Response to Anonymous Referee #1

This manuscript presents a thorough analysis of the spatial characteristics and temporal changes of surface ozone (O_3) over South Korea during the period 1999-2010. The KZ filter (used to decompose time series into short-term, seasonal and long-term components) is combined with linear regressions to examine the relationship between meteorological variables and ozone over different areas. The authors conclude that baseline temperature and insolation are relevant for the baseline ozone levels inland and in the Seoul Metropolitan Area (SMA), while the transport of regional background air masses impact on ozone concentrations on the coast. Their analyses of the probability of O_3 exceedances as a function of temperature or of the relationship between the exponential of the short-term component of O_3 with wind speed are very interesting. They also use singular value decomposition (SVD) to assess the possible impact of changes in NO_x on ozone concentrations. The authors are fair, they give credit to previous work and recognise most of the limitations of their analyses. This valuable study will contribute to improving our understanding of recent surface ozone changes and will also be useful for future projections.

I do not have any major objections to the scientific content, methods used or conclusions. I only have some comments and suggestions to clarify a few points (see section "Specific comments"). However the use of English and style should be improved. I have provided an annotated version of the paper to assist the authors with this, but I expect that somebody with good written English skills revises the language very carefully before the final submission. I will fully support the publication in *Atmos. Chem. Phys.* once this latter issue has been addressed by the authors.

→ We appreciate the reviewer for the valuable and constructive comments along with kind English corrections. As indicated in the following point-by-point responses, we have incorporated the reviewer's comments into our revised manuscript.
We have performed many additional analyses and provided supplementary figures, which make our study improved and stronger. New references are included and thus texts, figures, and tables are modified.

Specific comments

1. As indicated above, the authors give credit to previous work. However, further discussion about recent changes in tropospheric ozone over the Northern hemisphere is needed. Both in the introduction and conclusions the authors mention the recent increase in O_3 levels over the Northern Hemisphere and East Asia. This may be the general case for most of East Asia, but the picture for Europe or North America might not be so clear, in particular over the last decade. The authors cite papers such as Vingarzan et al. (2004). This paper already indicates that trends were not uniform in the years preceding 2004, although both they and more recent publications suggest that background ozone levels over the Northern mid-latitudes have continued to rise. Some authors (e.g. see below some papers by Samuel Oltmans) point to significant regional differences and to the flattening of O_3 levels in the Northern mid-latitudes over the last years. It is also known that ozone trends derived from different platforms are not always consistent with each other (see some relevant literature e.g. in the introduction of Saunouis et al., 2012), which might also be responsible for some of the regional differences. To conclude, I think the authors should include a short sentence (and cite one or two relevant

publications) to indicate that there is no clear consensus on the increase of ozone in the North hemisphere over the last decade.

→ We agree with the reviewer's comments. The background O_3 changes over the Northern Hemisphere are reported to have significant regional and temporal differences (e.g. Vingarzan, 2004; Oltmans et al., 2006; Oltmans et al., 2013). The temporal trends of O_3 in Europe, North Atlantic, North America, and even Japan do not show significant changes in recent decade while those in China are still increasing (e.g. Ding et al., 2008; Tang et al., 2009; Wang et al., 2009; Wang et al., 2012). We have added a description of the current O_3 trends into our revised manuscript and modified the text as follows:

In the Northern Hemisphere mid-latitudes where population, industry, and transportation are concentrated, the background O_3 levels 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.

2. Section 2.1. The authors mention that observations of O_3 and NO_2 are available at 290 sites while they use 124 of them. Is that selection based on data availability? Please provide details on the selection criteria.

→ 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 authors use $KZ_{29,3}$ to filter the short-term component (period smaller than 50 days). Then they do a meteorological adjustment of the baseline ozone concentrations and finally apply $KZ_{365,3}$ to extract the information for periods larger than around 1.7 yr. This looks

reasonable, but I wonder myself how sensitive results can be to the choice of the window length (m) and iterating times (p) used in the KZ filter. Could the authors explain how/why they have chosen those specific values of m and p? Were they looking for the mentioned periodicities (around 50 days and 1.7 yr)? Is this based on previous work? Or has this been done following trial and error?

→ We have used $KZ_{29,3}$ to filter out the short-term component of O_3 and $KZ_{365,3}$ to extract the long-term component of 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). In terms of filtering the short-term component, $KZ_{15,5}$ of which effective filter width is 33 days is also previously used in several studies (Eskridge et al., 1997; Rao et al., 1997; Milanchus et al., 1998; Wise and Comrie, 2005), although $KZ_{365,3}$ is commonly used for separation of the long-term component.
We further investigated the KZ filter method by using the power spectrum analysis. Figure S1 shows the power spectra of the log-transformed $O_{3\ 8h}$ and related meteorological variables (SI and T_{max}) with their baselines, which are filtered out by $KZ_{29,3}$. In the figure, the highest and second highest peaks of power spectral density appear at the period of 1 yr and 0.5 yr due to their periodic seasonal variations. On the other hand, the power spectra of which period is less than 50 days are relatively flat, similar to the white noise. Those high-frequency variations (short-term components) are well removed by $KZ_{29,3}$. In addition, the effective filter width of $KZ_{365,3}$ (1.7 yr) separates the seasonal periodicity and long-term variations of O_3 and meteorological variables.
It is found that both spatial distributions of R^2 between the baselines of $O_{3\ 8h}$ and meteorological variables (Fig. S2) and probabilities of high short-term component values ($\exp[O_{3\ ST}] > 1$) in each wind direction (Fig. S3) obtained by applying $KZ_{15,5}$ are very similar to Figs. 5 and 11 in the original manuscript obtained by applying $KZ_{29,3}$. Although the effective filter width of $KZ_{15,5}$ (33 days) is shorter than those of $KZ_{29,3}$ (50 days), statistical characteristics of the short-term and baseline components obtained by applying $KZ_{15,5}$ and $KZ_{29,3}$ are not much different. Therefore, $KZ_{29,3}$ is reasonably chosen to filter the short-term variation.

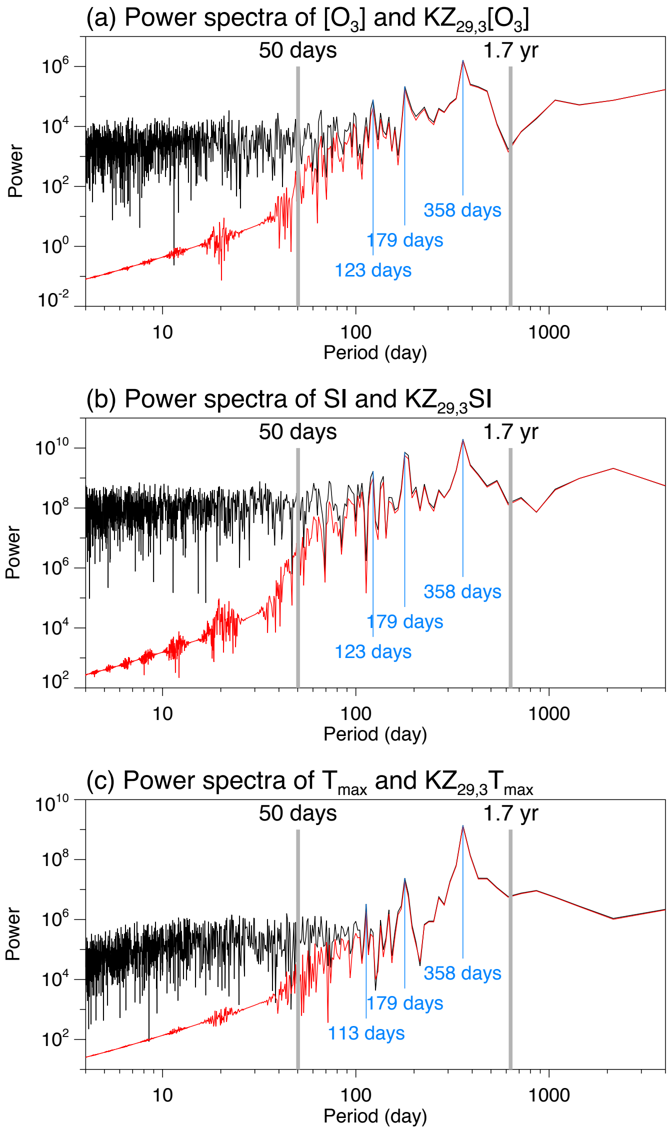

Figure S1. Power spectra of **(a)** log-transformed $O_{3\ 8h}$ time series ($[O_3]$) and its baseline ($KZ_{29,3}[O_3]$) at the City Hall of Seoul, **(b)** daily average surface insolation (SI) and its baseline ($KZ_{29,3}$SI), and **(c)** daily maximum temperature (T_{max}) and its baseline ($KZ_{29,3}T_{max}$) 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.

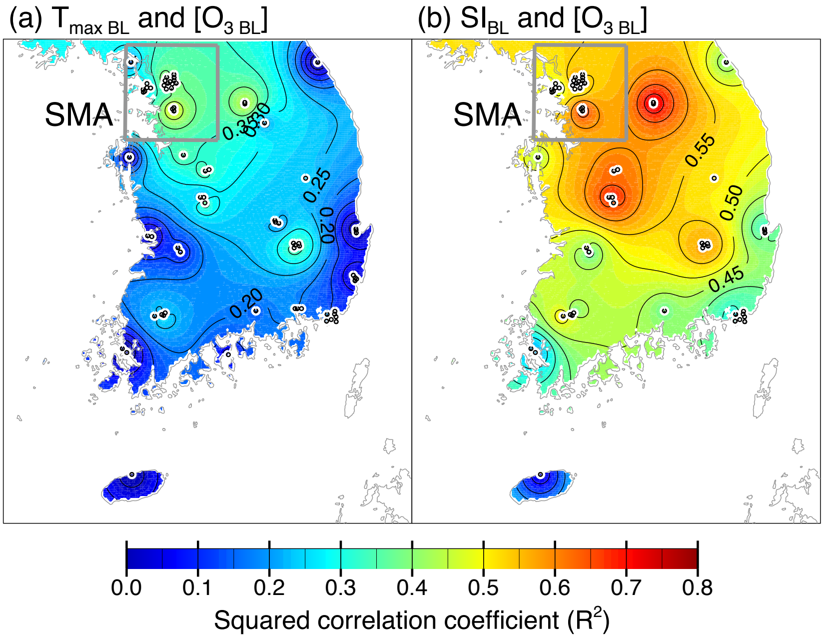

Figure S2. Spatial distributions of coefficients of determination (R^2) between baselines of $O_{3\ 8h}$ ($KZ_{15,5}[O_3]$) and **(a)** daily maximum temperature ($KZ_{15,5}T_{max}$) and **(b)** surface insolation ($KZ_{15,5}SI$). Each baseline is obtained by applying $KZ_{15,5}$ filter.

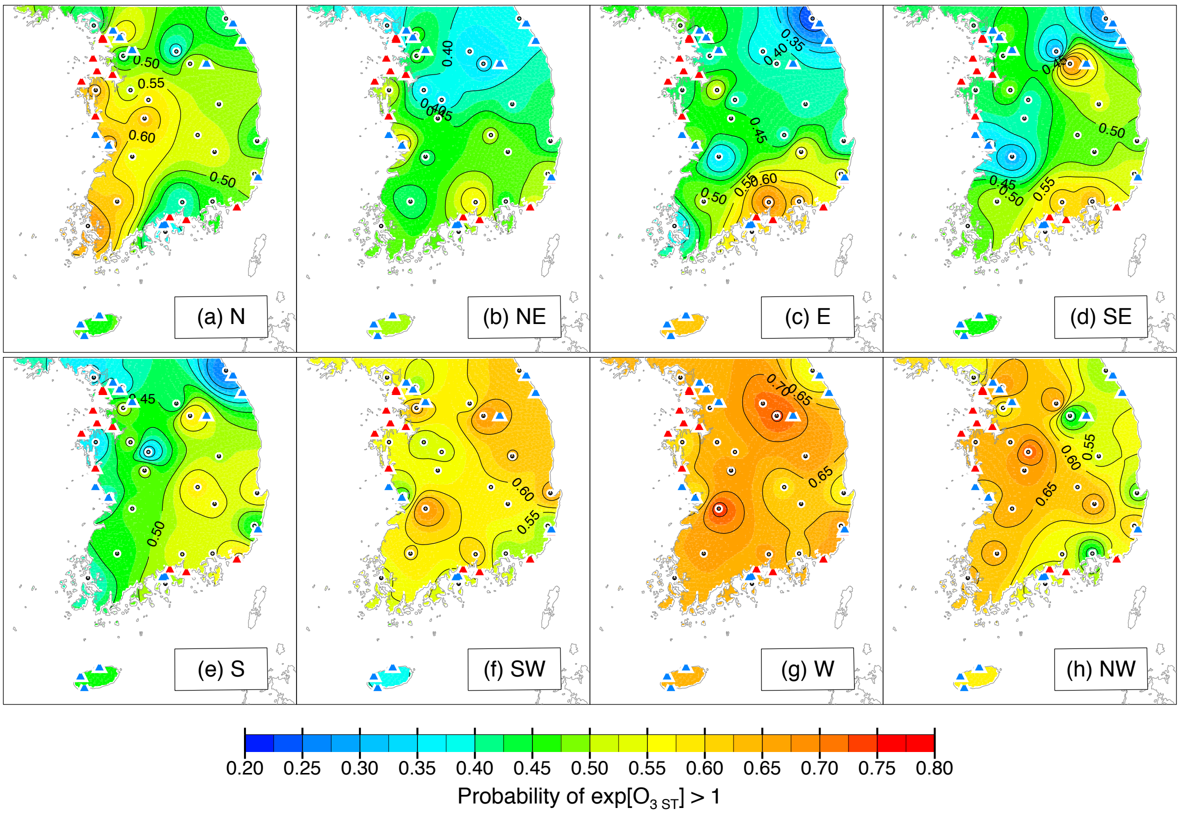

Figure S3. Spatial distribution of probabilities that exponentials of the short-term components will exceed 1 ($\exp[O_{3\ ST}]>1$) for each wind direction (WD). Each short-term component is obtained by applying the $KZ_{15,5}$ filter.

4. I understand that the residual "delta(t)" in equations (6) and (7) is not part of the long-term component and that it is the part of the seasonal component which cannot be explained by the meteorological regression model. Is this right? If so, shouldn't the authors test that the statistical characteristics of that residual are similar to that of white noise (e.g. normality, no autocorrelation, homoscedasticity)?

→ As in Eq. (5), the residual term $\varepsilon(t)$ is the difference between the baseline ($[O_{3\ BL}]$) and combined meteorological variables regressed on $[O_{3\ BL}]$ ($a_0 + \sum_i a_i\ MET_{BL}(t)_i$), and thus it contains both long-term and seasonal variability of O_3 (Figs. S4a and b). Their long-term variability is removed by $KZ_{365,3}$ as in Eq. (6) and the remaining $\delta(t)$ has some degree of seasonal variability related to the unconsidered meteorological variables in the multiple linear regression models (Figs. S4b and c). Therefore, the statistical characteristics of $\delta(t)$ are somewhat far from those of white noise as shown in its power spectrum (Fig. S4d). Compared to autocorrelation of the short-term component ($[O_{3\ ST}]$) denoted by red solid line in Fig. S4e whose statistical characteristics are close to those of white noise, that of $\delta(t)$ by black solid line shows clear periodicity.

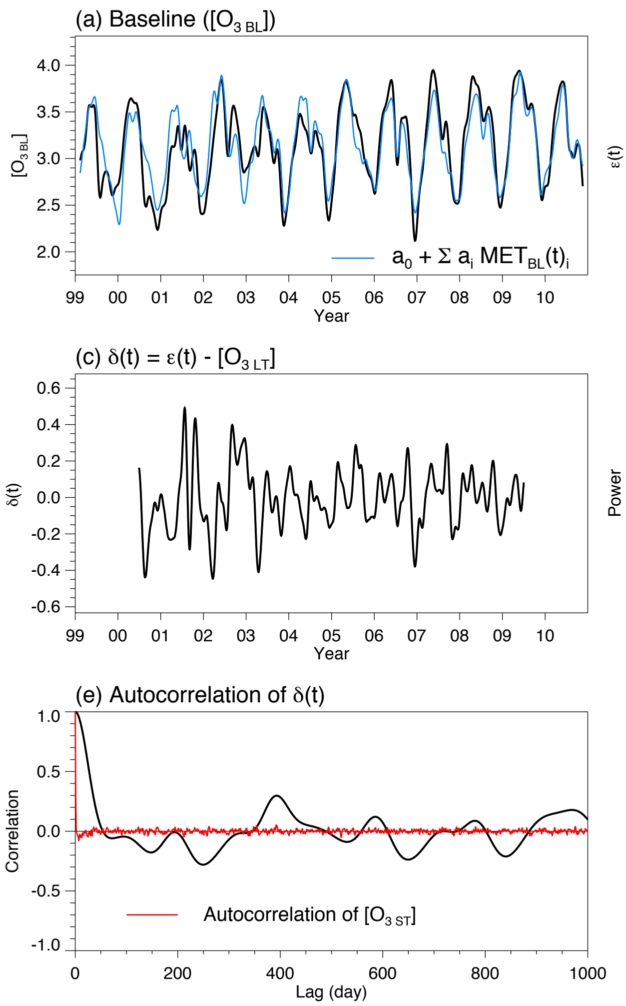

Figure S4. Time series of **(a)** baseline of log-transformed $O_{3\ 8h}$ ($[O_{3\ BL}]$) and combined meteorological variables regressed on the baseline ($a_0 + \sum_i a_i\ MET_{BL}(t)_i$), **(b)** $\varepsilon(t)$ and long-term component ($[O_{3\ LT}]$), and **(c)** $\delta(t)$ at the City Hall of Seoul. **(d)** Power spectrum of $\delta(t)$. **(e)** Autocorrelation of $\delta(t)$ and short-term component ($[O_{3\ ST}]$).

5. Section 3.2: The authors indicate that "The nationwide average of R-squared is 0.50 for surface insolation (SI), 0.29 for PS, 0.22 for T_{max}, 0.14 for TD, 0.05 for RH, and 0.03 for WS, respectively". R-squared is basically the variance explained by each variable. I think it would be very relevant to also know how much of the variance they are able to explain with all meteorological variables combined. Considering that they have used a multiple linear regression model of baseline ozone on the baseline of those meteorological variables (see eq. 5 or 7), why don't they also indicate the value of R-squared for that model?

→ Following the referee's suggestion, we have calculated the adjusted R^2, with the consideration of the number of parameters and degrees of freedom, between the baseline of $O_{3\ 8h}$ ($[O_{3\ BL}]$) and the multiple linear regression model ($a_0 + \sum_i a_i\ MET_{BL}(t)_i$). The adjusted R^2 values and their spatial distribution are represented in Table S1 and Fig. S5. As shown in Table S1, the multiple linear regression models with six meteorological variables explain 51% of the total variance of $[O_{3\ BL}]$ nationwide, but the averaged values of adjusted R^2 are much higher in the inland region (0.61) or SMA (0.62) than in the coastal region (0.37).

Table S1. Adjusted coefficients of determination (adjusted R^2) between baselines of $O_{3\ 8h}$ ($[O_{3\ BL}]$) and multiple linear regression models ($a_0 + \sum_i a_i\ MET_{BL}(t)_i$) at 25 cities over South Korea for the period 1999–2010. The cities are categorized into three groups: 10 coastal cities, 11 inland cities, and 4 cities in the Seoul Metropolitan Area (SMA). Numbers in bold fonts indicate correlations significant at the 95% level or higher.

Coastal region			Inland region			SMA		
Cities	City code	Adjusted R^2	Cities	City code	Adjusted R^2	Cities	City code	Adjusted R^2
Busan*	BS	**0.408**	Andong	AD	**0.628**	Ganghwa	GH	**0.389**
Changwon	CW	**0.533**	Cheonan	CN	**0.674**	Incheon*	IC	**0.577**
Gangneung	GN	**0.449**	Cheongju	CJ	**0.674**	Seoul*	SU	**0.693**
Gunsan	GS	0.164	Daegu*	DG	**0.703**	Suwon	SW	**0.818**
Jeju	JJ1	**0.337**	Daejeon*	DJ	**0.724**			
Mokpo	MP	**0.427**	Gumi	GM	**0.563**			
Pohang	PH	**0.398**	Gwangju*	GJ	**0.570**			
Seosan	SS	**0.506**	Jecheon	JC	**0.589**			
Ulsan*	US	0.186	Jeonju	JJ2	**0.404**			
Yeosu	YS	**0.251**	Jinju	JJ3	**0.396**			
			Wonju	WJ	**0.799**			
Average		0.366	Average		0.611	Average		0.619
Nationwide Average		0.514						

* Major metropolitan cities in South Korea (Seoul, Busan, Daegu, Incheon, Gwangju, Daejeon, and Ulsan).

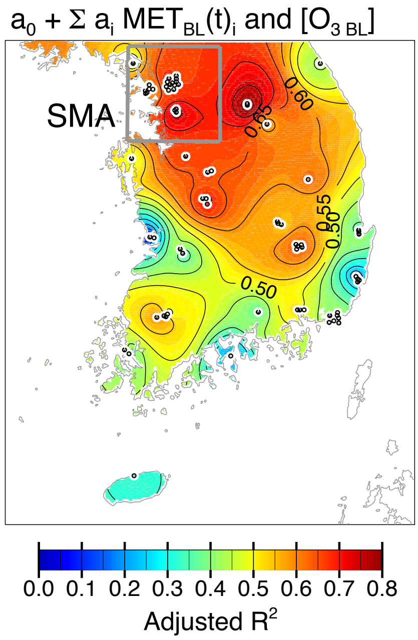

Figure S5. Spatial distribution of adjusted coefficient of determination (adjusted R^2) between baselines of $O_{3\ 8h}$ ($[O_{3\ BL}]$) and multiple linear regression models ($a_0 + \sum_i a_i\, \mathrm{MET}_{BL}(t)_i$).

To include the adjusted R^2 for the models in the revision, we have inserted Fig. S5 and Table S1 into Fig. 5 and Table 2 respectively. The lines 9–13 on page 1204 thus have been revised as follows:

As a result of the multiple linear regression, coefficients of determination (R^2) between baselines of $O_{3\ 8h}$ and each meteorological variable, as well as adjusted R^2 for the multiple linear regression models, were calculated for 72 air quality monitoring sites distributed in 25 cities nationwide and summarized in Table 2. The nationwide average of adjusted R^2 is 0.51 and that of R^2 is 0.50 for SI, 0.29 for PS, 0.22 for T_{max}, 0.14 for TD, 0.05 for RH, and 0.03 for WS, respectively.

The line 21 on the same page also has been changed as follows:

The spatial distributions of R^2 for T_{max} and SI, as well as the adjusted R^2 for the combined meteorological effects, are represented in Fig. 5.

6. A question on the choice of variables used. In section 3.2, the authors show that R-squared values are higher for insolation than for T_{max}, and mention the more indirect effect of T on O_3 production (Dawson et al., 2007). But have they tried to use daily T averaged at daytime instead of T_{max}? Please note that this is just a suggestion, not a major concern. I do not expect the authors to modify their analysis at this stage. Only if this is not too onerous it would be interesting to know if with a different choice the explained variance may improve. It might also be good to briefly introduce why some of the meteorological variables indicated in the paragraph above (e.g. TD, RH) are used.

→ Following the referee's suggestion, we have applied multiple linear regression models using the daytime average temperature (T_{day}). Newly calculated R^2 for T_{day} and adjusted R^2 for the regression models are shown in Fig. S6. Figs. S6a and b for T_{day} is very similar in the magnitude value and spatial pattern of the R^2 and adjusted R^2 to Fig. 5a in the original manuscript and Fig. S5 for T_{max} respectively. There are no significant differences or improvement with the different choice.

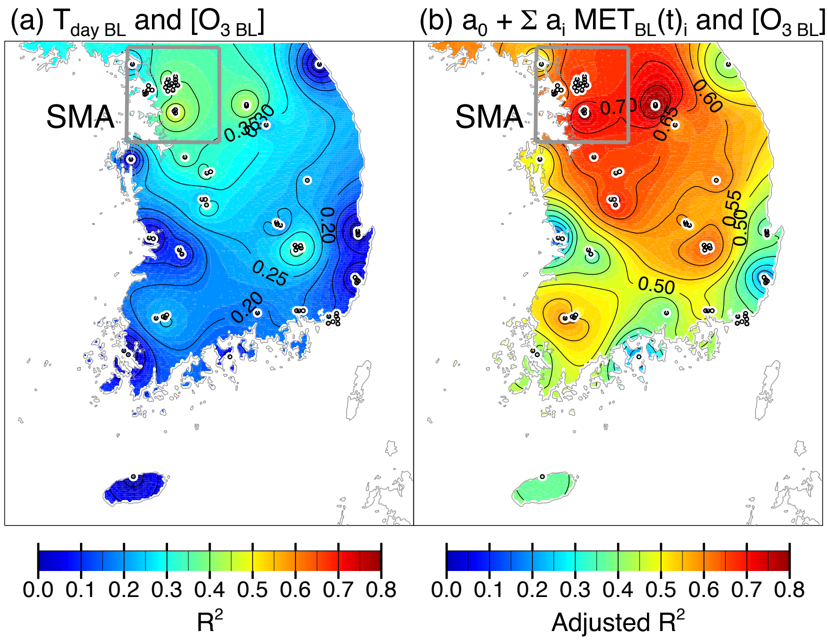

Figure S6. Spatial distributions of **(a)** R^2 between baselines of $O_{3\ 8h}$ ($[O_{3\ BL}]$) and daytime average temperature ($T_{day\ BL}$) and **(b)** adjusted R^2 between $[O_{3\ BL}]$ and multiple linear regression models ($a_0 + \sum_i a_i\ MET_{BL}(t)_i$).

In the revised manuscript, we have modified the lines 6–9 on page 1196 to briefly introduce the choice of meteorological variables as follows:

The meteorological variables used in this study include common factors related to the O_3 variations such as temperature (°C), surface insolation (W m^{-2}), relative humidity (%), and wind speed (m s^{-1}) (e.g. Ordóñez et al., 2005; Camalier et al., 2007; Jacob and Winner, 2009). Dew-point temperature (°C) and sea-level pressure (hPa) are additionally applied for multiple linear regression models as other previous studies have done (e.g., Thompson et al., 2001; Shin et al., 2012). Finally wind direction (16 cardinal directions) is used to reveal its relationship with short-term changes in O_3.

7. Similar question (about the impact of T and surface insolation SI on O_3) but from a different perspective. I understand that the correlations in Fig. 5 are done for all baseline data, considering the warm and cold seasons. I expect the surface insolation to have a stronger impact than T in winter, since it will favour the vertical mixing of pollutants and reduce the O_3 loss by titration while the effect of temperature might be not so clear. Might it be that T becomes much more relevant during the high ozone season (May–October) and that for that period the values of R-squared for $[O_{3\ BL}]$ with T_{BL} and SI_{BL} become much closer than shown in Fig. 5?

→ First of all, we would like to change the terminology 'high O_3 season' to 'months of frequent high O_3 events' in the revised manuscript to prevent confusion. As in the last paragraph of Sect. 2.2, O_3 concentrations in South Korea are highest in spring (March to June) and also high in autumn (September and October) although the clear sky insolation is maximum in summer. While high O_3 episodes frequently occur with clear sky and high temperature condition during May to October, averaged O_3 concentrations in summer are lower than those in spring. It is because frequent precipitation events related to the East Asian summer monsoon often reduce both insolation and precursors. To focus on the high O_3 events and wind directions, we examined the short-term component of O_3 during May to October in Sect. 3.5.

The referee expected that the impacts of temperature and insolation on O_3 variations would be different in cold and warm seasons. In Fig. S7, R^2 between baselines of $O_{3\ 8h}$ ($[O_{3\ BL}]$) and daily maximum temperature ($T_{max\ BL}$) or daily average insolation (SI_{BL}) for each season are presented. The figure shows that the referee's comment is qualitatively true despite of the mostly unclear effects of temperature in summer (Fig. S7c) and insolation in spring (Fig. S7f). It should be noted that the correlations in winter and summer are naturally small because the variables usually have little intra-seasonal variations in the baseline time scale (period larger than 50 days) compared to the transitional seasons. Especially in summer, the sporadic extremes of high O_3 and temperature are removed by the KZ-filter, and this is one reason for the low correlation between $[O_{3\ BL}]$ and $T_{max\ BL}$ in Fig. S7c. On the other hand, the low correlation between $[O_{3\ BL}]$ and SI_{BL} in spring (Fig. S7f) may reflect multiple factors to contribute to the high springtime O_3 in the Northern Hemisphere such as episodic stratospheric intrusion, transport by the continental outflow, and photochemical reactions of accumulated precursors during the winter. It will be further studied and discussed in future research.

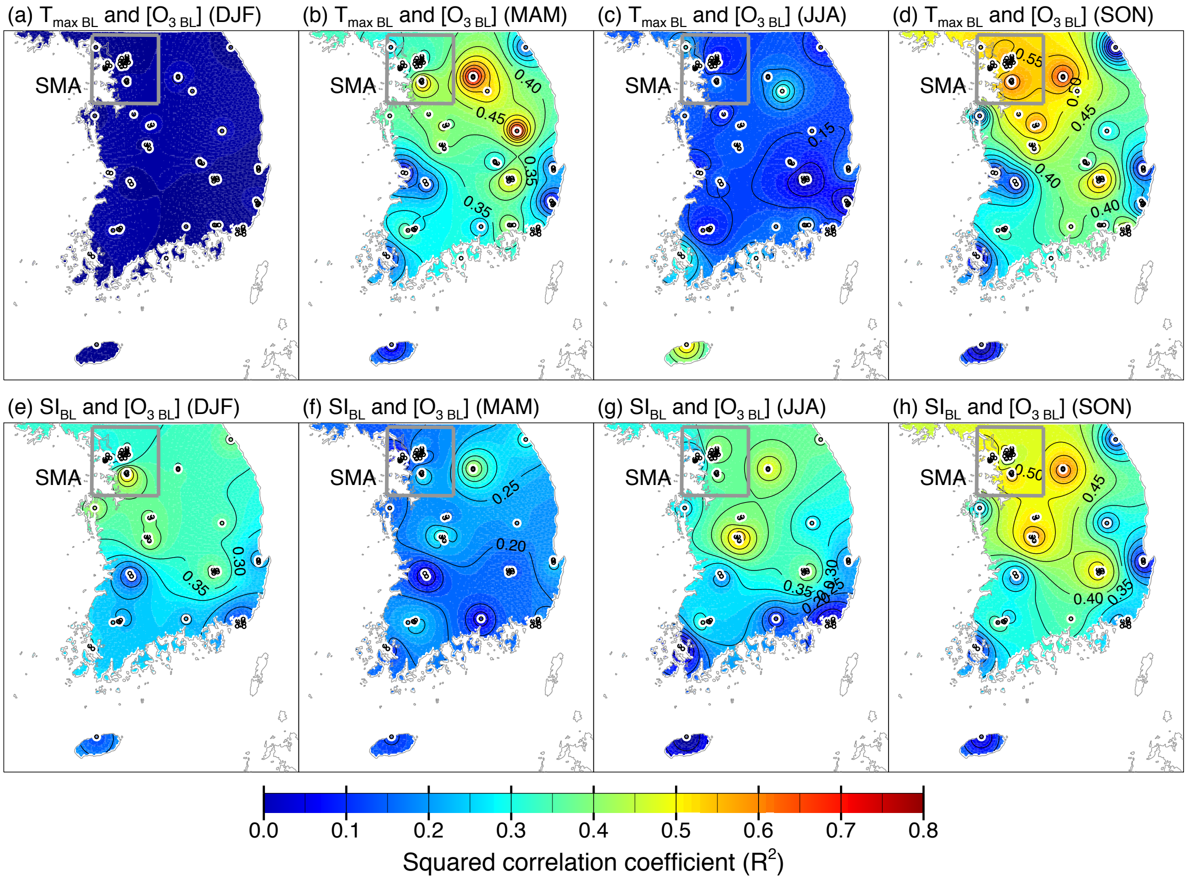

Figure S7. Spatial distributions of R^2 between baselines of $O_{3\ 8h}$ ([$O_{3\ BL}$]) and daily maximum temperature ($T_{max\ BL}$) in **(a)** winter, **(b)** spring, **(c)** summer, and **(d)** autumn, and R^2 between [$O_{3\ BL}$] and daily average insolation (SI_{BL}) during **(e)** winter, **(f)** spring, **(g)** summer, and **(h)** autumn for the period 1999–2010.

8. Another comment following the one before: In the conclusions the authors say "The high meteorological influences in the SMA and inland regions are related to effective photochemical activity, which results from large local precursor emissions and stagnant condition with low wind speeds". This would be true during the high O_3 season, but most of the time they show results for the whole year.

→ Figure S7 shows that the effects of insolation are high in the SMA and inland regions except during the springtime when multiple factors contribute the high O_3 levels. Compared to the coastal region, the SMA and inland regions have large local emissions and stagnant condition, which leads to the higher correlations between insolation and O_3 appear in the SMA and inland region during the winter, summer, and autumn. Therefore, the sentence pointed out by the referee is roughly true.

9. Throughout the paper the authors mention that meteorological effects (temperature and surface insolation) on ozone levels are high at the inland and SMA cities and low at the coastal cities, where the wind speed and long-range transport are more relevant. For instance, they finish section 3.2 with the sentence "Therefore, the meteorological effects on the O_3 productions become more important in the inland region where the wind speeds are lower". It is very clear to the reader what they mean by this. However, I would also consider the wind

speed to be a meteorological effect and therefore I am not sure the terminology they use is the most appropriate one. Is there another possible way of expressing this?

→ We concur with the referee's point. The term 'meteorological effects' used in the manuscript mainly represents the effects of temperature and insolation on the O_3 changes. However, it also partly includes the effects of wind speeds. Therefore, due to the usage of the term the sentence pointed out by the referee may cause confusion. To prevent that confusion, we have changed the term in the sentence with 'effects of temperature and insolation'.

10. Section 3.4 (Relative contributions of O_3 variations in different time-scales). As indicated by the authors, Figs. 9a and 9b illustrate the negative relationship between the relative contributions of [$O_{3\ ST}$] and [$O_{3\ SEASON}$]. They say "the large relative contributions of [$O_{3\ ST}$] at the coastal cities indicate the stronger effects of the synoptic-scale transport of background O_3 there". The highest contribution of [$O_{3\ ST}$] is in the North East, where I believe there is only one ozone monitoring site. Figure 1c shows the location of that site, close to the coast (on the East) but also to the mountains (on the West). I have a couple of considerations: (a) Is there any particularity about the location and topography of that site that might cause the very high relative contribution of the short-term component there? (b) Taking into account the lack of other O_3 monitoring sites in the proximities, I assume the spatial interpolation performed with the AIDW method might not work so well for the elevated area in the North East; this could affect any of the contour plots shown in the paper. This will not affect the validity of the main conclusions of the manuscript, but it might be worthwhile to acknowledge it.

→ (a) The monitoring site pointed out by the referee is at Gangneung, a northeastern coastal city. High and steep mountains on the west and sea on the east represent topographical characteristics of the city. In the short-term time scale, combined effects of wind directions related to eastward moving synoptic weather systems and 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 clear sky and strong vertical mixing over the region. Since the westerly winds contain the precursors emitted from the SMA and inland regions, 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.

(b) We agree with the referee's comment and have added a sentence in section 2.3, line 22 on page 1200 as follows:

In addition, mapping with a few monitoring sites combined with complex mountainous terrain can also distort the actual distribution of data, especially in the northeastern part of South Korea.

Technical corrections

The list of technical corrections would be too long to list here. As indicated above I have provided a pdf version of the paper with annotated changes (see supplement). Please do not pay attention to the formatting (the text was simply copied from the ACPD printer-friendly version to a text editor), but to the changes annotated in that document.

→ We really appreciate the referee's help on this. Following the referee's comment, we have also modified the lines 11–16 on page 1197 as follows:

For the clear separation of the components, we applied KZ-filter to the daily log-transformed O_3 as in Rao and Zurbenko (1994) and Eskridge et al. (1997), instead of the raw O_3 concentrations. While the short-term component separated by the KZ-filter using raw O_3 data still shows clear seasonality, use of $ln(O_3)$ makes the short-term component stationary and nearly independent of the seasonal influence by stabilizing variance (Rao and Zurbenko, 1994; Rao et al., 1997).

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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.

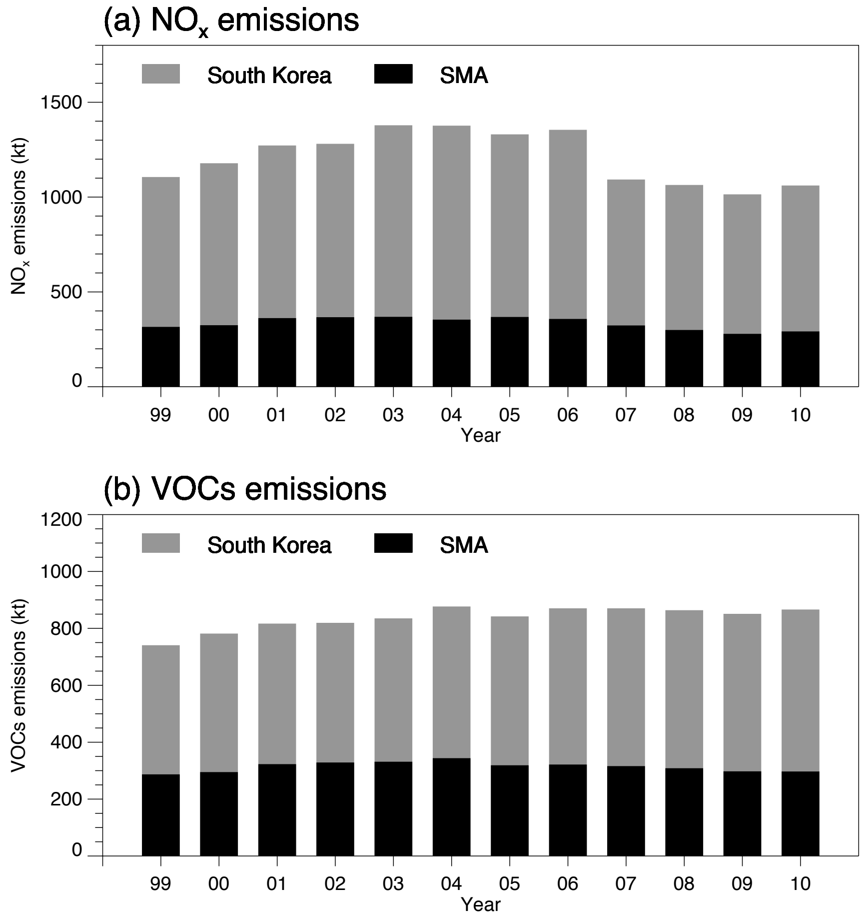

Figure S1. Total anthropogenic emissions of **(a)** NOx and **(b)** VOCs in the SMA and in South Korea (KMOE, 2013).

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.

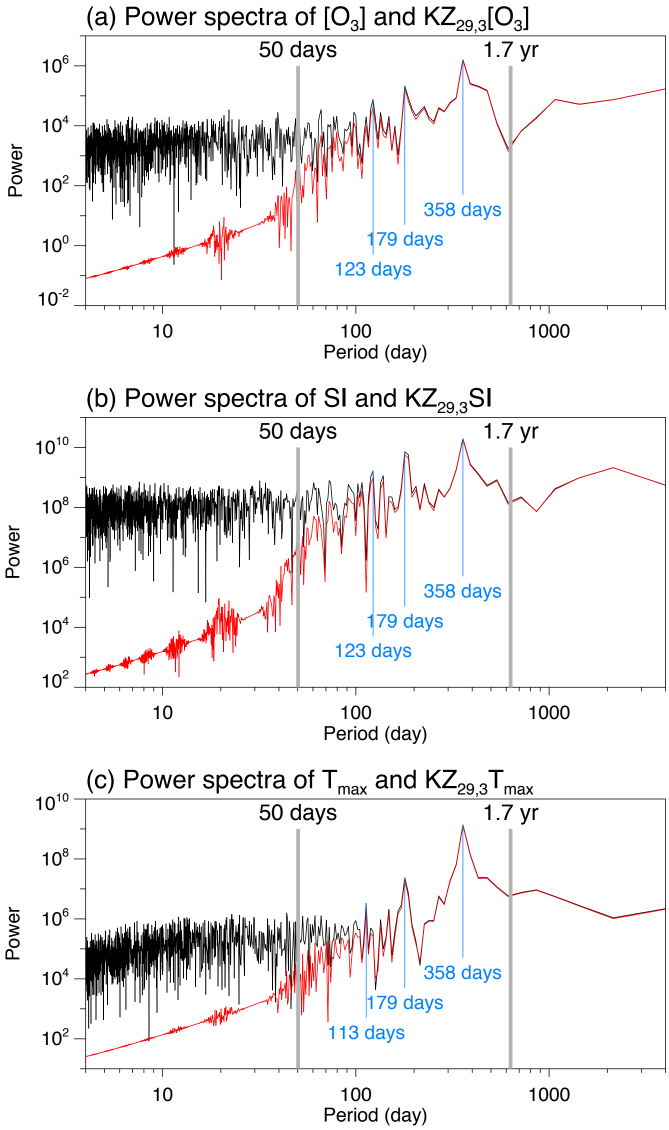

Figure S2. Power spectra of **(a)** log-transformed $O_{3\ 8h}$ time series ([O_3]) and its baseline ($KZ_{29,3}$[O_3]) at the City Hall of Seoul, **(b)** daily average surface insolation (SI) and its baseline ($KZ_{29,3}$SI), and **(c)** daily maximum temperature (T_{max}) and its baseline ($KZ_{29,3}T_{max}$) 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 $KZ_{29,3}$.

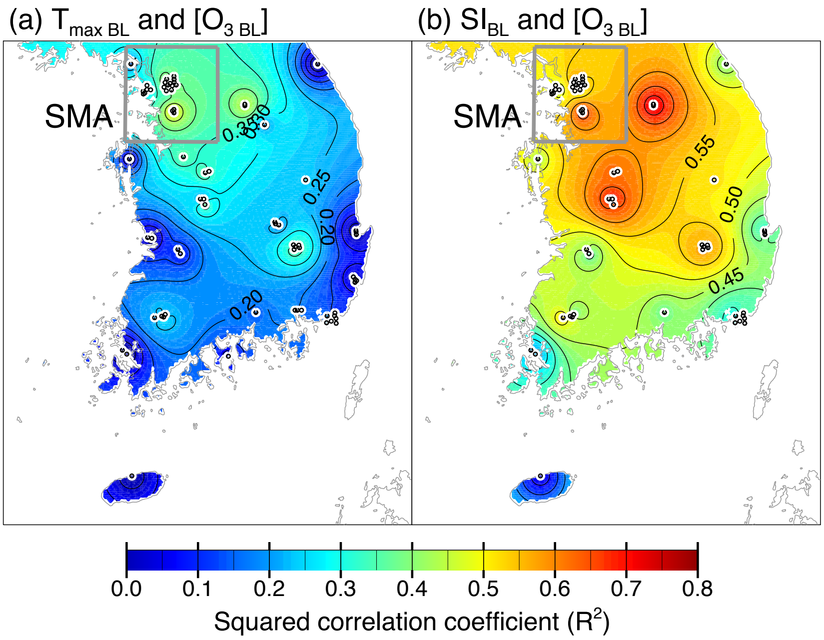

Figure S3. Spatial distributions of coefficients of determination (R^2) between baselines of $O_{3\ 8h}$ ($KZ_{15,5}[O_3]$) and **(a)** daily maximum temperature ($KZ_{15,5}T_{max}$) and **(b)** surface insolation ($KZ_{15,5}SI$). Each baseline is obtained by applying $KZ_{15,5}$ filter.

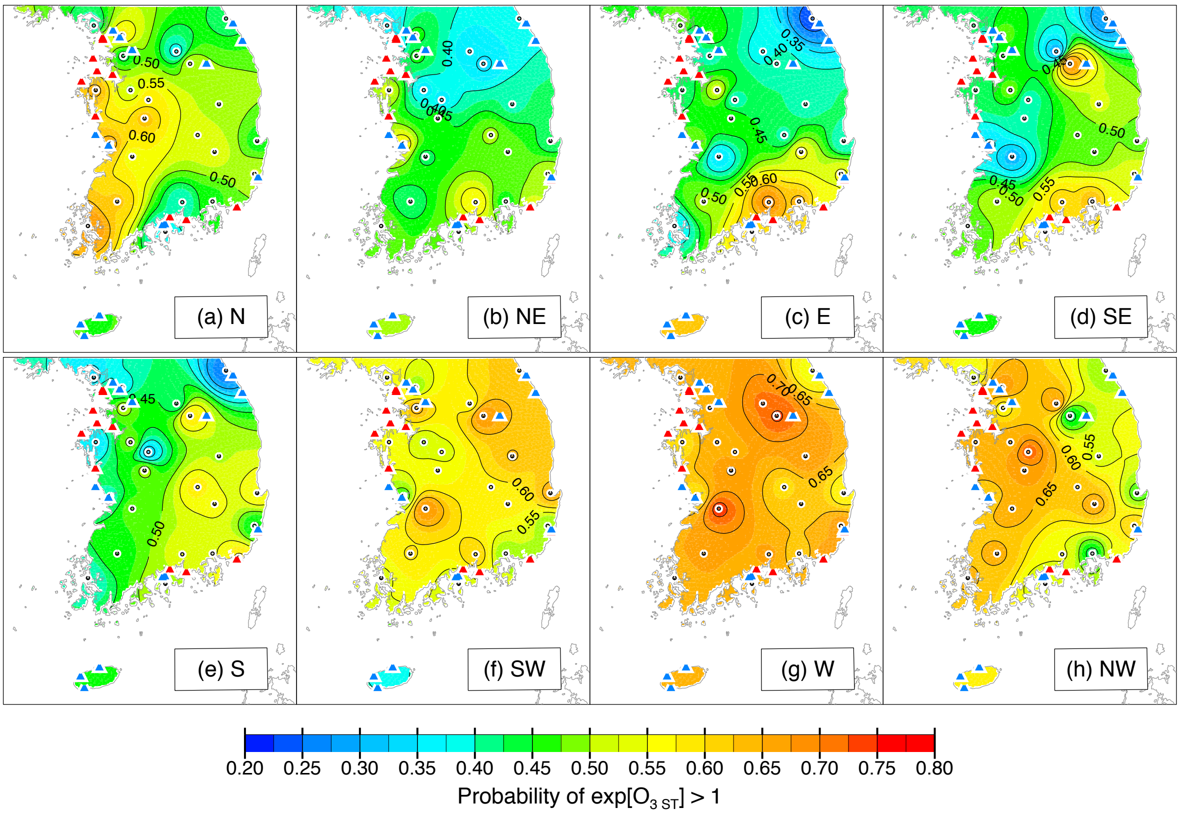

Figure S4. Spatial distribution of probabilities that exponentials of the short-term components will exceed 1 (exp[$O_{3\ ST}$]>1) for each wind direction (WD). Each short-term component is obtained by applying the $KZ_{15,5}$ filter.

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 population, industry, and transportation are concentrated, the background O_3 levels 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. However, in the coastal region where the NO_x concentrations are low (Fig. 3b), the less titration and nitrate formation at nighttime lead to 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.

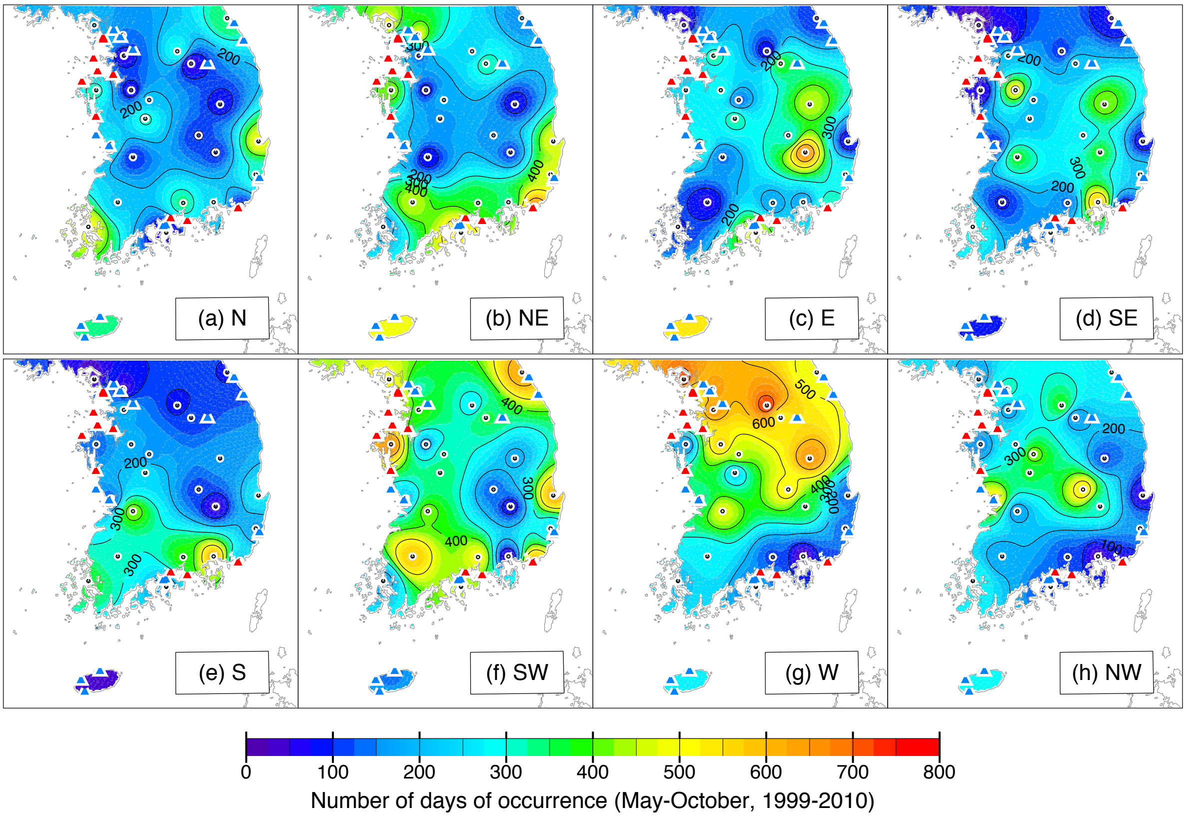

Figure S5. Spatial distributions of number of days in each wind direction (WD) during the months of frequent high O_3 events (May–October) for the period 1999-2010.

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.

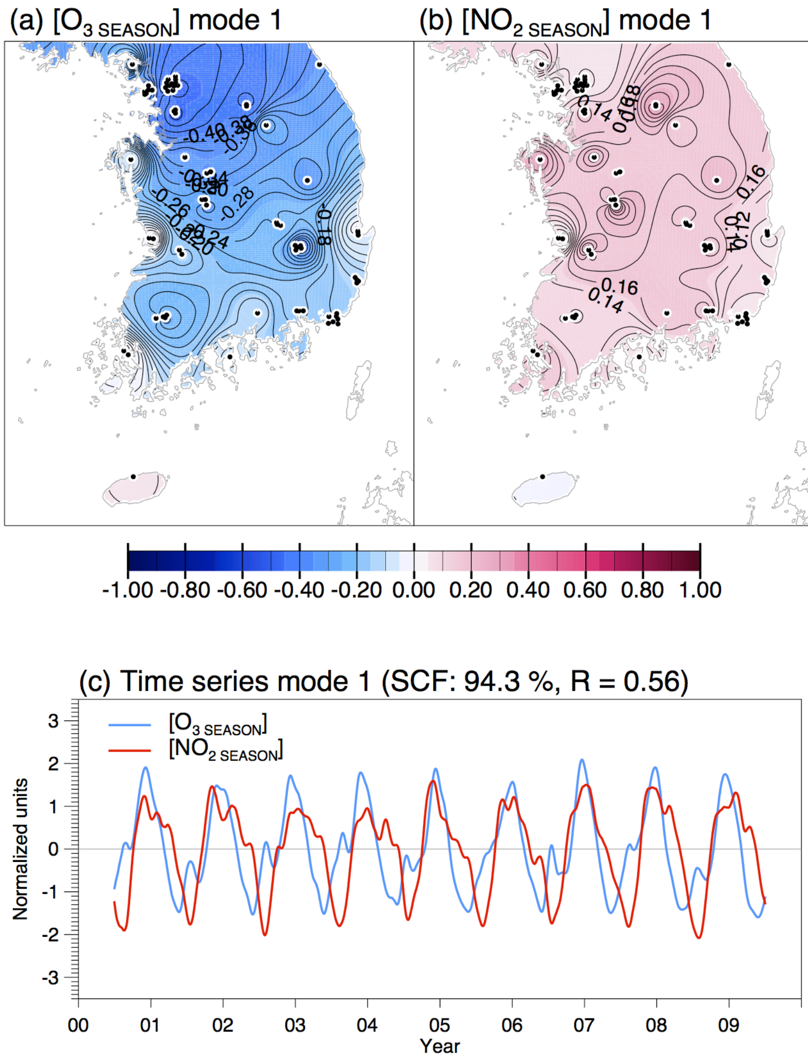

Figure S6. The first leading mode of SVD between the seasonal components of **(a)** daily maximum 8-h average O_3 ([O_3 SEASON]) and **(b)** daily average NO_2 ([NO_2 SEASON]) with **(c)** time series of the SVD expansion coefficient associated with [O_3 SEASON] mode (blue line) and [NO_2 SEASON] mode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

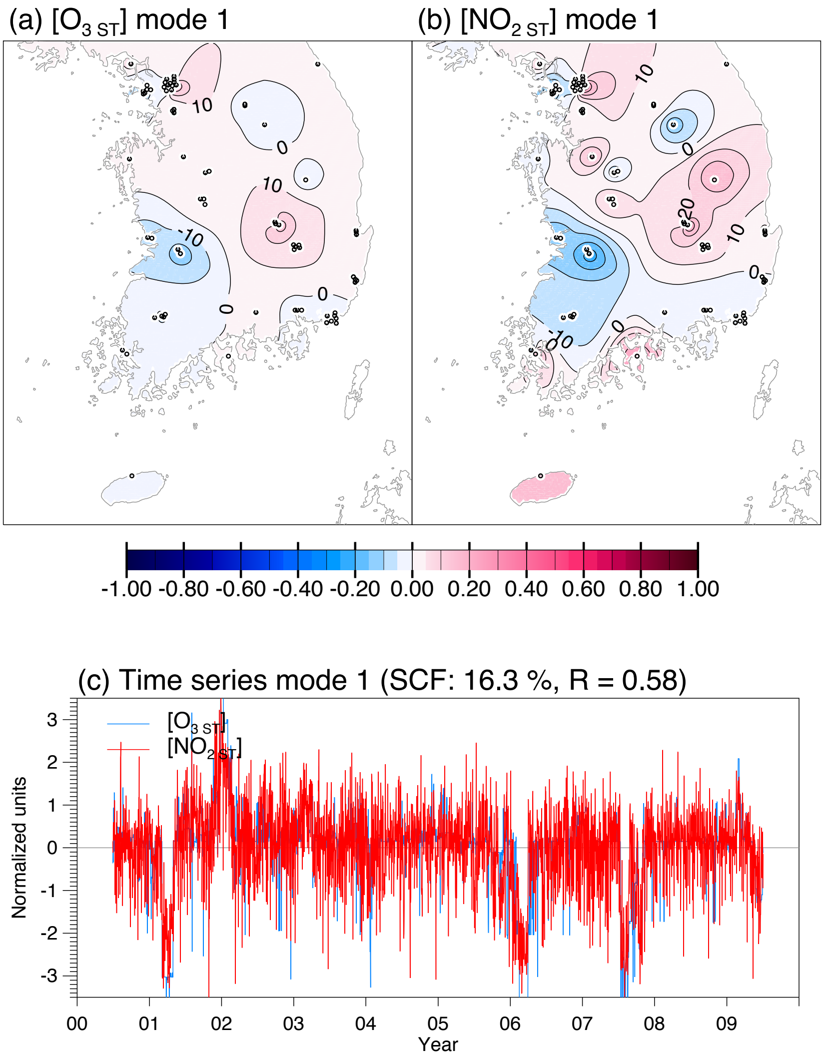

Figure S7. The first leading mode of SVD between the short-term components of **(a)** daily maximum 8-h average O_3 ([$O_{3\ ST}$]) and **(b)** daily average NO_2 ([$NO_{2\ ST}$]) with **(c)** time series of the SVD expansion coefficient associated with [$O_{3\ ST}$] mode (blue line) and [$NO_{2\ ST}$]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

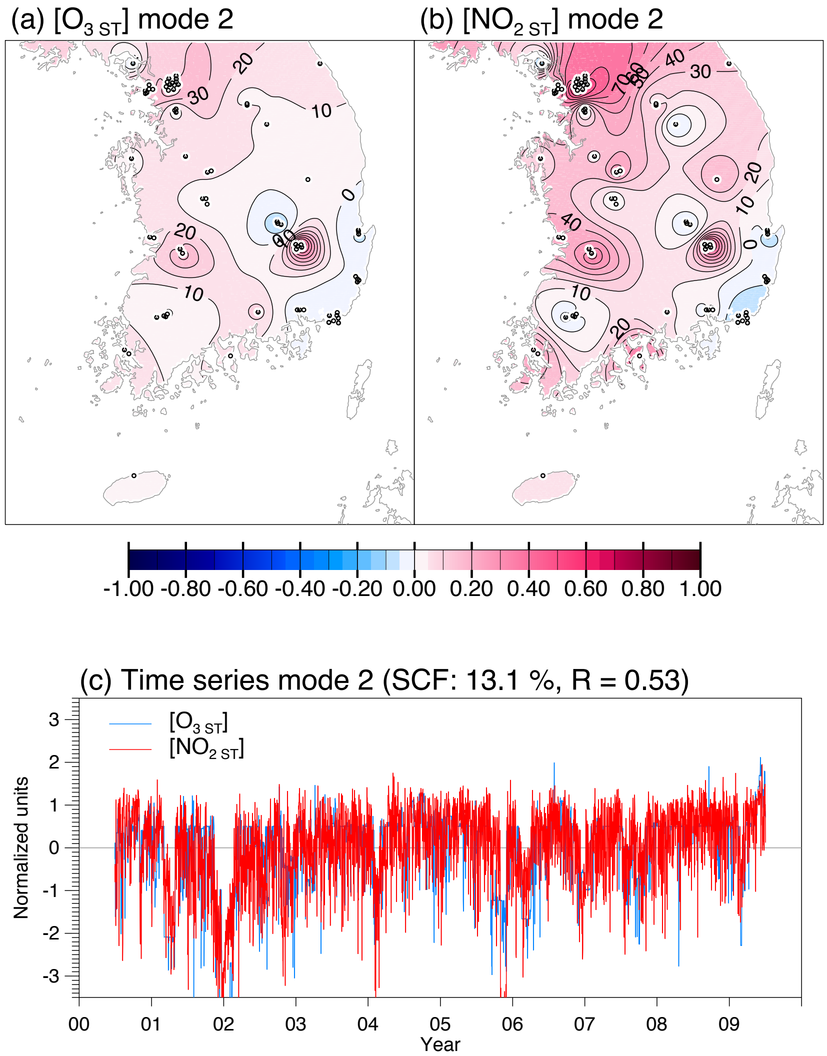

Figure S8. The second mode of SVD between the short-term components of **(a)** daily maximum 8-h average O_3 ([$O_{3\ ST}$]) and **(b)** daily average NO_2 ([$NO_{2\ ST}$]) with **(c)** time series of the SVD expansion coefficient associated with [$O_{3\ ST}$] mode (blue line) and [$NO_{2\ ST}$] mode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

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