Response to Anonymous Referee #2

In the manuscript, the authors used various statistical tools such as the K-Z filter and multiple linear regression method to the 18 year long-term data of the criteria air pollutants and meteorological variables. They could separate short term variations and long-term variation. Further, out of the long-term trend, they could separate the meteorological and emission driven trends. In addition they calculated local emission driven and transported components from the emission driven part based on the continuity equation approach.

It is a well organized manuscript and the results are of great importance since this study result can be complementary to the 3-dimensional chemical transport modeling results and, thus, it can provide a scientific background for effective policy development in South Korea.

However, there are several points that should be improved and clarified. Thus, I recommend the manuscript be accepted for the publication in the Journal with minor revisions. Specific points are:

We appreciate the reviewer's careful reading, valuable comments, and constructive suggestions. We modified the original manuscript as following the reviewer's comments and suggestions. Each response to the reviewer colored in blue and changes in the manuscript colored in red.

1. Abstract needs revision to further emphasize the scientific significances of the study.

We revised the last sentence of the abstract (L27–29 on p.1) as follows:

The present results not only reveal an important role of synoptic meteorological conditions on the episodic air pollution events but also give insights into the practical effects of environmental policies and regulations on the long-term air pollution trends. As a complementary approach to the chemical transport modeling, this study will provide a scientific background for developing and improving effective air quality management strategy in Seoul and its metropolitan area.

2. It would make the manuscript more valuable to compare the results with other the study results for South Korea in which the same statistical tools have been used (for example, Shin et al., AAQR, 12, 93, 2012). Also, it would be nice to refer and compare the study results on the influence of meteorological parameters on the air pollutant level (for example, Lim et al., JKOSAE, 28, 325, 2012).

Thanks for the suggestion. To add a brief comparison with the previous results from Shin et al. (2012), we modified L2–4 on p.14 as follows:

In terms of O_3, positive $\frac{\partial}{\partial t}\left(\mathrm{O_3}\,{}_{8\mathrm{h}}{}_{\mathrm{LT}}^{\mathrm{emis(T)}}\right)$ during the 2000s (Fig. 5e) implies that the transport of regional background O_3 played an important role in the meteorologically-adjusted long-term $O_{3\,8h}$ trend ($\mathrm{O_3}\,{}_{8\mathrm{h}}{}_{\mathrm{LT}}^{\mathrm{emis}}$) during the period. This result is consistent with the previous study on O_3 in Seoul for 2002–2006 using the OZone Isopleth Plotting Package for Research (OZIPR) and the KZ filter (Shin et al., 2012). However, because $\mathrm{O_3}\,{}_{8\mathrm{h}}{}_{\mathrm{LT}}^{\mathrm{emis(L)}}$ has been gradually changed from the decreasing phase to the increasing phase over the analysis period (Fig. 5e), the recent changes in $\mathrm{O_3}\,{}_{8\mathrm{h}}{}_{\mathrm{LT}}^{\mathrm{emis}}$ in the 2010s are probably more related to the local secondary production rather than the background O_3 transport.

Also, a short discussion of comparison with the dispersion effect of winds on PM_{10} and NO_2 levels reported by Lim et al. (2012) is inserted after L11 on p.12 as follows:

The long-term component of wind speed (WS_{LT}) in Seoul was increased by ~0.9 m s^{-1} from 2002 to 2012 (Fig. 6a). Previous statistical research on the dispersion effects of winds reported that the fraction of concentrations reduced by wind speed rising of 2 m s^{-1} to 3 m s^{-1} was ~24% for PM_{10} and ~9% for NO_2 (Lim et al., 2012). These ratios are comparable to the decrease of meteorology-related long-term concentrations

($\chi_{\mathrm{LT}}^{\mathrm{met}}$) in PM_{10} and NO_2 by ~30% and ~10% for the period, respectively ($\Delta X_{\mathrm{LT}}^{\mathrm{met}}$ $(= \Delta\chi_{\mathrm{LT}}^{\mathrm{met}}/\chi_{\mathrm{LT}}^{\mathrm{met}})$ is −0.3 for PM_{10} and −0.1 for NO_2; Fig. 5a and d).

3. In Table 1, add a part on explaining why the sum of the variances is not 100%. Also, in Table 4, it would better to make the sum of the trends of 'emis' and 'met' be equal to the total.

Explained variances in Table 1 are identical to coefficients of determination (R^2) between the original time series (X) and each decomposed component (X_{ST}, X_{SN}, and X_{LT}), which represent how much of the variability of X is accounted for by the variability of each component. The explained variances of the decomposed time series should sum to 100%, if X_{ST}, X_{SN}, and X_{LT} were completely independent of each other. However, because the components decomposed by the KZ filter still have some minor correlations among them albeit very weak, the explained variance of each component (R^2 with X in percentage) are contributed not only by the variance itself but also by the covariances with other components.
In the revised version, to clarify this, we replaced the explained variances in the original Table 1 with the proportions of each variance and covariance to the total variances. The sum of variances of and covariances among the short-term, seasonal, and long-term components are exactly the same as the total variances of the original time series (see the formula in the revised Table 1).
Following the modified Table 1, "(~50–70%)" in L19 on p.8 is now "(~46–68%)." Also, the original version of Table 1 is modified to the R^2 values and is now added to the supplement as Table S2.

Table 1: Total variances ($Var(X)$) of log-scale times series for Seoul average concentrations of PM_{10}, SO_2, NO_2, CO, and $O_{3\ 8h}$, and relative contributions (%) of variances of and covariances (Cov) among each component to $Var(X)$. Daily data for the period of July 2000 to Jun 2015 that X_{LT} data is available were used.

	PM_{10}	SO_2	NO_2	CO	$O_{3\ 8h}$
$Var(X)$[a]	0.2998	0.1345	0.1400	0.1540	0.4068
$Var(X_{\mathrm{ST}})$	63.98%	48.92%	67.94%	46.19%	42.96%
$Var(X_{\mathrm{SN}})$	22.07%	40.58%	23.86%	34.66%	47.06%
$Var(X_{\mathrm{LT}})$	8.74%	4.50%	3.64%	14.32%	5.00%
$Cov(X_{\mathrm{ST}}, X_{\mathrm{SN}})$	2.91%	2.53%	2.20%	2.00%	2.17%
$Cov(X_{\mathrm{SN}}, X_{\mathrm{LT}})$	−0.29%	0.47%	0.09%	0.42%	0.32%
$Cov(X_{\mathrm{ST}}, X_{\mathrm{LT}})$	−0.01%	0.00%	−0.01%	0.00%	0.01%
$Var(X_{\mathrm{LT}}^{\mathrm{emis}})$	2.49%	4.88%	1.80%	4.97%	1.90%
$Var(X_{\mathrm{LT}}^{\mathrm{met}})$	2.85%	0.09%	1.08%	4.45%	1.52%
$Cov(X_{\mathrm{LT}}^{\mathrm{emis}}, X_{\mathrm{LT}}^{\mathrm{met}})$	1.70%	−0.23%	0.38%	2.45%	0.79%

[a] Values of variances of each log-scale concentration time series
$Var(X) = Var(X_{\mathrm{ST}}) + Var(X_{\mathrm{ST}}) + Var(X_{\mathrm{ST}}) + 2[Cov(X_{\mathrm{ST}}, X_{\mathrm{SN}}) + Cov(X_{\mathrm{SN}}, X_{\mathrm{LT}}) + Cov(X_{\mathrm{ST}}, X_{\mathrm{LT}})]$

Table S2. Coefficients of determination (R^2) between each component and original time series (X) for Seoul average concentrations of PM_{10}, SO_2, NO_2, CO, and $O_{3\ 8h}$. Daily data for the period of July 2000 to Jun 2015 that X_{LT} data is available were used.

Components	PM_{10}	SO_2	NO_2	CO	$O_{3\ 8h}$	Notes
X_{ST}	0.699	0.541	0.724	0.503	0.474	Short-term components
X_{SN}	0.276	0.468	0.287	0.397	0.522	Seasonal components
X_{LT}	0.081	0.055	0.038	0.152	0.057	Long-term components
$X_{\mathrm{LT}}^{\mathrm{emis}}$	0.064	0.054	0.030	0.124	0.043	Emission-related X_{LT}
$X_{\mathrm{LT}}^{\mathrm{met}}$	0.069	0.003	0.019	0.107	0.040	Meteorology-related X_{LT}

In Table 4, the sum of the linear trends of $X_{\mathrm{LT}}^{\mathrm{emis}}$ and $X_{\mathrm{LT}}^{\mathrm{met}}$ should be exactly the same as the X_{LT} trend. However, the rounding after the calculation that we used here can induce tiny difference of ±1 in the last decimal place of both $\frac{\partial X_{\mathrm{LT}}}{\partial t}$ and the sum of $\frac{\partial}{\partial t} X_{\mathrm{LT}}^{\mathrm{emis}}$ and $\frac{\partial}{\partial t} X_{\mathrm{LT}}^{\mathrm{met}}$. For example, ${\mathrm{PM}_{10}}_{\mathrm{LT}}^{\mathrm{emis}}$ trend ($-1.6883300\%\ \mathrm{yr}^{-1}$) and ${\mathrm{PM}_{10}}_{\mathrm{LT}}^{\mathrm{met}}$ trend ($-1.9456153\%\ \mathrm{yr}^{-1}$) exactly sum to ${\mathrm{PM}_{10}}_{\mathrm{LT}}$ trend ($-3.6339453\%\ \mathrm{yr}^{-1}$). However, the trends rounded to the second decimal place are $-1.69\%\ \mathrm{yr}^{-1}$ for ${\mathrm{PM}_{10}}_{\mathrm{LT}}^{\mathrm{emis}}$ and $-1.95\%\ \mathrm{yr}^{-1}$ for ${\mathrm{PM}_{10}}_{\mathrm{LT}}^{\mathrm{met}}$, and thus their sum is $0.01\%\ \mathrm{yr}^{-1}$ larger than the rounded-off ${\mathrm{PM}_{10}}_{\mathrm{LT}}$ trend ($-3.63\%\ \mathrm{yr}^{-1}$). For the result value, we think that the rounding after the calculation is more accurate than the rounding before the calculation. Therefore, we left Table 4 as the original version.

4. In eq. (9), should it be a residual term or S_{LT} contains all terms except advection?

By Eq. (9), the term S_{LT} should contain all effects on the change rate of long-term trend ($\frac{\partial X_{\mathrm{LT}}}{\partial t}$), except the changes of inflow and outflow by horizontal advection ($-\vec{V}_{\mathrm{LT}} \cdot \nabla X_{\mathrm{LT}}$). The residual effects are source (surface emissions and atmospheric secondary production related to $X_{\mathrm{LT}}^{\mathrm{emis(L)}}$ and partly $X_{\mathrm{LT}}^{\mathrm{met}}$), sink (physical and chemical scavenging affected by $X_{\mathrm{LT}}^{\mathrm{met}}$), and diffusion (accumulation and dissipation), as well as the vertical advection term ($-w_{\mathrm{LT}} \frac{\partial X_{\mathrm{LT}}}{\partial z}$). The term $-w_{\mathrm{LT}} \frac{\partial X_{\mathrm{LT}}}{\partial z}$ is negligible because the long-term trend of vertical motion (w_{LT}) is close to zero. Therefore, S_{LT} can be regarded as the sum of source term (directly related to $\frac{\partial}{\partial t} X_{\mathrm{LT}}^{\mathrm{emis(L)}}$) and sink and diffusion terms (affected by the long-term meteorological effect; $\frac{\partial}{\partial t} X_{\mathrm{LT}}^{\mathrm{met}}$).

5. What would be the main reason for CO(LT,Met) and SO_2(LT,Met) to show different trends though both of them are primary air pollutants?

The local meteorological factors such as wind speed, relative humidity, insolation, and temperature are not directly related to the regional transport of air pollutants, and thus can affect mainly on the contribution of local source pollutants rather than transported pollutants. In other words, if the local emissions at a local place are assumed to be zero, the local meteorological effects on accumulation and secondary production ($X_{\mathrm{LT}}^{\mathrm{met}}$) in the long-term air pollution trend (X_{LT}) will be negligible. As we wrote in the third paragraph on p.11, the emission intensity of CO in Seoul (215 t/km^2) is higher than those in the upwind source areas (e.g., 98 t/km^2 for the Chinese eastern coast), while that of SO_2 in Seoul (7 t/km^2) is much lower compared to those in the upwind source areas (e.g., 14 t/km^2 for the Chinese eastern coast). Therefore, the long-term SO_2 trend (${\mathrm{SO}_2}_{\mathrm{LT}}$) is much less contributed by the changes in meteorological effects on its local emission (${\mathrm{SO}_2}_{\mathrm{LT}}^{\mathrm{met}}$) than by the changes in its regional emissions (${\mathrm{SO}_2}_{\mathrm{LT}}^{\mathrm{emis}}$). As we describe in L9–13 on p.13, the interannual variation of satellite-based SO_2 level over China is similar to ${\mathrm{SO}_2}_{\mathrm{LT}}$ (and ${\mathrm{SO}_2}_{\mathrm{LT}}^{\mathrm{emis}}$) in this study (Fig. 5c) and supports the large influence of the regional background SO_2 on the long-term SO_2 trend in Seoul.

6. The different trends of NO_2 and NO_x might be caused not only by the reasons explained in the manuscript but also with changing oxidative potential of the atmosphere (for example, Kim and Lee, AAQR, in press).

We appreciate the reviewer's comment. L8–L12 on p.11 was now modified as follows:

Interestingly, NO_2 level has been stabilized despite increasing of the number of vehicles in Seoul from 2.3 million in 1999 to 3.1 million in 2016 probably owing to implementation of natural gas vehicles and low emission diesel engines, and NO_x (= NO + NO_2) level has been even decreased from ~70 ppb to ~50 ppb for the same period (Shon and Kim, 2011; Kim and Lee, 2018). Such an increase of NO_2 to NO_x ratio implies that additional conversion of NO to NO_2 occurs somewhere before the emission (e.g., exhaust line of the vehicle) or in the atmosphere. Although further evidence is required, this can be attributable to expanding of diesel particulate filter (DPF) and diesel oxidation catalyst (DOC) usage for diesel vehicles or increase of the atmospheric oxidative potential in the SMA (Alvarez et al, 2008; Kim and Lee, 2018).