Response to Anonymous Referee #2

This manuscript shows an interesting study on investigating the characteristics of surface ozone variations over South Korea in the past decade. The Kolmogorov-Zurbenko filter (KZ-filter) is used in the time series analysis based on air quality data and meteorology records. The comprehensive analyses present the diversity of spatio-temporal variability over different regions and the impact of meteorological factors. This study explains the variability in detail, which makes it a very valuable study for surface ozone change in South Korea. The text is concisely written. I recommend its final publication after addressing several concerns that have been identified as noted in the suggestions below.

→ We thank the reviewer for the careful reading and valuable and constructive comments. Following the referee's comments, original texts, figures, and tables have been modified. We have performed many additional analyses and provided supplementary figures, which make our study improved and stronger. Additional minor changes will be also included in the revised manuscript as indicated in our response.

Suggestions for addressing certain details necessary for the revised manuscript are also listed below:

1. Before doing meteorological-ozone correlation study, it is necessary to exclude possible effects of emission change on the surface ozone variability. Maybe a few sentence to discuss whether the local emission change is significant during study period.

In addition, you can include more associated study in discussing Asian ozone change due to industrial development. For example, Lei et al., 2012 shows that the future surface ozone change in Asia is more affected by changes in local emissions than that in remote emissions. (Lei, H., Wuebbles,D., Liang, X.-L., Olsen,S., 2012. Domestic versus international contributions on 2050 ozone air quality: how much is convertible by regional control?, Atmospheric Environment, V68, Pages 315-325, doi:10.1016/j.atmosenv.2012.12.002)

→ The NO_x and VOCs emissions from various anthropogenic sources in South Korea are recently assessed for the period of 1999–2010 by KMOE (2013). Figure S1 shows the anthropogenic emissions in South Korea nationwide as well as those in the SMA. It should be noted that the rapid decrease in NO_x emissions between the years 2006 and 2007 mainly results from applying different assessment method to the NO_x emissions from the energy industry before and after 2007. Although the estimated NO_x emissions and its trends for the total period are not so convincing, Fig. S1a shows significant increasing trends of NO_x emissions in the early 2000s. In terms of anthropogenic VOCs emissions, Fig. S1b also shows slight increasing trends in the early 2000s and the trends became flat in recent years. Such increases in precursor emissions must be a major factor to increasing O_3 levels in South Korea, possibly combined with even more rapidly increased precursor emissions in China. Recently, Zhao et al. (2013) have estimated that the NO_x emission in China was about 11 Mt in 1995 but that increased to about 26 Mt in 2010 (in their Fig. 7). Therefore, the elevated O_3 levels in East Asia primarily affected by recent increases in precursor emissions as we addressed in Sect. 3.1.

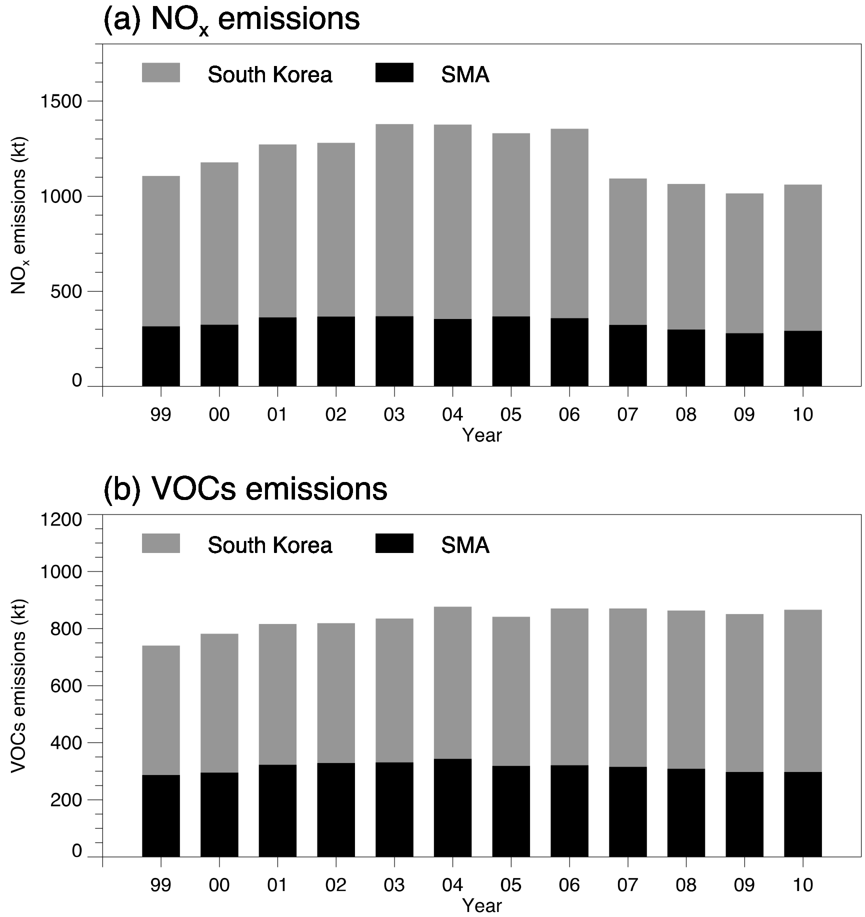

Figure S1. Total anthropogenic emissions of **(a)** NOx and **(b)** VOCs in the SMA and in South Korea (KMOE, 2013).

To emphasize this, we added sentences into line 26 on page 1202 as follows:

Recently, Zhao et al. (2013) have estimated that the NO_x emissions in China increased rapidly from 11.0 Mt in 1995 to 26.1 Mt in 2010, mainly due to the fast growth of energy consumption. The NO_x and VOCs emissions in South Korea also increased in the early 2000s. The estimated anthropogenic NOx and VOCs emissions are 1.10 Mt and 0.74 Mt in 1999 but 1.35 Mt and 0.87 Mt in 2006, respectively (KMOE, 2013).

Since the local precursor emissions are expected to be increased in future emission scenarios, such a local emission effects on the high O_3 levels will be still important in the future. Lei et al. (2013) have shown that the future changes in East Asian precursor emissions will affect O_3 levels in East Asia and will be even important for those in United States. Following the referee's suggestion, we added the following sentence into the introduction, at the last of the second paragraph on page 1193.

Experiments using a global climate-chemistry model with future emission scenarios by Lei et al. (2013) suggest that the increase in East Asian emissions will still be an important issue for the O_3 air quality in both East Asia and United States.

2. The study is based on observational air quality and meteorological data. The raw data may contains large uncertainty due to various reasons including inconsistence, diversity in

measuring method, geographic affects etc. which limits their representativeness in the analysis for a long term trend. I noticed that a quality control strategy may be used. As presented in the manuscript, it seems that only 124 sites out of 290 sites are used in the analysis. What is the quality control criteria? That needs to be clearly introduced.

→ By the year 2010, totally 290 air quality monitoring sites were run by the Korea Ministry of Environment (KMOE) and each local government in South Korea. The air quality monitoring network in 2010 consisted of 236 urban monitoring sites, 33 roadside monitoring sites, 16 suburban monitoring sites, and five background monitoring sites. The hourly data of O_3 and NO_2 measured at the 236 urban monitoring sites were quality-controlled and provided by the National Institute of Environmental Research (NIER). Among the measurement data at the 236 sites, those at 124 sites were chosen to be used in our study due to the continuous measurement for the period of 1999–2010, considering that 108 urban sites were newly installed after the year 2000 and four sites were closed before the year 2010. To make it clear, we revised the manuscript (page 1195, line 24 to page 1196, line 3) as follows:

The National Institute of Environmental Research (NIER) of South Korea provides hourly data of O_3 and NO_2 mixing ratios in the ppbv unit, which have been measured by ultraviolet absorption and chemiluminescence respectively. We here select 124 urban air quality monitoring sites over South Korea, based on data availability for the period 1999–2010, and analyze hourly time series of O_3 and NO_2 from each site. It is noted that our current analysis exclude other data from roadside measurement sites where data can be directly affected by the vehicle exhaust emissions and suburban and background sites located around South Korea.

3. The K-Z filter method is used. It is necessary to add text to clearly explain why the coefficients are selected (e.g. 29, 3, 365, 3), and how their value or uncertainty may affect your conclusion.

In this study, we used $KZ_{29,3}$ to filter the short-term component of O_3 and $KZ_{365,3}$ to extract the long-term component O_3, based on previous KZ-filter studies (Rao and Zurbenko, 1994; Rao et al., 1995; Flaum et al., 1996; Ibarra-Berastegi et al., 2001; Lu and Chang, 2005; Tsakiri and Zurbenko, 2011; Shin et al., 2012). Although $KZ_{365,3}$ is commonly used for the separation of the long-term component, $KZ_{15,5}$ of which effective filter width is 33 days is also used in several studies (Eskridge et al., 1997; Rao et al., 1997; Milanchus et al., 1998; Wise and Comrie, 2005). However, the use of $KZ_{29,3}$ and $KZ_{15,5}$ does not make any significant differences in our results because the signal of which period is less than 50 days are close to white noise, as shown in the power spectra of the log-transformed $O_{3\ 8h}$ and related meteorological variables (SI and T_{max}) in Fig. S2.

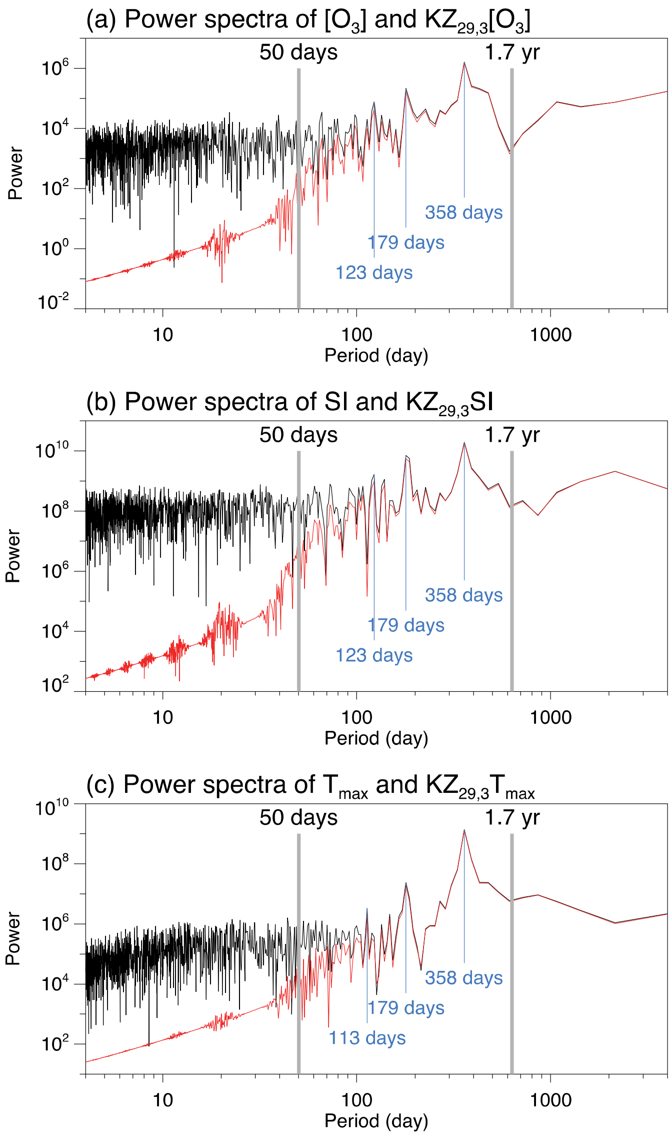

Figure S2. Power spectra of **(a)** log-transformed $O_{3\ 8h}$ time series ($[O_3]$) and its baseline ($KZ_{29,3}[O_3]$) at the City Hall of Seoul, **(b)** daily average surface insolation (SI) and its baseline ($KZ_{29,3}SI$), and **(c)** daily maximum temperature (T_{max}) and its baseline ($KZ_{29,3}T_{max}$) observed at the weather station in Seoul for the period 1999–2000. Each power spectra of original time series and its baseline obtained by $KZ_{29,3}$ filter are represented as black and red lines, respectively.

It is found that both spatial distributions of R^2 between the baselines of $O_{3\ 8h}$ and meteorological variables (Fig. S3) and the probabilities of high short-term component values for each wind direction (Fig. S4) obtained by applying $KZ_{15,5}$ are very similar to Figs. 5 and 11 in the original manuscript obtained by applying $KZ_{29,3}$.

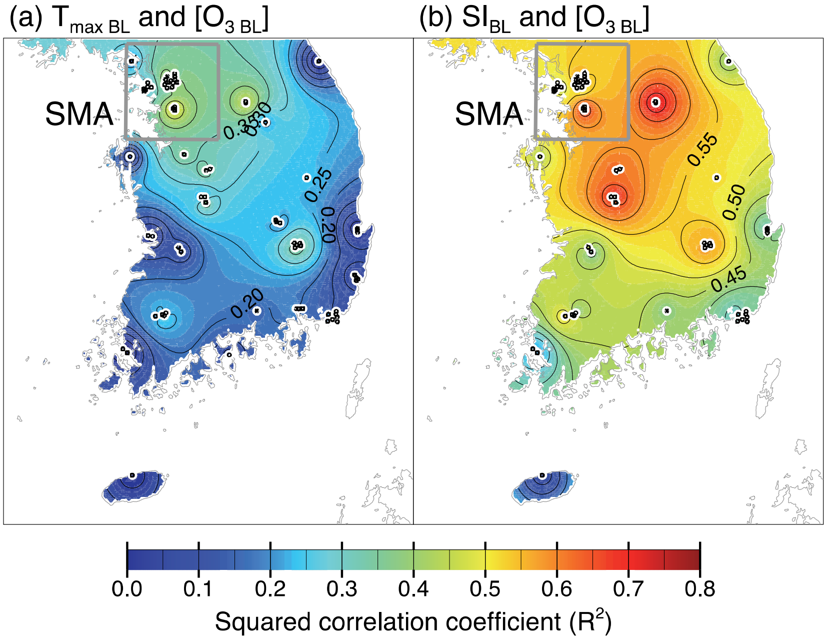

Figure S3. Spatial distributions of coefficients of determination (R^2) between baselines of $O_{3\ 8h}$ ($KZ_{15,5}[O_3]$) and **(a)** daily maximum temperature ($KZ_{15,5}T_{max}$) and **(b)** surface insolation ($KZ_{15,5}SI$). Each baseline is obtained by applying $KZ_{15,5}$ filter.

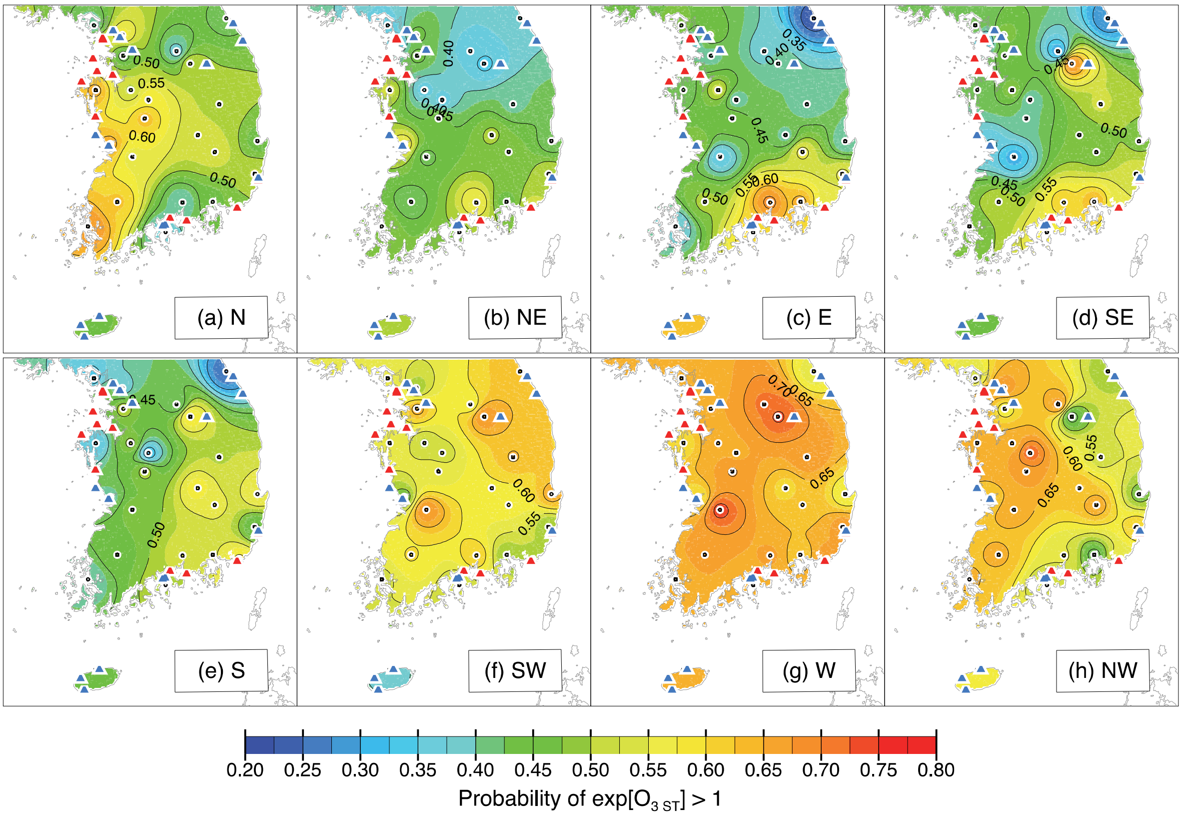

Figure S4. Spatial distribution of probabilities that exponentials of the short-term components will exceed 1 (exp[$O_{3\ ST}$]>1) for each wind direction (WD). Each short-term component is obtained by applying the $KZ_{15,5}$ filter.

To make it clear that we used the $KZ_{29,3}$ following previous studies, lines 4-5 on page 1198 is modified in the revised manuscript as follows:

In this study, we used the KZ-filter with the window length of 29 days and 3 iterations ($KZ_{29,3}$) following previous studies (e.g., Rao and Zurbenko, 1994) and decomposed daily *ln*($O_{3\ 8h}$) time series at the 124 monitoring sites.

4. It is also necessary to discuss the limitations on the K-Z filter method and resulted uncertainty in the result. For example, if considering the precipitation change as a driver for ozone variation, it is noted that precipitation has large seasonal and annual variability. Therefore, the statistic method filtered short-term component may contain contributions that were originally caused by seasonal precipitation system. It is better to discuss the quality of the analysis in the text.

→ In the present study, precipitation was not used as a predictor in the multiple linear regression models because of its discontinuity and sporadic nature in the time series. Instead, baselines of insolation, relative humidity, and dew-point temperature reflect the possible effects of rainfall on the O_3 variation in the seasonal time scale. For example, the seasonal component of $O_{3\ 8h}$ represents well the effects of seasonal precipitation system in July and August (Fig. 2c in the original manuscript). On the other hand, the effects of sporadic rainfall events on the O_3 short-term variation in other seasons are well filtered by the $KZ_{29,3}$, although we did not analyze their relationship in this study. Therefore, the use of KZ-filter can be an advantage rather than a limitation in view of the referee.

5. I suggest plotting fig 2(d) with a boarder value range and stating that the long term surface ozone change is not distinct in the text. In fact, previous studies on observed surface ozone change on other parts of world show that there is no clear increasing trend for surface ozone. Considering the biases in measurements, the significance of result need to be discussed.

→ We agree with the referee's point. However, since Fig. 2 is just an example for decomposition of O_3 time series into each component, we would prefer not to modify range of the figure but to add a sentence to note the small variance of long-term component O_3 in the last paragraph of Sect. 2.2 (page 1200, line 3) as follows:

It should be noted that [$O_{3\ LT}$] explains only 1.7% of the total variance of [O_3] at this site as its small ranges in Fig. 2d, while relative contributions of [$O_{3\ ST}$] and [$O_{3\ SEASON}$] are 58.3% and 32.7%, respectively. Therefore, the long-term changes in O_3 related to changes in local emission occupy only small fraction of the O_3 variations. The relative contributions of each component are further examined in Sect. 3.4.

As the referee pointed out, previous studies have reported that the temporal trends of O_3 in Europe, North Atlantic, North America, and Japan do not show significant changes in recent decades (e.g. Vingarzan, 2004; Oltmans et al., 2006; Oltmans et al., 2013). However, O_3 levels in China have significantly increased in recent years mainly due to the rapid economic growth and energy consumption (Ding et al., 2008; Tang et al., 2009; Wang et al., 2009; Wang et al., 2012). Such increasing trends of O_3 are also significant in South Korea (KMOE, 2012). We modified the manuscript (page 1193, lines 15–20) to introduce current O_3 trends in the mid-latitude Northern Hemisphere as follows:

In the Northern Hemisphere mid-latitudes where the most of the population, industry, and transport are concentrated on, the tropospheric O_3 increased during the late 20th century due to increases in anthropogenic precursors particularly nitrogen oxides (NO_x), but its trends show regional and temporal differences (Oltmans et al., 1998; Guicherit and Roemer, 2000; Vingarzan, 2004). Although the increasing trends of O_3 in Europe, North Atlantic, North America, and Japan have flattened over the past decade (Oltmans et al., 2006; Oltmans et al., 2013), there have still been concerns about elevated O_3 concentration in China owing to rapid economic growth and industrialization (Ding et al., 2008; Tang et al., 2009; Wang et al., 2009; Wang et al., 2012). Such recent increases of O_3 in China can affect the regional background O_3 levels in East Asia by transboundary transport of O_3 and its precursors.

6. In the section 3.2, daily minimum ozone in the coastal cities may not be good as an indicator for background ozone effect. Usually, the minimum surface ozone concentration occurs during nighttime. The change of minimum ozone is more affected by multiple factors, mainly including nighttime NO_y chemistry, temperature, nighttime surface mixing etc. High temperature accelerates the removal of ozone by NO_y chemistry, and thus associated with lower ozone concentration. There would have other environment factors and chemicals in affecting NO_y chemistry (e.g. Lei, H. and Wuebbles, D., Chemical Competition in Nitrate and Sulfate Formations and its effect on Air Quality, Atmos. Environ., 80, 472-477, doi:10.1016/j.atmosenv.2013.08.036.2013.), which indirectly affect the change of minimum ozone.

In addition, using wind speed at the time of minimum ozone occurs in the analysis can be meaningful. From figure 7, it seems that inland sites have higher wind speeds, while coastal sites have lower speeds. Low and High ozone concentrations occur on each region. This is more of a local characteristic. In addition, the minimum ozone usually occurs in nighttime. Therefore, there is no photochemistry as described in the manuscript. The description in this part may not stand. This part should be reanalyzed.

→ We concur with the referee's points that the daily minimum O_3 ($O_{3\ min}$) levels are also affected by multiple factors such as nocturnal NO_y chemical process related to nitrate formation or surface mixing, and therefore, $O_{3\ min}$ might be not a good indicator for background O_3 effects. In the polluted urban area, O_3 is removed by both NO_x titration ($NO+O_3 \rightarrow NO_2+O_2$) and nitrate formation ($NO_2+O_3 \rightarrow NO_3+O_2$) at nighttime. However, NO_x concentrations in coastal cities are lower than those in other inland cities due to surface mixing and ventilation by high wind speeds. Therefore, the less NO_x titration and nitrate formation induce the higher levels of $O_{3\ min}$ at nighttime in the coastal regions. In the revised manuscript, we modified manuscript (page 1205, line 8–18) to address the effects of nocturnal NO_y chemistry on the high $O_{3\ min}$ as follows:

As represented in Fig. 6a and Table 3, the $O_{3\ min}$ is high near the coast, low at the inland cities, and lowest in the SMA. In the polluted urban area, the O_3 concentration reaches near-zero minima during the night since O_3 is reduced by NO_x titration, nocturnal NO_y chemical process related to nitrate formation, and dry deposition in the absence of photochemical production. In the coastal region, however, the NO_x concentrations are low (Fig. 3b), and thus the less titration and nitrate formation at nighttime induce the higher $O_{3\ min}$ levels. In addition, transport of O_3 from the regional background could also keep high levels of O_3 during the night (Ghim and Chang, 2000). Frequency distributions of O_3 concentrations in previous studies suggested that O_3 levels at the coastal cities such as Gangneung, Jeju, Mokpo, Seosan, and Yeosu are affected by the background O_3 transport, unlike Seoul where the effect of local precursor emission is dominant (Ghim and Chang, 2000; Ghim, 2000). Therefore, combined effects of the low NO_x levels and transport of the regional background O_3 influence the high $O_{3\ min}$ near the coast.

The lines 21-23 on the same page are modified as follows:

These opposite patterns suggest that the meteorological effects on the O_3 production are negatively correlated with $O_{3\ min}$ for the South Korean cities.

The lines 28-29 on the same page are modified as follows:

Larger NO_x emissions in these southeastern coastal cities (Fig. 3b) induce lower $O_{3\ min}$ levels via NO_x titration and nocturnal NO_y chemical process.

7. In the section 3.4, the site of Gangneung locates on the back of mountain. The ozone change there would be more associated with the topography. I would suggest you to discuss these factors that have more sensitive influence to local surface ozone change.

→ As the referee pointed out, the topographical characteristics of Gangneung are an important factor for the high relative contribution of the short-term component. High and steep mountains on the west and sea on the east represent topographical characteristics of Gangneung. In the short-term time scale, wind direction changes by eastward moving synoptic weather systems, together with such topography, highly affect the short-term variation of O_3 in the eastern coastal region including Gangneung. In the prevailing westerly winds, orographic descent induces often warm, dry, and strong winds, which are favorable to the clear sky and strong vertical mixing over the region. Since the westerly winds contain the precursors emitted from the SMA and inland regions, the clear sky condition increases the O_3 levels during the daytime. In addition, the strong vertical mixing of high O_3 air from the upper troposphere compensates the O_3 loss by titration during the nighttime. On the other hand, in the easterly winds, orographic lift often forms fogs or clouds over the region and reduces the photochemical production of O_3. These short-term O_3 changes by the wind directions at Gangneung are also well represented in Fig. 11 of the original manuscript. We have modified lines 24–27 on page 1207 to add aforementioned information as follows:

Since [$O_{3\ ST}$] is related to synoptic-scale weather fluctuation (Rao et al., 1995; Rao et al., 1997), the large relative contributions of [$O_{3\ ST}$] at the coastal cities indicate the stronger effects of the eastward moving synoptic weather systems in there. Interestingly, the highest value of [$O_{3\ ST}$] contribution is appeared at a northeastern coastal city, Gangneung. High and steep mountains on the west of Gangneung induce often warm, dry, and strong westerly winds, which is favorable to the clear sky and strong vertical mixing over the region. Since the westerly winds contain the precursors emitted from the SMA, the clear sky condition increases the O_3 levels during the daytime. In addition, the strong vertical mixing of high O_3 air from the upper troposphere compensates the O_3 loss by titration during the nighttime. In the easterly winds, however, orographic lift often forms fogs or clouds over the region and reduces the photochemical production of O_3. Therefore, combined effects of wind directions related to synoptic weather systems and topography increase the short-term variability of O_3 at Gangneung.

Two other inland cities of Andong and Wonju also show topographic effects on the seasonal variability of O_3. Therefore we added a sentence in line 2 on page 1208 as follows:

The highest and second highest values of [$O_{3\ SEASON}$] contribution are appeared at Andong and Wonju located in the inland basin. Since the basin topography often traps pollutants and induces large annual ranges of temperature, seasonal variability of O_3 at two cities is larger than that of other inland cities.

8. It is very interesting to have the wind direction analysis in section 3.5. However, I have a general question on this part. Does some of the wind directions (e.g. easterly wind, northeasterly wind.) are much less occurring than westerly wind? If so, the discussion in the text should include this aspect.

→ Figure S5 shows the number of days in each wind direction during the analysis months of frequent high O_3 events (May–October), for the period 1999–2010. During the months, westerly and southwesterly winds are predominant as addressed in the last paragraph of the Sect. 3.5, while northerly, easterly, and southerly winds are much less occurring. In comparison with Figs. 11 and 12, Fig. 5S shows more local characteristics of prevailing wind directions in each region, except the predominant westerly wind. This implies that the high probability of high O_3 episodes in each wind direction is affected not by the local prevailing winds, but by relative directions of the local or remote emission sources (e.g. high probabilities in the western South Korea in northerly or the southeastern region in westerly). We think that this aspect is already contained in the text.

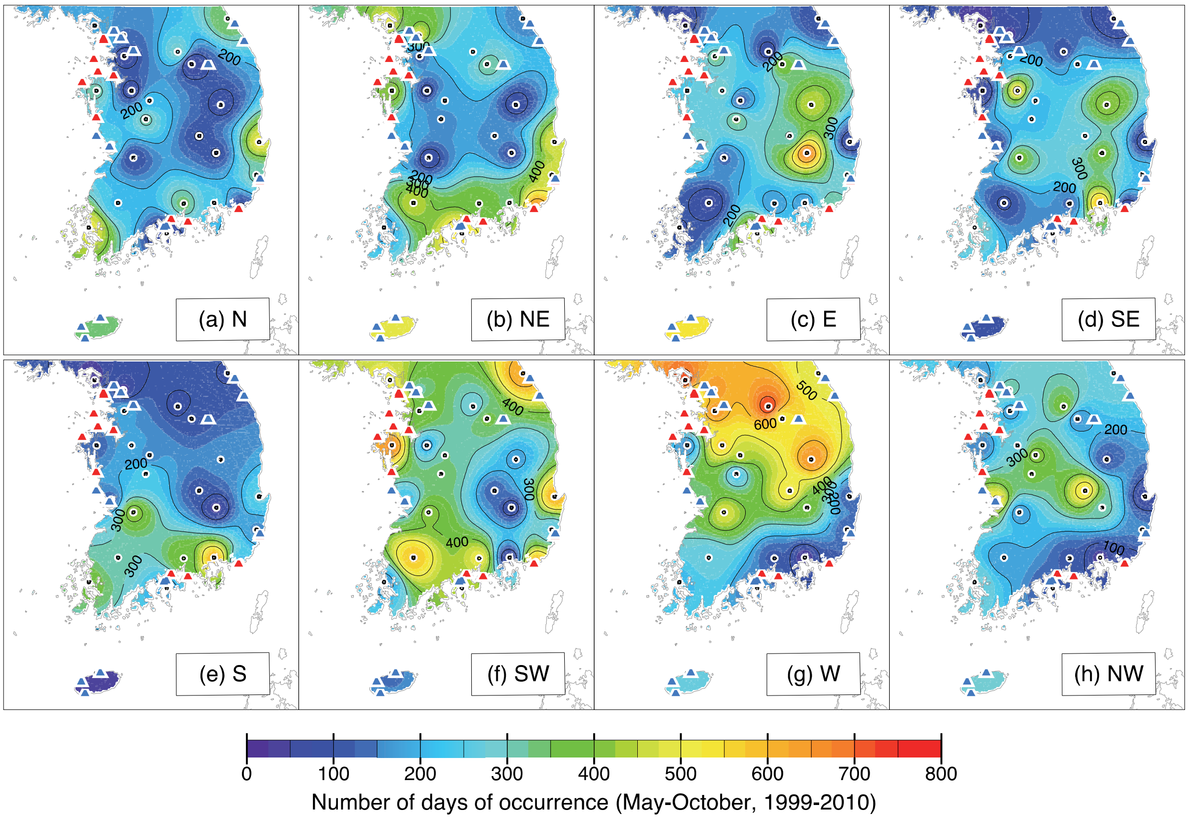

Figure S5. Spatial distributions of number of days in each wind direction (WD) during the months of frequent high O_3 events (May–October) for the period 1999-2010.

9. In the SVD analysis on 3.6, the long term ozone concentration has been filtered out a lot of information. It is clear that the short term variability over Korea takes more information for surface ozone change in Korea. Therefore, it is necessary to discuss this in the text. In particularly, I am very interested in the results by using the short term or seasonal ozone information in SVD.

→ We agree with the referee's points that the short-term and seasonal components take most of the information for the O_3 changes. Figure S6 shows the first mode of SVD expansion coefficients and their time series between the seasonal components of O_3 and NO_2. The first modes account for 94.3% of the total squared covariance and their time series clearly show typical seasonal characteristics of O_3 and NO_2 in South Korea. During the winter, reduced vertical mixing often traps the pollutants near surface and increases NO_2 levels while weak photochemical production with strong titration effects decreases O_3 levels. In the spring, on the other hand, O_3 levels are raised by photochemical reactions of accumulated precursors along with other factors such as transport by continental outflow or stratospheric intrusion. Due to wet scavenging of the pollutants, NO_2 levels show their annual minimum during the East Asian summer monsoon. In addition, reduced insolation by frequent precipitation together with relatively clean air decreases photochemical production of O_3 in summer.
The SVD with short-term components of O_3 and NO_2 is a bit complicated. Figures S7 and S8 shows the first and second leading modes of SVD and their time series for the short-term components of O_3 and NO_2. The squared covariance fraction of the first and second mode is 16.3% and 13.1%, respectively. As represented by the small squared covariance fractions of

each mode, the spatial patterns of O_3 and NO_2 short-term components are weakly coupled. However, the same signs of spatial patterns in many regions imply that the short-term variations of O_3 have some degree of relationship with the short-term variations of NO_2.
Despite some interesting features of SVD analysis with short-term and seasonal components, we did not include Figs. S6–S8 in the revised manuscript because we focused on the long-term variations of O_3 and NO_2 in Sect. 3.6.

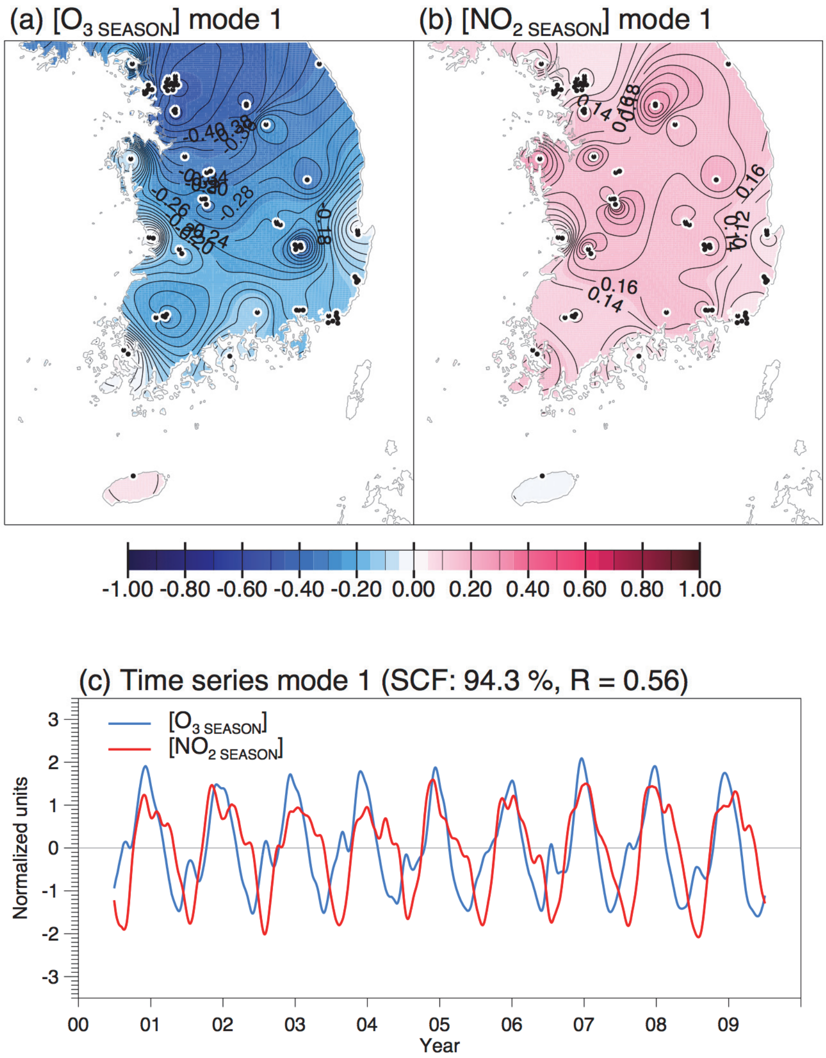

Figure S6. The first leading mode of SVD between the seasonal components of **(a)** daily maximum 8-h average O_3 ([$O_{3\ \mathrm{SEASON}}$]) and **(b)** daily average NO_2 ([$NO_{2\ \mathrm{SEASON}}$]) with **(c)** time series of the SVD expansion coefficient associated with [$O_{3\ \mathrm{SEASON}}$] mode (blue line) and [$NO_{2\ \mathrm{SEASON}}$] mode (red line).

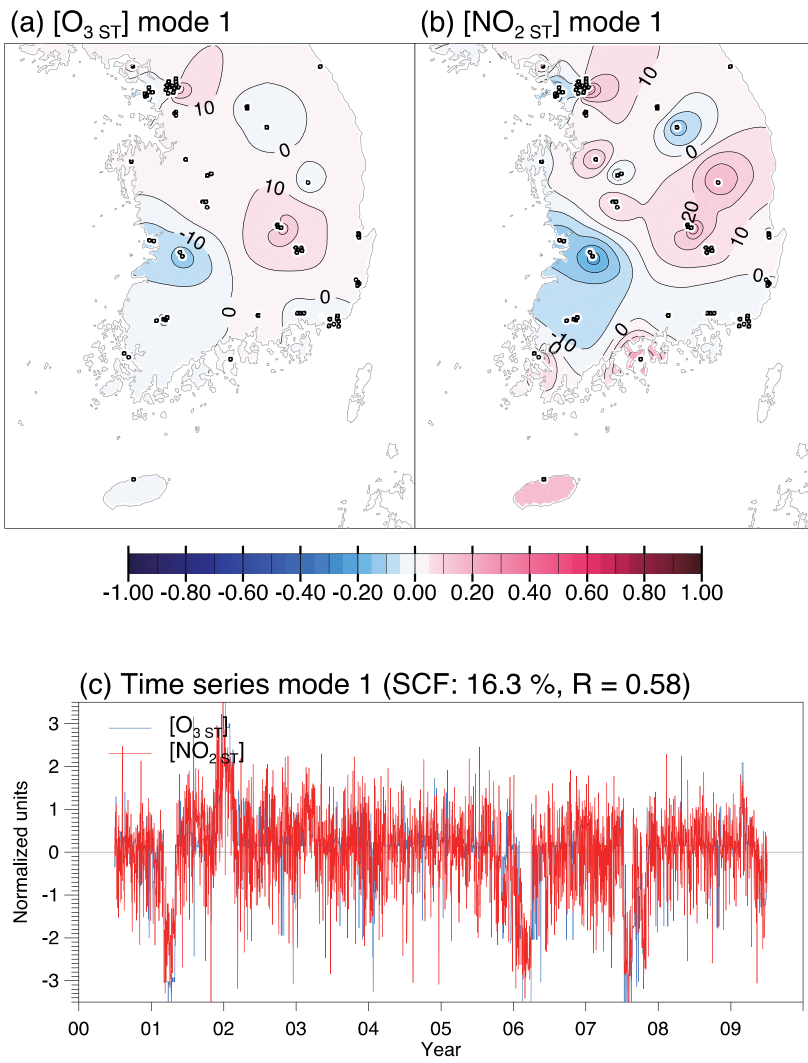

Figure S7. The first leading mode of SVD between the short-term components of **(a)** daily maximum 8-h average O_3 ([$O_{3\ ST}$]) and **(b)** daily average NO_2 ([$NO_{2\ ST}$]) with **(c)** time series of the SVD expansion coefficient associated with [$O_{3\ ST}$] mode (blue line) and [$NO_{2\ ST}$] mode (red line).

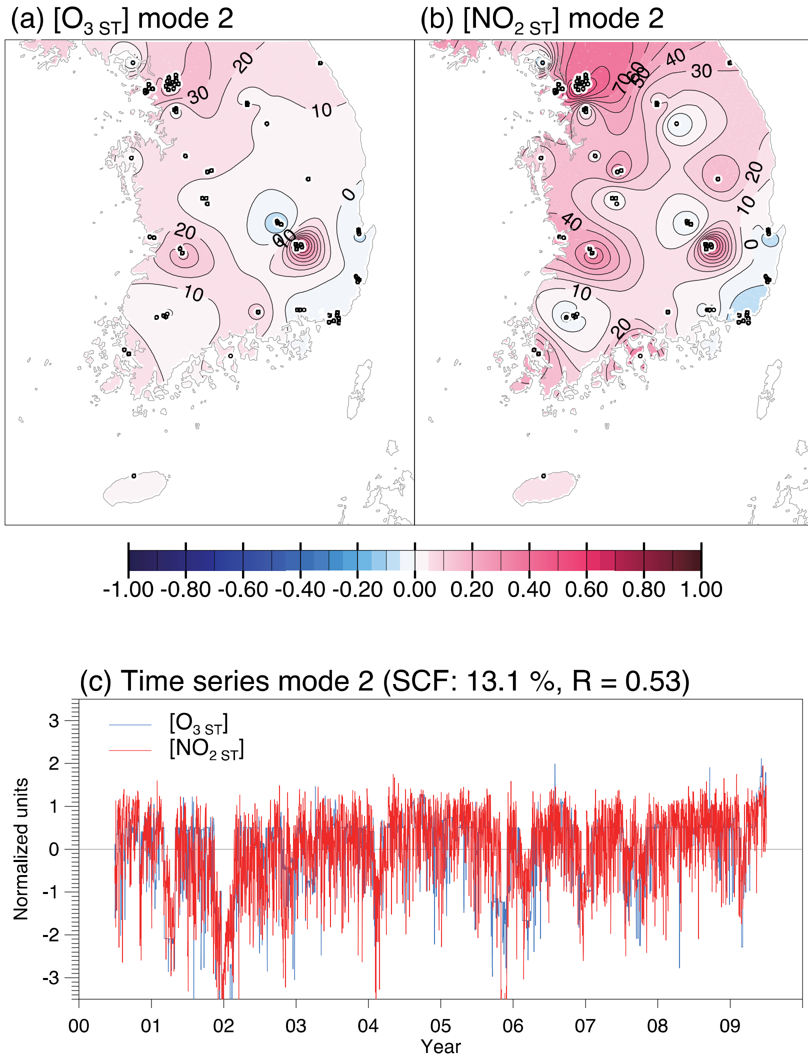

Figure S8. The second mode of SVD between the short-term components of **(a)** daily maximum 8-h average O_3 ([$O_{3\,ST}$]) and **(b)** daily average NO_2 ([$NO_{2\,ST}$]) with **(c)** time series of the SVD expansion coefficient associated with [$O_{3\,ST}$] mode (blue line) and [$NO_{2\,ST}$] mode (red line).

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