We are very grateful to the reviewers for their valuable comments and suggestions, which have helped us greatly in improving our manuscript. We have addressed all the comments and revised the manuscript accordingly. The point-to-point responses are provided below in Italic. The comparison of our manuscript between this version and the last version is also provided.

Anonymous Referee #1

General Comment

This paper examined the tropospheric ozone in East Asia in terms of the influence of various source regions, particularly focused on the regions outside East Asia, they called them as "foreign ozone". The topic of the paper is well within the scope of the journal. The model used and the methods to deduce the influence of each different source region are adequate and have been applied so far in various studies for similar purpose. The results shown in the paper are generally consistent with the facts published in the previous literatures. This gives a certain reliability to the analysis done in this paper, but at the same time, the novelty of this paper over those previous studies is not clearly shown in the manuscript. For example, the authors reviewed the roles of East Asian Monsoon on the foreign ozone in East Asia in "Introduction", but the findings of this paper written in "Abstract" is quite similar to what was reviewed there. The author argued that this paper provided a comprehensive assessment of the influence of foreign ozone on the East Asian tropospheric ozone, but if they just want a comprehensive assessment, writing a review paper is more suitable, and actually the review given in this paper is quite comprehensive. I suggest the authors should state more clearly what they achieved on top of the previous studies to be published in the journal.

This is my general concern.

Thanks for the reviewer's comments and suggestions. We added more statements in this revision in various sections on what is new in our study, mostly in the section of

Discussion and Conclusions. This study reveals the significant foreign influences on tropospheric ozone over East Asia through global atmospheric transport and gains some new insights. Firstly, this study comprehensively assessed foreign influences on tropospheric ozone over East Asia, while previous studies investigated ozone transport from one or a few foreign regions (X. Li et al., 2014; Chakraborty et al., 2015), that during one or a few seasons (Ni et al., 2018) or only surface ozone in East Asia (Wang et al., 2011). Comparisons with previous studies show that, despite some disagreements concerning various details, our results appear to be reasonable.

Secondly, we examined the foreign influence on ozone in East Asia throughout all tropospheric columns. The simulations show that the concentration of foreign ozone increases 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 over the area (Myhre et al., 2017). Such an impact has been rarely documented (Sudo and Akimoto, 2007).

Thirdly, we highlight the influences of 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, significant correlations between the strength of EAM and the ozone transport from various foreign regions including Europe, South Asia, and Southeast Asia have been found. These findings provide further understanding of the mechanisms of the intercontinental transport of air pollutants to East Asia.

Major Comment:

Almost all analysis was done for the average over East Asia. As previous studies have shown that the relative contributions of various source regions can vary considerably depending on the location within East Asia. So, I cannot fully understand the meaning of such “East Asian averaged” contributions of various source regions. Actually, the latitudinal dependence in each foreign and native contributions were analyzed, but I guess the longitudinal dependence should be also large enough to be analyzed.

Thanks for the points. We have added two figures (Figures 3 and 8) to show how foreign ozone distributes horizontally over East Asia. The longitudinal variations of foreign ozone have been analysed as well (Figure S6).

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Figure 3 compares the fractional contributions of native and foreign sources for ozone over East Asia in terms of the annual mean. The fractional contribution of foreign ozone at high altitudes is greater than that at the surface, and can reach a regional mean of up to 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). The analysis has been added in the second paragraph of section 3.1.

Figure 8 presents how foreign ozone from different source regions distributes horizontally in the middle troposphere, illustrating significant foreign impacts on ozone over East Asia at the altitudes. 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. The analysis has been added in the first paragraph of section 3.2.1.

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, they are less obvious than the latitudinal variations, especially in winter (Figure 10 vs. Figure S6). In the East Asian middle troposphere (500 hPa), central Asian and South Asian ozone decreases with longitude in summer, varying insignificantly with *longitude in winter. At the East Asian surface, ozone from the two regions decreases with longitude. Longitudinal variations of ozone from North America, Europe, Africa, and Southeast Asia are less obvious than that for central Asia and South Asia. The analysis has been added in the last paragraph of section 3.2.3.*

Specific Comments:

- L45: The terms “native” and “foreign” are used before their definition is given in Table 2.

Thanks. Table 2 is moved into the supplement as Table S1. ‘Foreign ozone’ and ‘native ozone’ have been explained when the terms first appear.

- L83-85: Is the "transport" itself associated with thing other than meteorology such as emission and/or chemistry?

The ‘transport’ process itself is driven by the meteorology, specifically the atmospheric circulation or the wind field. The amount of ozone brought by the transport from the source region to the receptor region is related to the emissions and chemistry. Therefore, the influences of the transport on ozone over the receptor region is associated with all these factors. This sentence has been revised.

-L124-125: How did you treat CH4 chemistry in the model? Is it fully represented?

In the full chemistry simulation, the CH_4 *concentrations were fixed throughout the troposphere to annual zonal mean values in four latitudinal bands (90N-30N, 30N-Eq., Eq.-30S, and 30S-90S). The global annual mean anthropogenic emissions of* CH_4 *were from EDGAR v4.2. In our sensitivity simulations, the effects of* CH_4 *emissions on anthropogenic ozone were excluded. This limitation has been added in the discussion section.*

- L132-136: I don’t think these detailed definition of regions are necessary, since

Figure 1 shows them visually and they can be found in its caption.

Agree. The definition of regions is removed.

- L146 (formula (1)): How is the consistency between CTRL-EAnth-GLO and the sum in the denominator of the first term? How are they close to each other? Should be explained somewhere in the text.

In Equation (1), $\sum_{i=1}^{8}$ *(CTRL - EAnth-*X_i*) is the sum of the ozone response to the 100% perturbation for each region, CTRL - EAnth-GLO is the ozone response to the 100% perturbation for the globe. In the East Asian troposphere, ozone concentration from* $\sum_{i=1}^{8}$ *(CTRL - EAnth-*X_i*) is 0-4 ppbv (0-20%) higher than that from CTRL - EAnth-GLO (Figure S1). Figure S1 and explanations are added in this revision.*

- L159-160: The production and loss of ozone can vary considerably with in a day, so I imagine using daily production and loss data should have bad consequences on the simulated tagged ozone concentration. Did you check the validity for using the daily values for them?

Thanks for this point. Due to the diurnal variations of ozone and the nonlinearity in chemistry, using daily production and loss data is one of the uncertainties for the tagged ozone simulation. We have compared the simulations between the full chemistry run and the tagged ozone run to assess the overall uncertainty of tagged ozone simulation (Figures S2-S3). The results show that the difference of the ozone concentrations in the two simulations is within ±5%. Results from the two types of simulations were also compared in Han et al. (2018). We also listed this as one of the sources of uncertainties in the section of Discussion and Conclusions.

- Table 2: I don't think Table 2 could effectively explain the different definition of the terminology used in the manuscript. For me, the caption of Table 2 is easier to understand what you want to explain than Table2 itself and the descriptions in the main text (L194-210).

Table 2 has been moved into the supplement (Table S1). The terms are explained at their first appearance or along with the experiment description.

- L233-234: You can not compare the contribution of foreign ozone on whole East Asia and that on China

Thanks for this point. The sentence is removed.

- L248: How did you calculate the chemical lifetime? Explain it in the text.

'The chemical lifetime' has been revised to 'the lifetime'. For each model grid in the boundary layer, the lifetime of ozone was calculated by the daily average dry deposition and chemical loss rate of ozone. For each grid in the free troposphere, only the chemical loss rate of ozone is used. The explanation is added in the revision.

- L251-252: Only the dry deposition could be the cause of the difference? Are there any other causes which should be mentioned here?

The dry deposition and active chemical reactions of ozone in the boundary layer are the two main reasons for the shorter lifetime of ozone at the surface than in the free troposphere. The sentence has been revised accordingly.

- L267-269: This part should be more specific. Where is the East Asian trough? Which region of downdraft you are referring?

The East Asian trough in the middle troposphere is an important feature of the EAWM system. It locates along ~130-140°E in winter (see Figure 6a in Y. Zhu et al. (2017)). Downdrafts prevail behind the East Asian trough in winter from 100°E to 140°E. The locations of the East Asian trough and downdrafts are added in the text.

- L284-287: This sentence is quite hard to understand logically. The difference between foreign and foreign anthropogenic O3 is not mentioned in the previous sentences. I cannot understand what you want to mean here.

Thanks. The sentence is removed.

- L342: Why the contribution from Africa in winter can be so large? Why it can be larger than that from SAS SEAS where much closer to EAS.

Thanks for this question. Ozone transport from Africa to the East Asian middle and upper troposphere is mainly driven by the Hadley circulation and the subtropical westerlies. Unlike South Asia and Southeast Asia, Africa covers areas in the Northern Hemisphere (NH) and the Southern Hemisphere, with around three-quarters of the continent in the tropics. In NH winter, there still exists strong convection over the intertropical convergence zone (ITCZ) in Africa. In NH winter, the ITCZ in Africa uplifts biogenic emissions to the middle and upper troposphere, where is with high lightning NO_x *emissions. Therefore, ozone concentrations in the African middle and upper troposphere are relatively high. Furthermore, African ozone in the middle and upper troposphere can be efficiently transported to East Asia by the subtropical westerlies. Compared with Africa, although South Asian and Southeast Asia are more close to East Asia, the atmospheric circulations are less favorable to the ozone transport to East Asia (Figure S5). See Han et al. (2018) for more specific analysis of ozone transport from Africa to East Asia. We added explanations on this in the third paragraph of section 3.2.1 in this revision.*

- L442 (formula(8)): What is U'850?

Thanks. U'850 is the anomaly of the zonal wind at 850 hPa. The explanation is added.

- L484-487: Where should I look at in Figure 3 and 7 to find these values? Is it annual evaluation or seasonal? This is a quite blur sentence.

This sentence has been revised. The concentrations of foreign ozone and native ozone is compared on the annual average in this sentence. Such a comparison in the four seasons is added in section 3.1.

Anonymous Referee #2

This publication represents a robust analysis of contributions to surface and tropospheric ozone. While several prior studies have found similar results, and in this sense the study is maybe not completely new, the comprehensive analysis of contributors to ozone over Asia has clearly added value.

On the downside, I think it is regrettable that the authors have not attempted to align their study better with the HTAP2 source-receptor studies, that included harmonized simulations of emission sensitivities and responses over East Asia and a number of other world regions, updated and harmonized emissions, etc. As a result, it is becoming more difficult to evaluate uncertainties related to the use of one specific model compared to other models.

Nevertheless, I find the overall analysis convincing, the material well presented, and therefore recommend to publish the manuscript in ACP, with some suggestions for minor revision presented below.

Minor suggestions:

l. 17 East Asia defined as : : :.

We added the geographic boundaries to define the domain of East Asia in this study.

l. 48 the ecosystem=>ecosystems.

Thanks. The sentence has been revised.

l. 54 why?

This sentence is revised to explain why.

l. 57 I don’t find this in Fiore’s paper. Anyway as the ozone response depends on the emission reduction strength, the sensivity will depend on the magnitude of perturbation. In this sense perturbations that are close to the present situation (i.e. a 5, 10 or 20 %) are used for Source-receptor relationships, sometimes with a correction for larger perturbation sizes. These have been used e.g. in HTAP1, and HTAP2, and other studies by individual researchers also for Asia. A nice paper that combines source attribution and tagging paper: https://www.geosci-model-dev.net/11/2825/2018/. It can be used to address some of the uncertainties. I also note that it is not very clear around

Thanks for this point. It is better to call this method as "perturbation (sensitivity) method "as suggested by Butler et al. (2018). Reducing the emissions of ozone precursors in source regions to zero in this study is one of the scenarios of the perturbation method. In the manuscript, the 'emission zero-out' has been revised as 'emission perturbation'.

l. 154 how exactly the tagging was done- and there are several ways to do so.

In this study, ozone molecules were tagged based on the geographical model domains in which the ozone molecules are formed (Wang et al., 1998). Using the daily ozone production and loss data archived from a full chemical simulation conducted beforehand, net ozone production at each model grid was resolved. For a specific source region A, ozone produced in A is labelled as a tracer. The tracer excludes the ozone molecules formed outside A. Therefore, how the tracer distributes spatially directly show the ozone transport from A to the outside. The amount of the tracer in a receptor region can be directly attributed to ozone production in A. Overall, the tagged ozone simulation in this study tracks ozone produced in the troposphere over each of the defined source regions along its transport into a receptor region. The explanation is added in the last paragraph of section 2.

l. 98. It is confusing to talk about trends when you really talk about interannual variability.

Thanks for the point. The discussion about the trend has been deleted.

l. 127 I notice that this resolution is meanwhile not really state-of-the-art. Mention already here that you also do higher resolution sensitivity simulations

Thanks. We added statements on our run at higher resolutions.

l. 130 Unfortunately the different choice of regions compared to HTAP1 or HTAP2 does not help in comparing to other model simulations.

Definition of the study domains in this study is slightly different to that in HTAP1, but with similarities (Table R1). The comparison has been added.

Table R1. Comparison of the definition of the study domains between this study and HTAP1.

	This study	*HTAP1*
East Asia	*95°E-150°E, 20°N-60°N*	*95°E-160°E, 15°N-50°N*
North America	*170°W-65°W, 15°N-70°N*	*125°W-60°W, 15°N-55°N*
Europe	*15°W-50°E, 35°N-70°N*	*10°W-50°E, 25°N-65°N*
South Asia	*60°E-95°E, 5°N-35°N*	*50°E-95°E, 5°N-35°N*

142-145 I guess what the authors are doing here is introducing a correction factor so that the sum of the individual region 100 % perturbations nicely sums up to a global perturbation? How large are these corrections?

Yes. In Equation (1), $\sum_{i=1}^{8}$ *(CTRL - EAnth-*X_i*) is the sum of the ozone response to the 100% perturbation at each of the defined regions, CTRL - EAnth-GLO is the ozone response to the 100% perturbation for the globe. In the East Asian troposphere, ozone concentration from* $\sum_{i=1}^{8}$ *(CTRL - EAnth-*X_i*) is 0-4 ppbv (0-20%) higher than that from CTRL - EAnth-GLO (Figure S1). For each source region, the correction over East Asia is less than 1 ppbv. The magnitude of these corrections are added.*

l. 164. Linoz. Does this effectively mean a constant yearly influx of stratospheric ozone by 484 Tg in these variation. Note that it is likely that there is a correlation between large scale circulation (and I guess monsoon as well) and strat-trop exchange.

Thanks. The influx of stratospheric ozone to the troposphere varies interannually and is ~484 Tg in 2005. The explanation has been added.

l. 137 Why were 2006/2007 chosen? And not more recent years (e.g. HTAP's 2010)

Thanks. In this study, GEOS-Chem was driven by GEOS-4 meteorological data, which has strong performance in simulating tropospheric ozone (Choi et al., 2017; Y. Zhu et al., 2017; Han et al., 2018). GEOS-4 covers 1985-2006, which is the study period here.

l. 157 No variation in chemistry. In the next line the authors explain this is achieved by extracting production/loss data for 2005. This is going to lead to inconsistencies (and hopefully identical results for 2005). This needs discussion.

Thanks. Daily ozone production and loss data in 2005 were used for all the years from 1986 to 2006. Therefore, in the simulations, the daily data in 2005 allow a seasonal variation, but no interannual variation in chemistry. Here "No variation in chemistry" means no interannual variation in chemistry. The sentence is revised to clarify the meaning.

l. 170. The results of using different resolutions and meteorological drivers, needs to be somehow included in the discussion of uncertainties.

Thanks. Added in the section of Discussion and Conclusions.

l. 194 here the operational definition of troposphere needs to be given.

Thanks. Added.

l. 202-204 In this case North American ozone is both from natural and anthropogenic emissions as well as stratospheric ozone that entered the North American region?

Thanks. For a specific region, ozone produced in the troposphere over that region is named after that region, such as 'North American ozone'. Ozone produced in the stratosphere and then entered the troposphere is labelled as 'stratospheric ozone', an independent tracer. So, North American ozone excludes stratospheric ozone that entered North America. The original Table 2 (present Table S1) explained this. We further clarified this in this revision.

l. 330 I recommend also to consider the results of Turnock et al, ACP, 2018, which discusses HTAP2 results.

Thanks. Added.

l. 346. This finding warrant a bit more discussion, given the similarity of emissions but longer distance compared to Europe.

Thanks. The expression has been revised. On annual average, North America and Europe contributes 5-13 ppbv (7-12%) and 5-7 ppbv (3-11%) to ozone in the East Asian middle and upper troposphere, respectively. The annual mean of North American ozone is higher than that of European ozone over East Asia at layers above 500 hPa.

To compare the two, we further conducted four sensitivity experiments. In two of simulations, biogenic emissions in North America and Europe were turned off, respectively. In another two simulations, lightning NO$_x$ *emissions in North America and Europe were turned off respectively. The difference between the results from the control experiment and those from these four sensitivity simulations are shown in Figure S4. It is demonstrated that the difference between North America and European ozone over East Asia is from both anthropogenic and natural emissions including biogenic and lightning sources. The analysis has been added in the first paragraph of section 3.2.1.*

l. 368 I guess that can well be, also given the fact that many other models have larger stratospheric influxes. Would the conclusions change substantially if the number would be double. It is unclear to me how stratospheric ozone influx is accounted in the various attribution methods (e.g. influx of ozone In Europe, is that European ozone as well?)

Thanks for the points. In this study, stratospheric ozone is ozone produced in the stratosphere and then transported into the troposphere. Stratospheric-to-troposphere ozone flux is the amount of the stratospheric ozone that entered the troposphere. European ozone is the ozone produced in the European troposphere. So, stratospheric-to-troposphere ozone flux over Europe is excluded in European ozone. Stratospheric ozone transported downward to the troposphere is showed in Figures 9 and 10. The values of ozone concentrations from different source regions (Figures 9a-9d) would not changed if the stratospheric ozone is doubled. We assessed the fractional contribution of ozone from different source regions in Figures 9e-9h and Figures 9i-9l, respectively. A change in the influxes of stratospheric ozone would not affect the results in Figures 9e-9h but in Figures 9i-9l. This is explained in section 2 in this revision.

l. 410 It is not so clear why you would need 3x2 monsoon indices. If they are so different, please summarize what aspects they would represent stronger. Please also clarify how monsoon (summer phenomenon) indices also can have significant correlation with winter ozone.

Thanks. We used three monsoon indices for winter and three for summer, respectively. In each season, the three indices describe the features of the EAM from different perspectives. The monsoon indices were correlated with ozone variation in the same season. The monsoon indices in summer were not connected to wintertime ozone. The explanation has been added. We have also added the correlation coefficients for each *of the indices in Table S2, and the corresponding analysis is added in section 4.*

l. 540 It would have been great to use the HTAP2 compilation for 2008/2010- as did many other models. It would be good to mention at least the differences.

Thanks. In future studies, the HTAP2 emission inventory can improve the estimates of foreign influences on ozone over East Asia and their long-term trends. The difference between the anthropogenic emissions in this study and that for HTAP2 is briefly discussed in the last section. From 2000 to 2010 in EDGAR emission inventories, NO_x*, CO, and NMVOCs respectively changed by 9.5%, -1.2%, and 5.2% globally, and the three species decreased across North America and Europe but increased in East Asia (Turnock et al., 2018).*

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 ~~and its variations with space and time ~~using a global chemical transport model, GEOS-Chem, ~~for emission zero-out and ~~through the tagged ozone and emission perturbation simulations. The results show that foreign ozone ~~varies significantly with latitude, altitude, and season in the East Asian troposphere. The transport of foreign ozone ~~is transported to East Asia ~~occurs primarily~~(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, ~~where ~~the ~~concentration~~regional mean concentrations of foreign ozone ~~(~~range 32-65 ppbv~~) in East Asia is ~~, being 0.~~5-6~~8-4.8 times higher than ~~that of ~~native ozone ~~(~~concentrations of 11-18 ppbv~~) and has strong seasonality, being largest~~. Over the East Asian tropospheric column, foreign ozone appears most in spring and ~~lowest in winter. Foreign ozone in ~~least in summer when the South Asian High constrains North

American, European, and African ozone to north of 35°N of East Asia ~~increases rapidly with altitude.~~. 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 ~~average~~mean of foreign ozone ~~concentration~~concentrations is ~22.2 ppbv, which is comparable to its native counterpart of ~20.4 ppbv. The annual mean ~~concentration~~ 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). ~~The presence of foreign~~Foreign ozone concentrations at the East Asian surface ~~is~~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. ~~In summer~~Interannully, the ~~South Asian High facilitates the build-up of South Asian ozone in the East Asian upper troposphere and constrains North American, European, and African ozone to the regions north of 35°N. The interannual variations~~variation of foreign ozone at the East Asian surface ~~have been~~is found to be closely related to the intensity of the EAM. ~~When~~Specifically, the stronger the EAWM is ~~strong,~~in a winter, the more ozone is from North ~~American~~America and ~~European ozone are enhanced at~~Europe, because of the ~~East Asian surface, as the~~stronger subsidence behind the East Asian trough ~~becomes stronger. In strong EASM years,~~. In summer, ozone from South and Southeast ~~Asian ozone~~Asia is reduced ~~at the East Asian surface~~in strong EASM years due to weakened south-westerly monsoon ~~wind~~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), ~~climate (B. Li et al., 2016), economics (Lin et al., 2014), and the ecosystem (Zhang et al., 2017).~~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 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 ~~are increasing~~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; Nopmongcol et al., 2017) and Europe (Karamchandani et al., 2017; Knowland et al., 2017; Jonson et al., 2018). ~~Nevertheless,~~Therefore, research on ozone ~~transport~~outflow from ~~foreign regions into~~East Asia to the ~~East Asian troposphere~~rest of the world has ~~received~~been active, while relatively less attention has been paid to the ozone inflow from foreign regions to East Asia.

~~Previous studies have assessed foreign contributions to surface ozone in East Asia~~

~~by two types of~~ Two numerical simulations~~:~~ have been widely used for studying source-receptor relationships (Butler et al., 2018): emission ~~zero-out (also known as brute-force or~~ perturbation (sensitivity~~,~~) (Fiore et al., 2009) and ~~tagged ozone~~tagging tracer (Wang et al., 1998; Hou et al., 2014; Y. Zhu et al., 2017) simulations ~~(Liu et al., 2011).~~. The ~~emission zero-out~~perturbation simulation examines how ozone within a receptor region responds to a perturbation of ~~the~~ ozone precursor emissions in different foreign regions, while the tagged ozone simulation ~~tracks~~link ozone ~~produced~~ in ~~separate~~each of model grids with its source regions ~~along its transport into a receptor region. Based~~. Concerning air quality, most previous studies focused on ~~the simulations, some~~foreign influences on the surface-layer in East Asia. These studies ~~have revealed~~showed that the ~~concentration~~concentrations of ozone from foreign ~~ozone~~regions are larger at the East Asian surface ~~is larger~~ 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 ~~ozone~~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 (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~~, respectively.~~ . 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).

~~Concerning air quality, previous studies were mostly focused on foreign influences on the surface-layer in East Asia. How foreign ozone~~Limited~~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). However, limited~~ studies have suggested that foreign influences are much larger in the higher altitudes than at the surface in East Asia, such as ~~shown in~~ Y. Zhu et al. (2017) and Han et al. (2018) ~~for~~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 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 strongly latitude- and altitude-dependent radiative forcing of tropospheric ozone requires a further examination of the vertical variation of ~~foreign~~imported ozone in East Asia. Quantifying ~~foreign~~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.

~~The transport of foreign ozone to East Asia is associated with various factors, including emissions, meteorology, and chemistry in the source regions, along the transport pathways, and in East Asia (Han et al., 2018). Although previous studies have not fully clarified the drivers of the seasonal variation of foreign influences, they have specifically~~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 ~~foreign~~ ozone from different foreign regions to China. Y. Zhu et al. (2017) and Han et al. (2018) demonstrated the importance of the EAM to the vertical transport ~~in the EAM~~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. ~~Therefore, for better understanding of the transport of foreign ozone to East Asia, the role of~~(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.

~~On the decadal scale, foreign ozone has been found to significantly drive the interannual variations in tropospheric ozone over East Asia (Chatani and Sudo, 2011; Sekiya and Sudo, 2012) and even lead to increasing or decreasing trends over some regions of East Asia (Nagashima et al., 2017). Y. Zhu et al. (2017) and Han et al. (2018) found that interannual variations in the transport of North American and African ozone to East Asia are closely related to the variation of the EAM intensity. However, factors controlling the interannual variations of foreign ozone over East Asia have not been adequately studied.~~

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 ~~those~~that from one or a few source regions (X. Li et al., 2014; Y. Zhu et al., 2017; Han et al., 2018), ~~those~~that occurring during one or a few seasons (Ni et al., 2018), or ~~those~~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 ~~not well~~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. ~~It is desirable~~Therefore, this study aims to ~~systematically examine the~~address all of the research gaps and engage in a comprehensive assessment of foreign influences on tropospheric ozone in East Asia ~~according to space and time, as well as by source region.~~ .

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, ~~latitudinal~~horizontal, and vertical variations of these influences and explore the potential mechanisms. We also search for a link between the interannual variations of ~~the~~ foreign ~~ozone~~ influences and the EAM. In the following ~~portions of this paper~~, 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

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 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 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 "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 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.

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 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 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.

~~GEOS-Chem was run in two modes: the full chemistry and tagged ozone modes, corresponding to the emission zero-out and tagged tracer simulations. The former and the latter simulate anthropogenic ozone and overall ozone from foreign regions, respectively.~~ Table 1 describes the experiments conducted in this study. We divided the world into eight regions (Figure 1), including 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°E-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),~~, South Asia~~ (SAS, 60°E-95°E, 5°N-35°N),~~, Southeast Asia~~ (SEAS, 95°E-140°E, 10°S-20°N),~~, and the rest of the world ~~(ROW).~~. 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 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 ~~source~~ regions and in the ~~rest of~~whole world (*EAnth-GLO*). The anthropogenic ozone from a region X_i can be represented by the ~~world. As~~difference between *CTRL* and the sensitivity experiment for that region ($EAnth\text{-}X_i$) (X. Li et al., 2014). Because ozone does not linearly respond to the reduction of its precursors (Fiore et al., 2009), ~~to~~the sum of the ozone response to the 100% perturbation for each region $\sum_{i=1}^{8} (CTRL - EAnth\text{-}X_i)$ is not equal to the ozone response to the 100% perturbation for the globe (*CTRL - EAnth-GLO*). Specifically in this study, ozone concentration from $\sum_{i=1}^{8} (CTRL - EAnth\text{-}X_i)$ is approximately 0-4 ppbv (0-20%) higher than that from (*CTRL - EAnth-GLO*) over East Asia depending on altitude (Figure S1). Therefore, to fit the total anthropogenic ozone and isolate the relative contributions of anthropogenic emissions from different ~~source~~ 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:

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

where *EAnth-A* indicates the sensitivity experiment for *A*. The calculated *CON-A* is the ~~ozone contributed by the~~ anthropogenic ~~emissions~~ozone from a specific source region *A*~~. *EAnth-Xi* indicates one of the sensitivity experiments. The difference between *CTRL*~~ and ~~*EAnth-GLO* represents the total anthropogenic ozone.~~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 “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, ~~the meteorology was allowed to vary interannually, while there was no year-to-year variation in chemistry. We label this simulation~~labelled as ~~Fix-Chem in~~ Tagged-Ozone (Table 1~~. Note that seasonal variation in chemistry was allowed in the simulation. Daily~~), daily ozone production and loss data in 2005 were ~~extracted~~produced from the full chemistry simulation (CTRL~~). Then, Fix-Chem was conducted using the achieved ozone production~~) beforehand and ~~loss data. Fix-Chem~~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 ~~10~~ten tracers, i.e., ozone, ozone from the stratosphere (STR), and ozone produced in the troposphere over East Asia and ~~over~~ the seven foreign regions (Figure 1). ~~A~~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 (~~Linoz,~~ McLinden et al., 2000) was used in the calculation of stratospheric ozone~~, yielding a~~. The calculated global cross-tropopause ozone ~~of~~ 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.

~~To assess the sensitivity of the GEOS-Chem simulation to different meteorological data, spatial resolutions, and simulation modes, we ran GEOS-Chem driven by both GEOS-4 and GEOS-5 at two spatial resolutions (4° by 5° and 2° by 2.5° in latitude and longitude) in full chemistry mode and using GEOS-4 data at 4° by 5° resolution in tagged ozone modes.~~ Figure 2 compares the ozone vertical profiles from ~~these~~the simulations~~,~~ with different meteorological data and resolutions, averaged over East Asia by season. Among the ~~different meteorological data and resolutions, the simulated~~simulations, ozone profiles over East Asia were similar in shape and magnitude in ~~every season~~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 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 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 (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).

~~To clearly identify different sources of tropospheric ozone in East Asia, we define some terms used in this paper (Table 2). East Asia is the receptor region (Figure 1), and the regions outside East Asia are the source or foreign regions. Ozone in this paper refers to ozone in the troposphere, unless stated otherwise. Tropospheric ozone in East Asia consists of ozone produced in the troposphere from East Asia and foreign regions and ozone transported to the troposphere from the stratosphere. In this paper, we discuss the foreign influence on tropospheric ozone in East Asia from multiple perspectives (Table 2). (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", in this context, generally refers to ozone originally produced in foreign regions and distributed in the domain of East Asia. It also can refer to ozone originally produced in foreign regions and distributed 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". (3) The third is by the sources of ozone precursors, in which the term "anthropogenic ozone" refers to ozone produced from precursors with anthropogenic sources. "Natural ozone" is the sum of ozone produced in the troposphere from precursors with natural sources and ozone from the stratosphere. (4) In the fourth discussion 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 2. For example, North American anthropogenic ozone refers to ozone produced from anthropogenic precursors emitted from North America. The foreign influence is 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.~~

~~3 Seasonality~~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 ~~the seasonal variations of~~ native and foreign ozone over East Asia at the surface (Tables 2-3 and 4Figure 3) and in the East Asian troposphere (Figures 3~~ and 4). The tagged ozone simulation evaluates~~ -5). Figure 3 shows the horizontal variation of the ~~amount~~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 ~~generated from both~~ over East Asia by season. Figure 5 is the same as Figure 4, but for anthropogenic ~~and natural precursors, as well as the amount of~~ ozone~~ from the stratosphere (Figure 3, Table 3), while the emission zero-out simulation further specifies the amounts of~~ .

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 ~~from foreign regions (Figure 4, Table~~ (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 ~~concentration is~~ concentrations are ~22.2 ppbv, which ~~is~~are comparable to that of its native counterpart (~20.4 ppbv) (Table ~~3~~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 ~~3~~2, Figure ~~3~~4). Foreign ozone accounts for over 50% of ozone at the East Asian surface throughout the year, except in summer. This is ~~similar to~~in agreement with the estimate of 50-80% foreign contributions in spring made by Nagashima et al. (2010) and J. Li et al. (2016). ~~Differently, foreign~~Foreign anthropogenic ozone at the East Asian surface is 4.7 ppbv in the annual mean and peaks in spring~~, at~~ (6.4 ppbv, ~~and accounts for~~ 14.1% of ozone~~ (Table 4, Figure 4). This level~~) when it is comparable to ~~that of~~ native anthropogenic ozone ~~in spring, i.e., a~~ (6.9 ppbv~~ concentrations and a~~, 15.1~~% fractional contribution. These results are~~%) (Table 3, Figure 5). Wang et al. (2011) suggested a higher estimate of 12.6±2.3 ppbv in ~~agreement with~~the annual mean foreign anthropogenic ozone in China. Ni et al. (2018~~), who~~) estimated that foreign and native anthropogenic ozone were both approximately 6 ppbv at the surface in China in ~~2008, as determined through a GEOS-Chem simulation. However, Wang et al. (2011) suggested a higher estimate of 14.8±2.2 ppbv foreign anthropogenic ozone in China.~~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 ~~3 and 4, Figures 3 and 4).~~ 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.

~~In~~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 ~~ozone (47.3 ppbv, 72% of ozone) and foreign~~ anthropogenic ozone peaks in spring (9 ppbv~~, 15%) both peak in spring (Figures 3 and 4).~~ or 14% of overall anthropogenic ozone). 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%), while~~ 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. ~~The chemical~~Ozone lifetime ~~of ozone~~ 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 ~~5, the chemical~~ 6, ozone lifetime ~~of ozone~~ at the East Asian surface is longest in winter (~~32~~11.3 days), shortest in summer (~~3.5~~1.1 days~~)~~), and intermediate in spring (~~11~~4 days) and autumn (~~10~~3.8 days). ~~The chemical~~Ozone lifetime ~~of ozone~~at the East Asian surface is approximately ~~10~~9-28 days ~~longer~~shorter than that in the middle troposphere (500 hPa) ~~than in the boundary layer~~ (Figure ~~5). This is expected,~~

~~considering~~ 6), due to the dry deposition and more active chemical reactions of ozone ~~near~~in the ~~surface~~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 ~~6~~7, also see Zhang et al., 2006). Therefore, the combined effect of the ozone lifetime and the westerlies ~~would be~~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 ~~6d~~7d and ~~6h~~7h), both foreign ozone and foreign anthropogenic ozone in the East Asian upper troposphere are at minimum ~~values~~ in winter (Figures ~~3~~4 and ~~4~~5).

Because of the longer lifetime of ozone and the stronger westerly wind in the middle and upper troposphere (Figures ~~5~~6 and ~~6~~7), foreign ozone ~~is~~ concentrations there are 1-2 times higher ~~there~~ than at the surface (Figure ~~3~~4). Figure ~~6~~7 clearly shows that ~~the transport of~~ foreign ozone is transported to East Asia ~~occurs mainly~~mostly through the middle and upper troposphere. It also demonstrates that the seasonality of foreign ozone in different tropospheric layers, particularly ~~in layers~~ near the surface, is greatly impacted by ~~the~~ vertical transport. In ~~East Asia, the~~winter, the East Asian trough located around 130-140°E in the middle troposphere is an important feature of the EAWM system. The downdrafts behind the East Asian trough ~~in the EAWM~~(100°E-140°E) favour the descent of foreign ozone from upper levels to the surface. Winter has the strongest downdrafts, followed by spring (Figure ~~6~~7). Oppositely, in summer, the prevailing ascents in the EASM block foreign ozone from reaching the lower troposphere (~~Figure 6b~~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 ~~6b~~7b and ~~6f~~7f, see also Han et al., 2018). In addition, the downdrafts behind the European trough (~0-40 °E in Figures ~~6b~~7b and ~~6f~~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 ~~3~~4 and ~~4~~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.

~~Vertically, foreign ozone constantly increases with altitude during all seasons (Figures 3a-3d). The concentration of foreign ozone is larger than that of native ozone throughout the troposphere in spring, autumn, and winter and is above ~650 hPa in summer (Figures 3a-3d). In terms of the fractional contributions, foreign ozone (Figures 3e-3h) and foreign anthropogenic ozone (Figures 4e-4h) both peak in the middle or upper troposphere, depending on the season. In contrast, native ozone, either from all sources or from anthropogenic sources, is the largest near the surface and decreases with altitude. These large differences between foreign ozone and foreign anthropogenic ozone in the upper troposphere imply the importance of non-anthropogenic sources at these altitudes (Figure 3 vs. Figure 4, also see Liu et al., 2002; Aghedo et al., 2007).~~

3.2 Foreign ozone over East Asia by source region

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

~~In this section, we further examine the seasonality of foreign ozone~~ 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 ~~by source~~ 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~~. Tables 3 and 4,~~ 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, ~~show foreign ozone and foreign anthropogenic~~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 ~~(in ppbv~~ are higher than those of European ozone over East Asia at layers above 500 hPa, because of larger anthropogenic and natural emissions including 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 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).

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).

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.

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.

~~percentage)~~3.2.2 Foreign ozone at the East Asian surface by source region~~.~~

~~Annually, on~~

Tables 2 and 3 show, respectively foreign ozone and foreign anthropogenic ozone at the East Asian surface by source region. On annual average (Table ~~3~~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 ~~seasonalities~~seasonal variations of North American, European, and African ozone are similar, ~~with all~~i.e., decreasing from winter to summer and then increasing in autumn (Table ~~3~~2), forming a unimodal distribution. South Asian ozone exhibits a unimodal seasonality, as well (Chakraborty et al., 2015).

By ozone precursor, each of the foreign regions contributes less than 3 ppbv of anthropogenic ozone to the East Asian surface ozone ~~during~~in all seasons (Table ~~4~~3). On average, ~~annually,~~ the largest contributing regions are North America (27% of foreign anthropogenic ozone~~) and~~), followed by Europe (21%), ~~followed by~~ 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 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 ~~at,~~ in total, less than 1 ppbv throughout the year. It is noteworthy that ROW ozone generated from anthropogenic ~~sources~~precursors contributes little to the East Asian surface ozone (0.8% of surface ozone, Table ~~4), but~~3), whereas the contribution of ROW ozone from ~~all sources contributes~~both anthropogenic and non-anthropogenic precursors is much more ~~greatly~~ (14.2%, Table ~~3~~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 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 comparable with the simulations in Fiore et al. (2009) and Chakraborty et al. (2015). X. Li et al. ~~(2015), although there exist uncertainties in our results because of the chemical nonlinearity (Huang et al., 2017). The results are also similar to those in X. Li et al. (2014), who~~(2014) estimated that European and South Asian anthropogenic emissions can each contribute 2.4 and 1 ppbv to surface ozone in western China. ~~Holloway et al. (2008) reported that anthropogenic ozone from both North America and Europe ranges between 1-3 ppbv in various regions in East Asia.~~ Ni et al. (2018) suggested that ~~these two regions contribute~~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. ~~Simulations~~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.~~2 Foreign~~3 Latitudinal and longitudinal variations of foreign ozone in the East Asian ~~middle and upper~~ 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).

~~Figure 7 shows the vertical distributions of foreign ozone (in ppbv) in East Asia by region (Figures 7a-7d), along with the fractional contributions to ozone produced in the troposphere (in %) (Figures 7e-7h) and to ozone over East Asia at the corresponding altitudes (Figures 7i-7l). In the East Asian middle and upper troposphere, foreign regions contribute to 65-85% of ozone produced in the troposphere. When foreign ozone peaks at 500 hPa in winter, the ROW is the largest 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%). At 300 hPa, the concentrations of wintertime ozone from the ROW (17.3 ppbv), North America (10.9 ppbv), and Africa (8.0 ppbv) are all higher than those of native ozone (5.6 ppbv). The sum of the ozone from these three regions accounts for 71% of foreign ozone. Excluding the ROW, North America is the region that contributes the most ozone to the East Asian middle and upper troposphere.~~

~~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 during all seasons (Figure 7). European ozone decreases with altitude in the East Asian troposphere in winter, which is opposite to the vertical variations in the other seasons. The peaking season of central Asian ozone is summer in all tropospheric layers in East Asia, and in this season, its vertical maximum occurs in the middle troposphere.~~

~~Stratospheric ozone in the East Asian troposphere increases rapidly with altitude, is largest in winter, and is the second largest in spring (Figure 7). For instance, it rises from below 3 ppbv (5% of ozone) at the surface to 3-30 ppbv in the middle troposphere (5%-30%) and above 30 ppbv (>30%) in the upper troposphere in spring (Figures 7a and 7i). 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 magnitude and 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, at approximately 10-15 ppbv. 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.~~

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

~~Figure 8 shows how the fractional contributions of foreign ozone vary with latitude at the surface and in the middle troposphere. At the East Asian surface (Figure 8a-8d~~At the East Asian surface (Figure 10e-10h), the fractional contribution of foreign ozone~~ along all latitudes~~ 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~~, Figure 8b~~) and then southward to 25°N in autumn (31 ppbv, 74~~%, Figure 8c)~~%) and farther southward to 20°N in winter (28 ppbv, 61~~%, Figure 8d).~~%). 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 ~~concentration~~concentrations of stratospheric ozone at the East Asian surface ~~is~~are largest between 42°N and 46°N in spring (2.4 ppbv) and winter (2.7 ppbv).

~~In nearly all the tropospheric layers (Figure 8e-8h, the cases in the upper troposphere are not shown), South and Southeast Asian ozone is mostly distributed in the regions south of 35°N in East Asia. In contrast, ozone from North America, Europe, and central Asia mostly 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. In contrast, the fractional contributions of 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).~~

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 ~~so-called~~ Tibetan High, or Asian summer monsoon anticyclone~~ or Tibetan High~~), and its position and coverage are shown by the streamlines in Figure ~~9. Figure 9 illustrates that the~~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 ~~9f~~11f) (Vogel et al., 2015), being a reason for the summer maximum of South Asian ozone over the region (Figure ~~7b~~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 ~~Fix-Chem~~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 ~~Fix-Chem~~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 ~~3~~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 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 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}~~-GPH_{500}(30-45 °N, 125-145 °E) (2)

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

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

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

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

$$\delta=\frac{\| \bar{V}_1 - V_i \|}{\| \bar{V} \|}-2 \quad (7)$$

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

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), $\overline{V}_1$ and V_i are the January climatological and monthly wind vectors at a grid and $\overline{V}$ 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 ~~10~~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 ~~10a~~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 ~~10b~~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 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 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.

~~6~~5 Discussion and conclusions

~~In this numerical study using~~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~~, respectively~~ of ozone precursors, using the emission ~~zero-out~~perturbation and tagged ozone simulations~~.~~, respectively. The distributions of foreign ozone in East Asia were characterized in time (seasonally) and space (horizontally and vertically ~~and latitudinally) and time (seasonally~~). 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.

~~The transport of foreign~~Foreign ozone is transported to East Asia ~~occurs~~ mainly through the middle and upper troposphere because of the longer lifetime of ozone and the stronger westerlies at these layers than in the ~~northern hemisphere.~~lower troposphere. In the East Asian middle and upper troposphere (700-200 hPa), the ~~concentration~~annual mean concentrations of foreign ozone ~~is~~are 0.~~5-6~~8-4.8 times higher than that of native ozone, ~~with the former~~ as foreign ozone ranging ~~between~~in 32-65 ppbv (65-85% of ozone produced in the troposphere) and ~~the latter~~native ozone ranging ~~from~~in 11-18 ppbv (Figures ~~3~~4 and ~~7).~~ 9).

At the East Asian surface, the annual mean ~~concentrations of foreign ozone (22.2 ppbv) and~~ of native ozone (20.4 ppbv) ~~are~~is comparable ~~(Table 3).~~ to that of foreign ozone (22.2 ppbv) (Table 2), most of which is transported from North America and Europe~~ are the two major contributing regions of surface ozone in East Asia. Considering only~~. Regarding anthropogenic ozone only, the ~~total~~annual mean of foreign ~~influence~~anthropogenic ozone (4.7 ppbv, 43~~%)~~% of total anthropogenic ozone) is slightly lower than its native counterpart (6.3 ppbv, 57%) ~~in terms of the annual mean~~ (Table ~~4~~3). Three-quarters of~~ the~~ 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 ~~foreign ozone~~ in East Asia, and thus, its seasonality is dominated by the EAM system (Figure ~~6~~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 ~~3~~2).

~~In nearly all tropospheric layers~~In the East Asian troposphere, foreign ozone from North America, Europe, and central Asia generally increases with latitude from 20°N to 60°N~~ in East Asia, while a decrease of~~, whereas foreign ozone ~~with latitude is observed for the regions of~~from South Asia and Southeast Asia ~~(Figure~~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 ~~9~~11).

The interannual variations of foreign ozone at the East Asian surface ~~were~~is found to be closely related to the EAM strength (Figure ~~10). When~~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 ~~reduce the~~enhance outflow of native ozone ~~by enhancing~~ and thus reduce its ~~export. When~~concentrations in East Asia. In summer, if the EASM is strong ~~in summer~~, the weakened south-westerly monsoon wind enhances ~~the~~ native ~~contribution by decreasing~~ ozone ~~export from China~~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 ~~reveals~~revealed the significant foreign influences on tropospheric ozone over East Asia through global atmospheric transport. ~~While~~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 ~~investigated ozone transport~~most focused on the influences from one or a few foreign regions (X. Li et al., 2014; Chakraborty et al., 2015~~), that~~; Zhu et al., 2017; Han et al., 2018), or during one or a few seasons (Ni et al., 2018~~)~~), or ~~only~~on the foreign influence at the East Asian surface ~~ozone in East Asia (~~or boundary layer (Holloway et al., 2008; Wang et al., 2011~~), a comprehensive assessment is provided in~~; Hou et al., 2014). At the East Asia surface, this study~~. Our results are generally consistent~~ compares reasonable with ~~those~~most of the previous studies ~~and appear to be reasonable. For example, we concluded that foreign ozone contributes ~50% of springtime surface ozone in East Asia, and this is in agreement with Nagashima et al. (2010) and J~~, although~~. Li et al. (2016). However,~~ there are some disagreements ~~with previous studies concerning~~ in various details. ~~Upon considering the foreign anthropogenic ozone influence, our estimates for North America and Europe are slightly lower than those of Yoshitomi et al. (2011), probably resulting from differences in the emission inventories and numerical models. The simulated stratospheric influence appears weaker than in some previous studies (Nagashima et al., 2010; Wang et al., 2011), possibly due to the underestimation of ozone in the~~

~~lowermost stratosphere by GEOS-Chem (Jaeglé et al., 2017).~~

We examined the foreign influence on ozone in East Asia throughout ~~all tropospheric columns. Such 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).~~the entire tropospheric column. The simulations show that the ~~concentration~~concentrations of foreign ozone ~~increases~~increase remarkably with altitude and is much higher than its native counterpart in the middle and upper troposphere ~~(Figure 7), implying the significant role that foreign ozone may play in climate change in East Asia.~~. 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).

~~In this~~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 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, 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 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~~),~~) and the INTEX-B Asia emissions in 2006 (Zhang et al., 2009~~), and the NEI05 for North America in 2005. Our simulations in spring agree with those in Ni et al. (2018), who used the global inventory EDGAR v4.2 in 2008.~~). However, the anthropogenic emissions have significantly changed globally in the last decade~~,~~, (Jiang et al., 2018), especially in East Asia~~.~~ (Zheng et al~~. (2018) suggested that because of clear air actions, the anthropogenic emissions in China during~~., 2018). In the EDGAR emission inventories, global NO_x, CO, and NMVOCs from 2000 to 2010~~-2017~~ changed by 9.5%, -1.2%, and 5.2%, respectively and the three species decreased ~~by 17% for NO_x~~across North America and ~~27% for CO and~~Europe, while increased ~~by 11% for NMVOC.~~ in East Asia (Turnock et al., 2018). Moreover, ~~the anthropogenic NO_x and CO emissions in North America are decreasing (Jiang et al., 2018). In addition,~~ there have been some changes in the global natural emissions~~ of ozone precursors~~, such as in biogenic NMVOC emissions (Chen et al., 2018). ~~Therefore, updated emissions inventories can improve future estimates of foreign influences on ozone over East Asia~~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 ~~impact~~impacts on the interannual variation of foreign ozone in East Asia, ~~we specifically underscored~~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~~ further~~ 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

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

Competing interests

The authors declare that they have no conflict of interest.

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