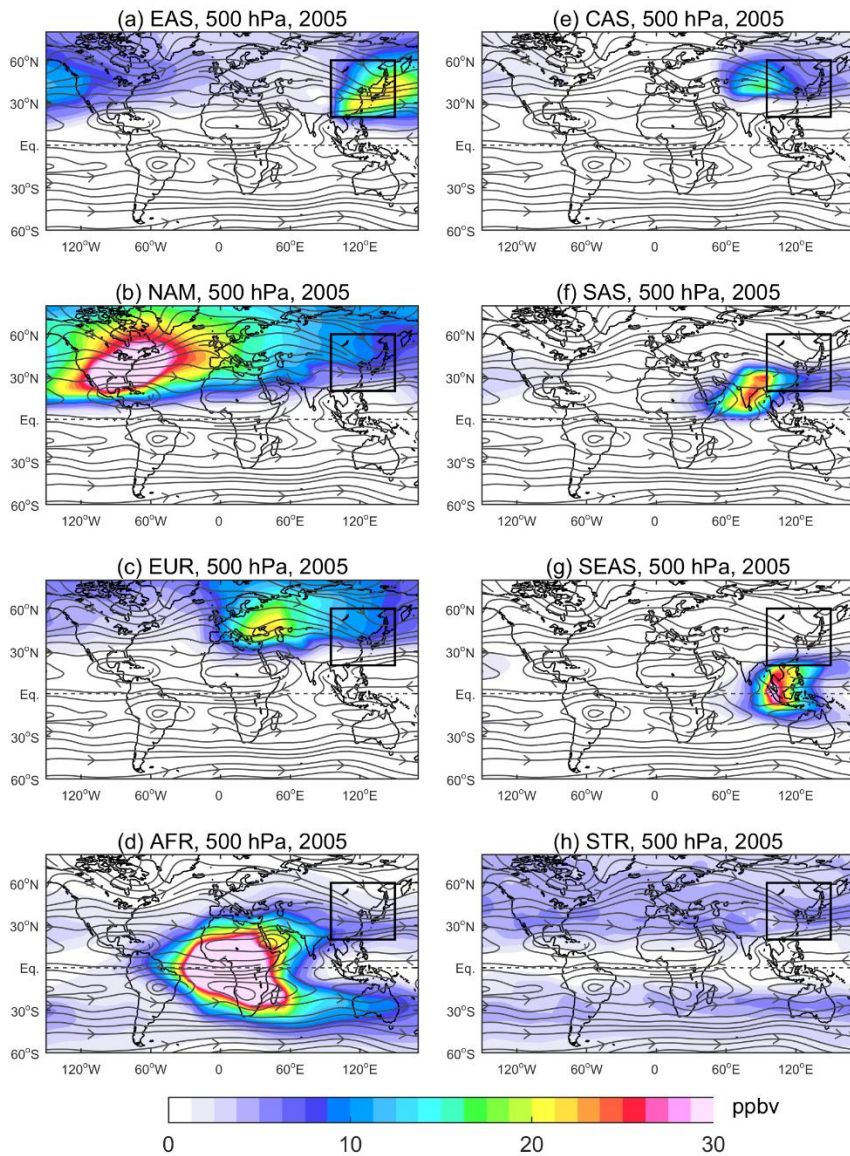
Figure 8. Horizontal distributions of ozone produced in different regions in the middle troposphere (500 hPa) averaged in 2005. The boxed area indicates East Asia. The abbreviations are for (a) East Asia (EAS), (b) North America (NAM), (c) Europe (EUR), (d) Africa (AFR), (e) central Asia (CAS), (f) South Asia (SAS), (g) Southeast Asia (SEAS), and (h) the stratosphere (STR).
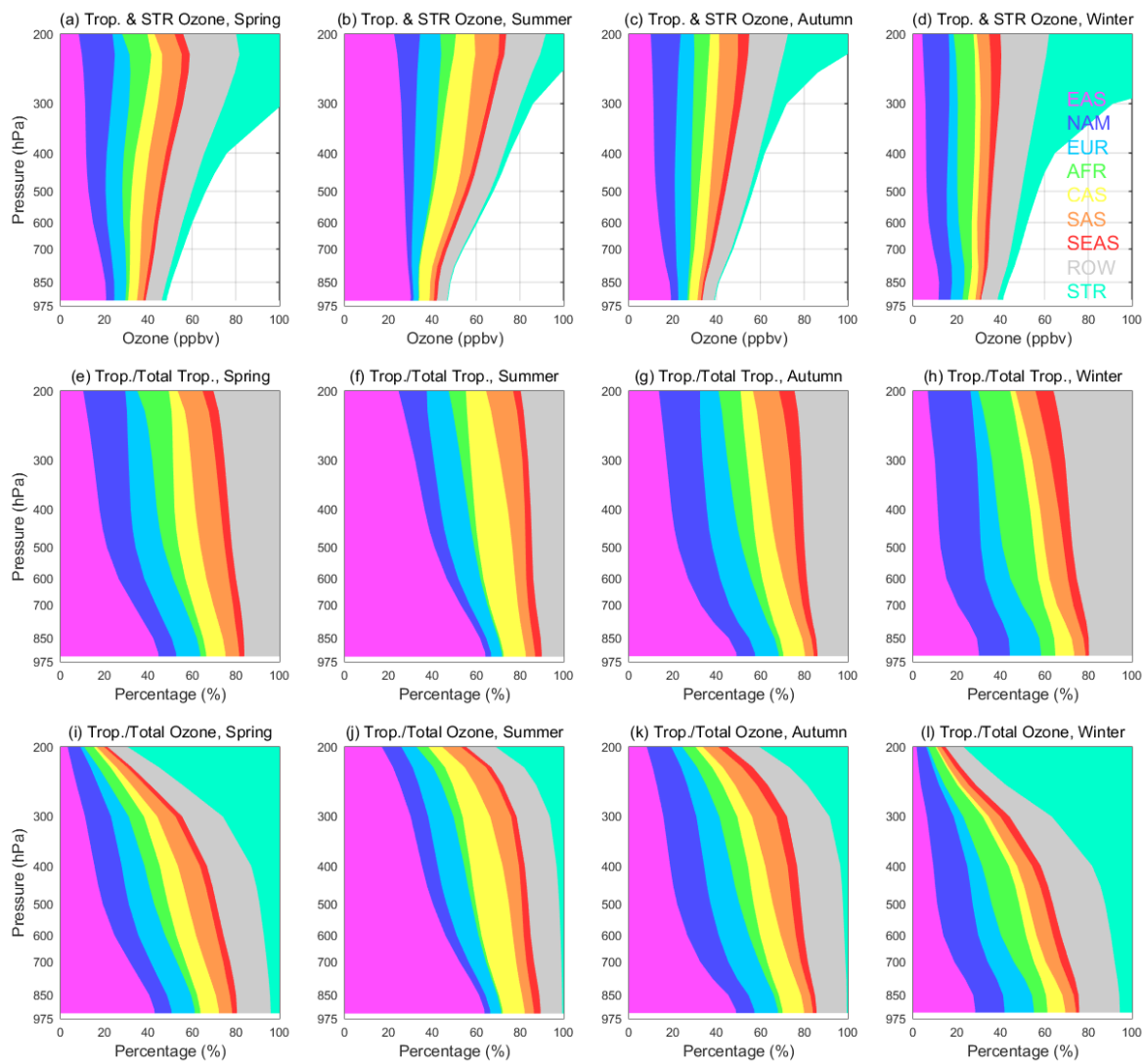
Figure 9. (a-d) Vertical variations of ozone (in ppbv) over East Asia from native and foreign sources by season. (e-h) The same as (a-d), but in terms of the factional contributions (in %) to ozone produced in the troposphere. (i-l) The same as (a-d) but in terms of the fractional contributions (in %) to ozone over East Asia. All values are the means over East Asia in 2005. The abbreviations are for East Asia (EAS), North America (NAM), Europe (EUR), Africa (AFR), central Asia (CAS), South Asia (SAS), Southeast Asia (SEAS), the rest of the world (ROW), and the stratosphere (STR).
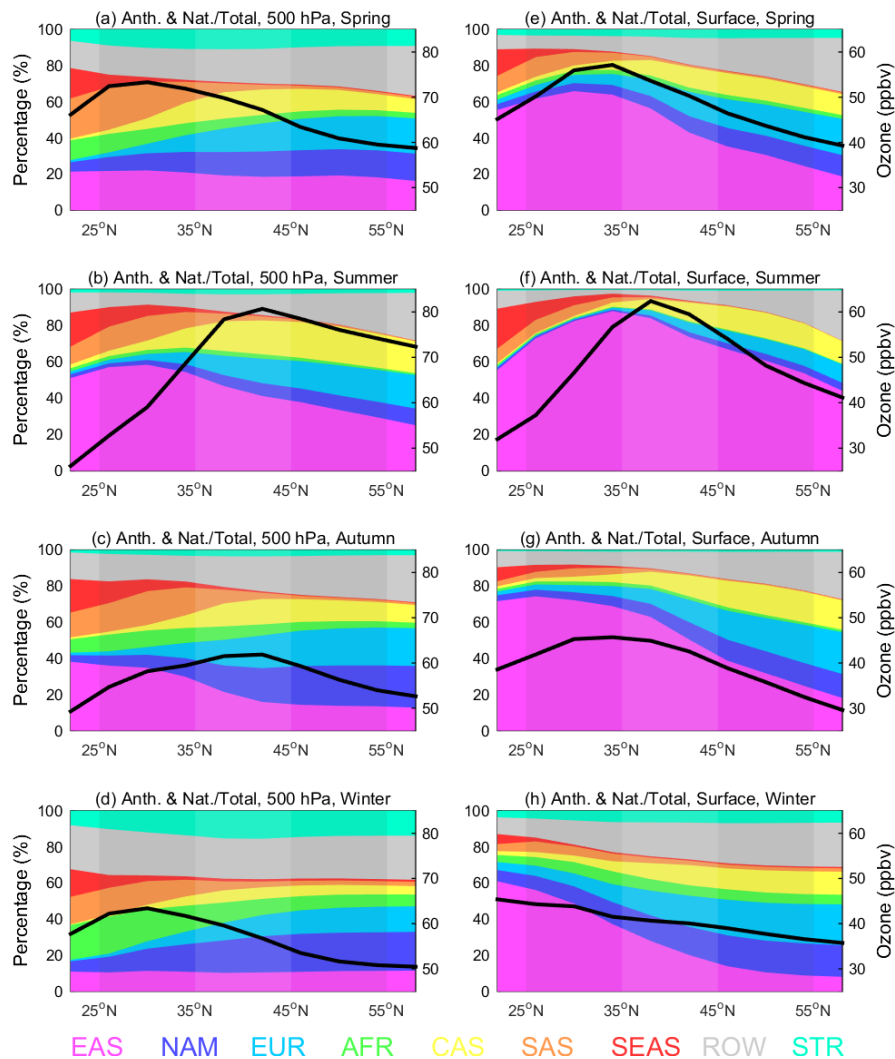
Figure 10. Latitudinal variations of the fractional contributions (in %, left y-axis) of native and foreign sources to ozone in East Asia in the middle troposphere (500 hPa) (a-d) and at the surface (e-h) in 2005. The black lines indicate ozone concentrations averaged over East Asia (in ppbv, right y-axis). The abbreviations are for East Asia (EAS), North America (NAM), Europe (EUR), Africa (AFR), central Asia (CAS), South Asia (SAS), Southeast Asia (SEAS), the rest of the world (ROW), and the stratosphere (STR).

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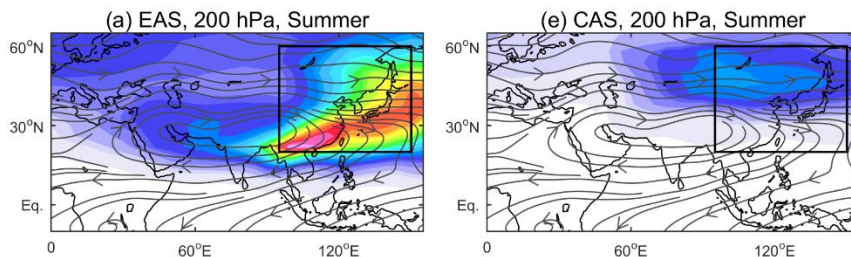
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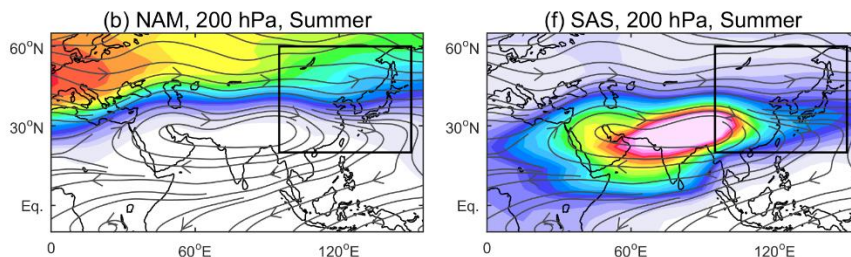
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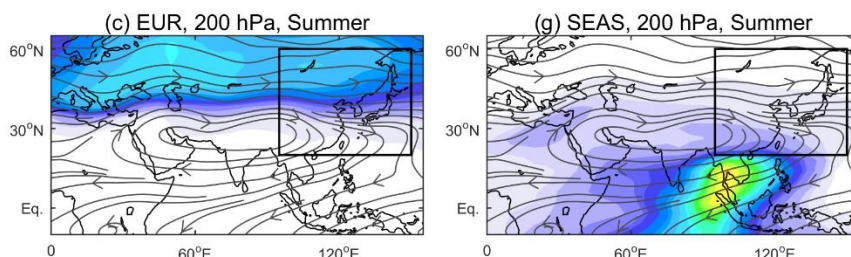
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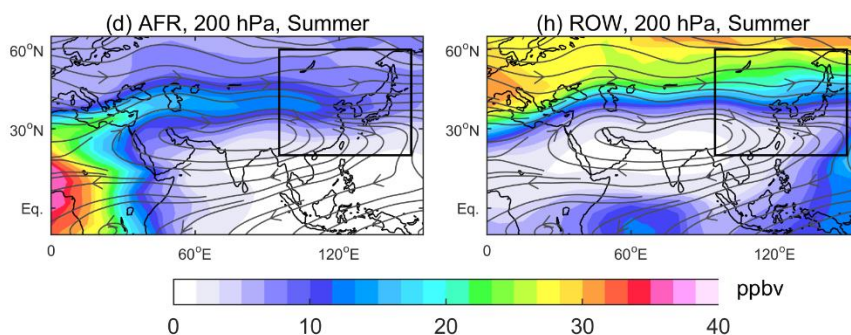
Figure 11. Horizontal distributions of ozone produced in different regions overlaid with streamlines in

the upper troposphere (200 hPa) in the summer of 2005. The boxed area indicates East Asia. The

abbreviations are for (a) East Asia (EAS), (b) North America (NAM), (c) Europe (EUR), (d) Africa

(AFR), (e) central Asia (CAS), (f) South Asia (SAS), (g) Southeast Asia (SEAS), and (h) the rest of the

1210   world (ROW).
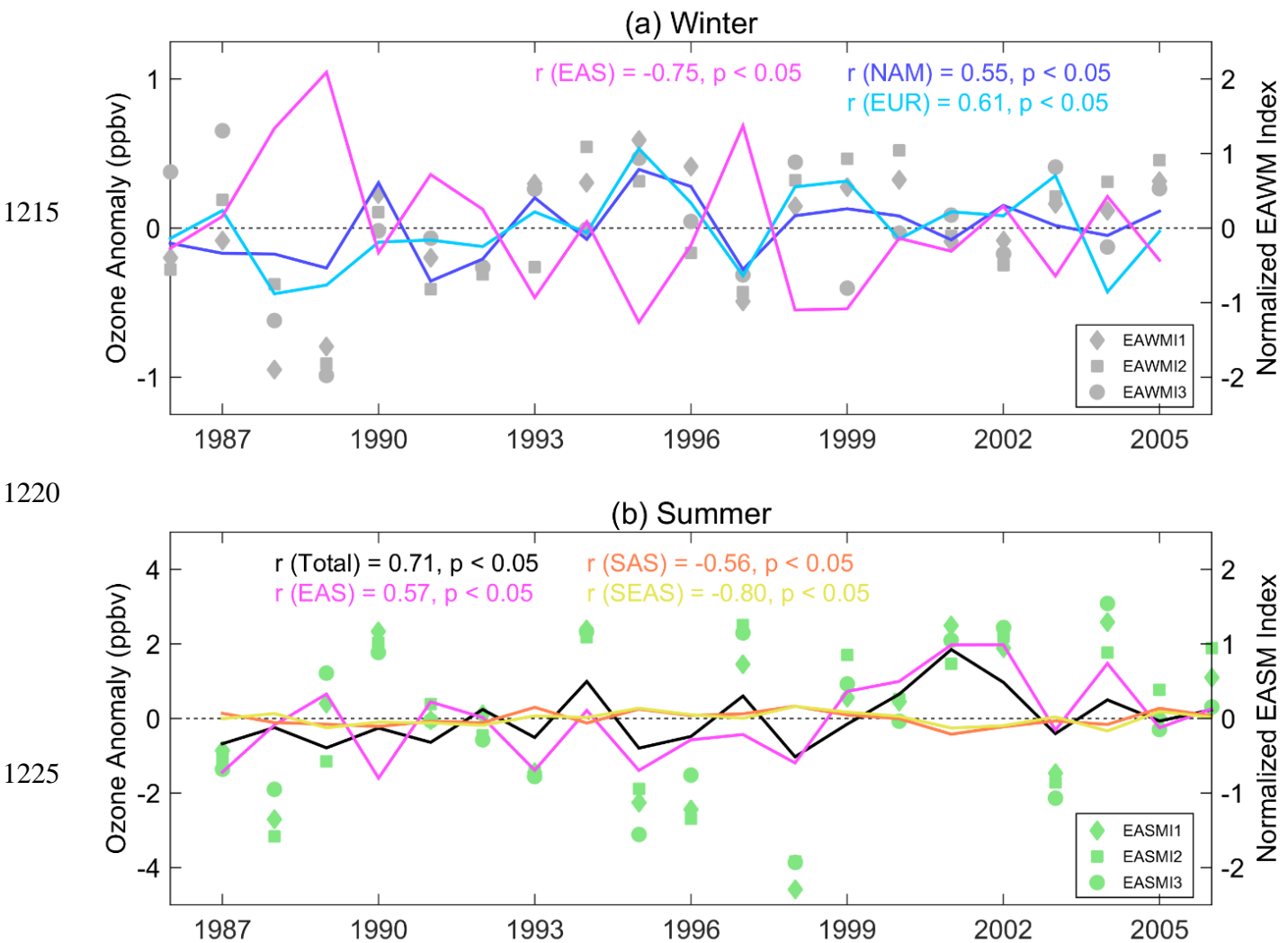
Figure 12. Interannual variations of the anomalies of foreign ozone (in ppbv, left y-axis) from various regions at the East Asian surface driven by meteorology (Fix-Chem) in winter (a) and summer (b). The values are averaged over 110-145 °E, 22-46 °N in winter and 95-130 °E, 22-46 °N in summer, where the correlations are the most significant. The correlation coefficients calculated with the three sets of monsoon indices in each season are averaged. The abbreviations are for East Asia (EAS), North America (NAM), Europe (EUR), South Asia (SAS), and Southeast Asia (SEAS).