

## **Responses to the Referee #2's Comments**

Thank you very much for your providing valuable comments on this work. We have improved the original manuscript following your comments.

This paper uses an offline meteorological and air quality modeling system to investigate how the urban heat island effect alters the chemical evolution of ozone. This study builds upon similar findings from previous studies; however, the complex variations of topography and coastal environment could also alter the impact of the urban head island on the evolution of ozone and its precursors. There have been few such studies, so content of this paper should be useful to the air quality modeling community. There are still a number of issues, described next, that need to be addressed by the paper.

### **Major Comments:**

1) While the results of the paper are interesting and show that adequately including a treatment of the urban boundary layer is important for air quality purposes, the air quality simulations depend upon the meteorological simulations that are presented in another paper currently under review (Ryu and Baik, 2012). It is not known how well the model represents key factors, such as the magnitude of the UHI, differences in boundary layer height between the city and outside of the city, and winds in the city core that will affect the conclusions drawn in this study. Either this paper needs to be put on hold until the first paper is accepted and in press (and available for reviewers to access) or the authors need to include key results from that study that demonstrate the model performs reasonably well in representing observed temperature and winds in the urban area. Readers need to know if there are sufficient observations to verify the meteorological model predictions, otherwise, the present study is simply more of a modeling exercise that may or may not represent the true impact of the UHI on air quality predictions.

The paper, Ryu and Baik (2012), is now in press and available online. We updated the corresponding reference in the revised manuscript. In Ryu and Baik (2012), the simulated meteorological variables are evaluated against observation dataset. The WRF-SNUUCM coupled model showed satisfactory performances in reproducing the diurnal variations of

observed wind and temperature both in the city core and outside the core. The PBL height is reasonably well simulated by the model as compared with that observed in an urban site and that observed in a rural site (~50 km away from the urban core) although the validation result is not given in Ryu and Baik (2012). Note that the YSU PBL scheme showed a better performance in simulating PBL height in the urban site than the MYJ PBL scheme. Please, refer to the paper in the following website:

<http://journals.ametsoc.org/doi/abs/10.1175/JAMC-D-12-0157.1>

2) Some additional discussion is needed at the end of the paper to stress that the authors have examined one case and put this study into the proper context. They do mention that this case is for fair weather conditions when one might expect a larger impact of the UHI on the local meteorology. However, it would also be interesting to know how strong synoptic forcing needs to be that would overwhelm the UHI and the effects on air quality presented in this study.

Following your comment, we included the point you mentioned and stressed the necessity of further studies at the end of the paper. We agree with your proposal that quantitative analyses on how strong synoptic forcing is required to offset the effects of urban land-surface forcing on air quality need to be undertaken. In previous studies, it was found that urban-induced circulations (or urban heat island circulations) are detectable under calm or light wind conditions. A critical wind speed for the development of such circulations has been reported to be  $\sim 3\text{--}4 \text{ m s}^{-1}$  (Wong and Dirks, 1978). By performing numerical simulations, Lemonsu and Masson (2002) showed that when the synoptic wind speed reaches  $7 \text{ m s}^{-1}$  the urban-induced circulation cannot develop. The magnitude of strong synoptic forcing that would overwhelm the UHI and corresponding effects on air quality in the present study area can be different from that examined in previous studies. Therefore, further in-depth studies under various synoptic conditions needs to be carried out through comprehensive numerical simulations.

3) The authors clearly show that representing the urban impacts on meteorology can affect ozone chemistry in a noticeable way. However, it is not clear how this effect compares to other uncertainties in air quality predictions. For example, choice of PBL parameterization

could also introduce differences in ozone concentrations and uncertainties in emission inventories will impact predictions of ozone. How does the impact of the UHI compare to other commonly known uncertainties in air quality predictions? Some context is needed. It would be interesting to see Figure 2b repeated but using the simulation replacing the urban area with cropland. Presumably the results would be worse if the modeling system was adequately representing the overall ozone chemistry, transport, and mixing in the region. It is possible that the statistics would not look much different which would stress the difficulty in identifying the impact of the UHI using standard metrics by themselves.

Following your comment, we examined changes in ozone concentration owing to the choice of PBL parameterization scheme and changes in ozone precursor emissions. In the present study, the YSU PBL scheme is used and this is mentioned in the revised manuscript. To examine an uncertainty owing to the choice of PBL parameterization scheme, the MYJ PBL scheme is chosen and the results are presented in Table B1. The difference in area-averaged O<sub>3</sub> concentration between the baseline simulation with the YSU PBL scheme and the simulation with the MYJ PBL scheme is small in the daytime (3 ppb) but large in the nighttime (11 ppb) (Table B1).

To examine possible uncertainties in O<sub>3</sub> prediction associated with emission uncertainties, simulations in which anthropogenic NO<sub>x</sub> and anthropogenic VOC emissions are altered by 20% are performed. We considered four types of scenarios with 20% decrease in NO<sub>x</sub> emission (denoted by NO<sub>x</sub>0.8), 20% increase in NO<sub>x</sub> emission (denoted by NO<sub>x</sub>1.2), 20% decrease in VOC emission (denoted by VOC0.8), and 20% increase in VOC emission (denoted by VOC1.2). When either NO<sub>x</sub> emission or VOC emission is altered, the other precursor emission is set to be the same as the emission in the baseline simulation. In Table B1, it is found that the change in O<sub>3</sub> concentration owing to the change in the precursor emissions is larger in the daytime than in the nighttime and that O<sub>3</sub> concentration is more sensitive to the change in the NO<sub>x</sub> emission than to the change in the VOC emission. Four additional simulations in which the urban areas are replaced with cropland areas are performed with the same emission scenarios (Table B1). Even though the precursor emissions are altered, the impact of urban land-surface forcing is substantial for each emission scenario. So, the changes in precursor emissions do not change the conclusions drawn in this study. In summary, the changes in O<sub>3</sub> concentration owing to urban land-surface forcing are significant as compared with those owing to the choice of different PBL parameterization schemes and

those owing to the changes in O<sub>3</sub> precursor emissions.

Following your comment, the statistics for the NO-URBAN simulation in which the urban areas are replaced with cropland areas are given in Fig. B1. The performance of reproducing O<sub>3</sub> concentration is worse in the NO-URBAN simulation than in the baseline (URBAN) simulation. Therefore, the presented results suggest that the research approach adopted in this study is proper to examine the impacts of urban land-surface forcing.

4) The title refers to “air quality”, but really only deals with ozone chemistry. Other factors of interest to air quality, such as particulate matter, are not investigated. So I recommend changing the title from “air quality” to “ozone and its precursors” to better represent the contents of the paper.

We changed the title to “Impacts of urban land-surface forcing on ozone air quality in the Seoul metropolitan area” to better represent the contents of this study.

5) While the paper is well organized, there are numerous grammatical errors, some of which are pointed out in the specific comments. I likely did not catch all of them and suggest that the authors find an editor to help them with the final manuscript.

We corrected grammatical errors. Thank you.

#### **Specific Comments:**

Page 25794, lines 23-24: That may be true for some studies, but the impact of the UHI on simulated near-surface winds has been hard to verify in observations.

We agree with you, but urban-modified winds have been detected and/or simulated generally under weak synoptic wind conditions. The previous studies cited in the paper showed relatively strong UHI effects on near-surface winds.

Page 25795, line 19: “Advanced Research” is not needed.

Following your comment, “Advanced Research” is deleted.

Page 25795, line 26: Refer to Figure 1a. I assume the WRF grids area the same as the CMAQ grids?

The grid sizes used in the CMAQ model simulation are the same as those in the WRF model simulation. However, the domains used in the two model simulations are not the same as each other. The sizes of the domains in the CMAQ model simulation are a little bit smaller than those in the WRF model simulation because the perimeters of the domains in the WRF model simulation are trimmed.

Page 25796, lines 4-5: Change sentence to “The National Center for Environmental Prediction (NCEP) final analyses are used for the initial and boundary conditions.”

The sentence is changed.

Page 25796, line 12, Change “model” to “represent”.

Following your comment, “model” is changed to “represent”.

Page 25796, lines15-16: Perhaps just say the domains are the same as those used for WRF.

The domains are not the same as those used in the WRF model simulation. Please, see the response to the third specific comments.

Page 25796, lines 17-18: What are the default profiles for CMAQ? Perhaps include this information in a table for the most relevant species. Is long-range transport significant for this case and how might that affect the model results?

Following your comment, the default profiles for some relevant species are present in Table B2. This table is included in the supplement. In general, the effects of long-range transport of pollutants from China can be significant when the prevailing wind is westerly. In the present case, however, we consider that the effects of long-range transport are rather insignificant because the prevailing wind is easterly. Under the influence of easterly flow, little pollutants are transported to the Korean Peninsula.

Page 25797, lines 1-9: It would be useful to include a table summarizing the total emissions of NO<sub>x</sub>, VOCs, and other relevant species for the innermost grid for 2007 and 2008.

Figure B2 shows the total annual emissions of NO<sub>x</sub>, VOC, CO, and SO<sub>x</sub> in 2008. This is included in the supplement. We added the reduction ratios of the total annual emissions of NO<sub>x</sub> and VOC in 2008 to those in 2007 in the revised manuscript as follows.

“As compared with the total annual emissions in 2007, the total annual emissions of NO<sub>x</sub> (VOC) in 2008 are reduced by 37% (12%) in Seoul.”

Page 25797, line 15: Change the phrase to “is the rate at 1500 LT on a weekday (Thursday).”

Following your comment, it is changed to “is the rate at 15:00 LT on a weekday (Thursday)”.

Page 25799, line 7: Change “observed ones” to “observations”.

Following your comment, “observed ones” is changed to “observations”.

Page 25800, line 21-22: I disagree with the terminology of UHI here, and subsequently in the text. UHI usually refers to a horizontal temperature gradient between the urban area and the surroundings. The author should use another term or simply call this a temperature difference between the simulations; otherwise, use of UHI introduces confusion in the description of the model results.

Following your comment, the terminology of UHI that indicated the temperature difference between the two simulations is not used in the revised manuscript. We rephrased the term and replace it with “difference in air temperature between the two simulations” in the revised manuscript. However, we use the terminology of UHI that refers to a horizontal temperature difference between the urban area and the surroundings.

Page 25800, line 23: Change “appears in the nighttime” to “was simulated at night”.

The sentence and the terms you pointed out are reworded not to use the terminology of UHI.

Page 25801, lines 5 - 17: What is missing in this description is whether building wake effects

slow the winds down. Since the urban canopy model is not described it is hard to tell whether those effects are included.

In the numerical simulation, the effects of wind speed reduction above and within an urban canopy layer owing to the existence of buildings are considered. The effects of buildings on wind speed reduction are parameterized in the urban canopy model, and the details are described in Ryu et al. (2011). The content that the increased drag owing to buildings is included in the urban canopy model is added in Sect. 2.1.

Page 25802, line 2: Change “influences significantly” to “significantly influences”.

Following your comment, “influences significantly” is changed to “significantly influences”.

Page 25802, line 2-3: Change “hence urban-modified” to “hence the urban-modified”.

Following your comment, “hence urban-modified” is changed “hence the urban-modified”.

Page 25802, line 7: Change “concentration is” to “concentrations are”.

The corresponding sentence is changed as follows. “On average, O<sub>3</sub> concentration is 16 ppb higher in the nighttime (from 00:00 to 05:00 LT and from 20:00 to 24:00 LT) and is 13 ppb higher in the daytime (from 06:00 to 19:00 LT) in the URBAN simulation than in the NO-URBAN simulation.”

Page 25802, line 14: Change “less” to “reduced”.

Following your comment, “less” is changed to “reduced”.

Page 25802, lines 26-28: This sentence needed to be reworded. I think the authors mean to say that vertical mixing dominates in both simulations, so that the reduced destruction of O<sub>3</sub> by NO in the URBAN simulation is not that significant.

Following your comment, the sentence is reworded as follows. “The downward vertical mixing of O<sub>3</sub> from the residual layer dominates in both simulations, which leads to the small difference in O<sub>3</sub> concentration between the two simulations, so the effect of the reduced O<sub>3</sub>

destruction by NO in the URBAN simulation seems to be insignificant.”

Page 25804, line 2: What is “the cloud process”. Be more specific.

Following your comment, specific cloud processes are included in the revised manuscript.

Page 25804, line 10-11: This is a poorly worded sentence. Please fix.

The sentence is reworded as follows. “In the URBAN simulation, the chemical loss of O<sub>3</sub> is reduced in the surface layer.”

Page 25804, line 22: change “the more O<sub>3</sub> production in the surface layer, and the more” to “the enhanced O<sub>3</sub> production in the surface layer, and the enhanced”.

Following your comment, it is changed to “the enhanced chemical production of O<sub>3</sub> in the surface layer, and the enhanced”.

Page 25804, lines 23-25: Change sentence to “In addition to the urban-modified boundary layer, the urban-induced local circulation contributes . . .”

Following your comment, it is changed.

Page 25804, lines 24: I don’t know what the term urban-induced/-modified means. Induced and modified seem redundant.

The urban-induced circulation means a local circulation induced by a city itself, and the “urban-modified” means that a city modifies pre-existing local circulations that can develop without a city such as sea-breeze and valley-breeze circulations. In this study, we consider the two kinds of local circulations separately.

Page 25804, lines 28-29: Change to “In the NO-URBAN simulation, the reduction in O<sub>3</sub> by dry deposition and chemical processes in the surface layer is compensated by the downward vertical mixing of O<sub>3</sub> from the upper layers. The near-surface removal of O<sub>3</sub> by dry deposition is less in the URBAN simulation than the NO-URBAN simulation.”



Following your comment, those sentences are changed.

Page 25812, line 7: Change “Under the fair weather condition” to “Under fair weather conditions”.

In the present study, we conducted a case study under a specific weather condition. So, “the fair weather condition” indicates the specific weather condition that we considered. For this reason, we would like to retain the phrase.

Page 25813, lines 13-15 This sentence seems to be redundant with the phrase in the previous sentence on fair weather conditions.

Following your comment, the sentence is deleted.

References: I noticed at least one reference cited in the text that was not included in the reference list. The authors should check all references to make sure they are properly cited.

We have checked all the references cited and the reference list.

Table B1. Daily-, daytime-, and nighttime-averaged near-surface O<sub>3</sub> concentration averaged over the urban analysis area in the baseline simulation (denoted by Base (YSU)) that is identical to the URBAN simulation, in the simulation using the MYJ PBL scheme (denoted by MYJ), in the simulations in which the NO<sub>x</sub> emissions are decreased by 20% and increased by 20% (denoted by NO<sub>x</sub>0.8 and NO<sub>x</sub>1.2, respectively), in the simulations in which the VOC emissions are decreased by 20% and increased by 20% (denoted by VOC0.8 and VOC1.2, respectively), and in the NO-URBAN simulation (denoted by Base NO-URBAN). Four additional simulations in which the urban areas are replaced with cropland areas are performed under  $\pm 20\%$  changes in NO<sub>x</sub> and VOCs emissions (denoted by NO<sub>x</sub>0.8 NO-URBAN, NO<sub>x</sub>1.2 NO-URBAN, VOC0.8 NO-URBAN, VOC1.2 NO-URBAN). The concentration unit is ppb.

	daily average	daytime average	nighttime average
Base (YSU)	44	61	23
MYJ	38	58	12
NO <sub>x</sub> 0.8	51	70	26
NO <sub>x</sub> 1.2	37	51	19
VOC0.8	40	55	21
VOC1.2	47	66	24
Base NO-URBAN	30	48	7
NO <sub>x</sub> 0.8 NO-URBAN	37	59	9
NO <sub>x</sub> 1.2 NO-URBAN	24	38	5
VOC0.8 NO-URBAN	27	43	6
VOC1.2 NO-URBAN	33	53	7

Table B2. The CMAQ default concentration profiles for the relevant species at the four boundaries. The levels of the 1<sup>st</sup>, 8<sup>th</sup>, and 15<sup>th</sup> layers correspond to ~17, ~580, and ~1500 m, respectively. The concentration unit is ppb. The description of the lumped VOC groups can be found in Carter (2000).

	West boundary			North boundary			East boundary			South boundary		
	1 <sup>st</sup> layer	8 <sup>th</sup> layer	15 <sup>th</sup> layer	1 <sup>st</sup> layer	8 <sup>th</sup> layer	15 <sup>th</sup> layer	1 <sup>st</sup> layer	8 <sup>th</sup> layer	15 <sup>th</sup> layer	1 <sup>st</sup> layer	8 <sup>th</sup> layer	15 <sup>th</sup> layer
O <sub>3</sub>	3.5×10 <sup>1</sup>	4.1×10 <sup>1</sup>	4.6×10 <sup>1</sup>	3.5×10 <sup>1</sup>	3.6×10 <sup>1</sup>	4.3×10 <sup>1</sup>	3.0×10 <sup>1</sup>	3.6×10 <sup>1</sup>	4.3×10 <sup>1</sup>	3.0×10 <sup>1</sup>	3.6×10 <sup>1</sup>	4.3×10 <sup>1</sup>
NO	8.3×10 <sup>-2</sup>	8.3×10 <sup>-2</sup>	7.1×10 <sup>-2</sup>	8.3×10 <sup>-2</sup>	7.1×10 <sup>-2</sup>	3.0×10 <sup>-2</sup>	0	0	0	1.0×10 <sup>-2</sup>	7.1×10 <sup>-3</sup>	0
NO <sub>2</sub>	1.7×10 <sup>-1</sup>	1.7×10 <sup>-1</sup>	1.4×10 <sup>-1</sup>	1.7×10 <sup>-1</sup>	1.4×10 <sup>-1</sup>	6.0×10 <sup>-2</sup>	1.0×10 <sup>-2</sup>	7.1×10 <sup>-3</sup>	0	1.0×10 <sup>-2</sup>	7.1×10 <sup>-3</sup>	0
PAN	1.5×10 <sup>-1</sup>	1.5×10 <sup>-1</sup>	1.3×10 <sup>-1</sup>	1.0×10 <sup>-1</sup>	8.6×10 <sup>-2</sup>	5.0×10 <sup>-2</sup>	1.5×10 <sup>-2</sup>	1.5×10 <sup>-2</sup>	1.5×10 <sup>-2</sup>	1.5×10 <sup>-2</sup>	1.5×10 <sup>-2</sup>	1.5×10 <sup>-2</sup>
HCHO	4.0×10 <sup>-1</sup>	4.0×10 <sup>-1</sup>	4.0×10 <sup>-1</sup>	2.5×10 <sup>-1</sup>	2.5×10 <sup>-1</sup>	2.4×10 <sup>-1</sup>	2.5×10 <sup>-1</sup>	2.5×10 <sup>-1</sup>	2.4×10 <sup>-1</sup>	2.5×10 <sup>-1</sup>	2.5×10 <sup>-1</sup>	2.4×10 <sup>-1</sup>
ALK1	1.0×10 <sup>-1</sup>	1.0×10 <sup>-1</sup>	9.3×10 <sup>-2</sup>	1.0×10 <sup>-1</sup>	9.4×10 <sup>-2</sup>	6.6×10 <sup>-2</sup>	1.0×10 <sup>-1</sup>	1.0×10 <sup>-1</sup>	9.1×10 <sup>-2</sup>	1.0×10 <sup>-1</sup>	1.0×10 <sup>-1</sup>	9.1×10 <sup>-2</sup>
ALK2	4.0×10 <sup>-2</sup>	4.0×10 <sup>-2</sup>	3.7×10 <sup>-2</sup>	2.0×10 <sup>-2</sup>	1.9×10 <sup>-2</sup>	1.3×10 <sup>-2</sup>	6.0×10 <sup>-3</sup>	6.0×10 <sup>-3</sup>	5.4×10 <sup>-3</sup>	6.0×10 <sup>-3</sup>	6.0×10 <sup>-3</sup>	4.8×10 <sup>-3</sup>
ARO1	1.0×10 <sup>-2</sup>	5.0×10 <sup>-3</sup>	4.4×10 <sup>-3</sup>	1.0×10 <sup>-3</sup>	1.0×10 <sup>-3</sup>	1.0×10 <sup>-3</sup>	0	0	0	0	0	0
ARO2	3.0×10 <sup>-3</sup>	1.5×10 <sup>-3</sup>	4.0×10 <sup>-4</sup>	2.0×10 <sup>-4</sup>	1.7×10 <sup>-4</sup>	7.1×10 <sup>-5</sup>	0	0	0	0	0	0
CO	8.0×10 <sup>1</sup>	8.0×10 <sup>1</sup>	8.0×10 <sup>1</sup>	8.0×10 <sup>1</sup>	8.0×10 <sup>1</sup>	7.7×10 <sup>1</sup>	8.0×10 <sup>1</sup>	8.0×10 <sup>1</sup>	7.9×10 <sup>1</sup>	7.0×10 <sup>1</sup>	7.0×10 <sup>1</sup>	7.0×10 <sup>1</sup>

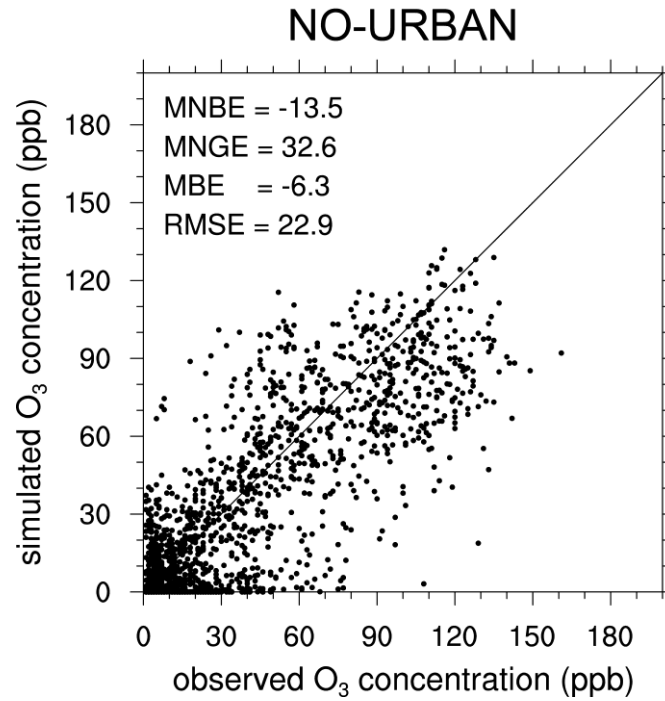


Fig. B1. Scatter diagram of observed O<sub>3</sub> concentrations and simulated O<sub>3</sub> concentrations on 24 June 2010 in the NO-URBAN simulation. The MNBE and MNGE refer to the mean normalized bias error and mean normalized gross error, respectively, and their units are %. The MBE and RMSE refer to the mean bias error and root-mean-square error, respectively, and their units are ppb.

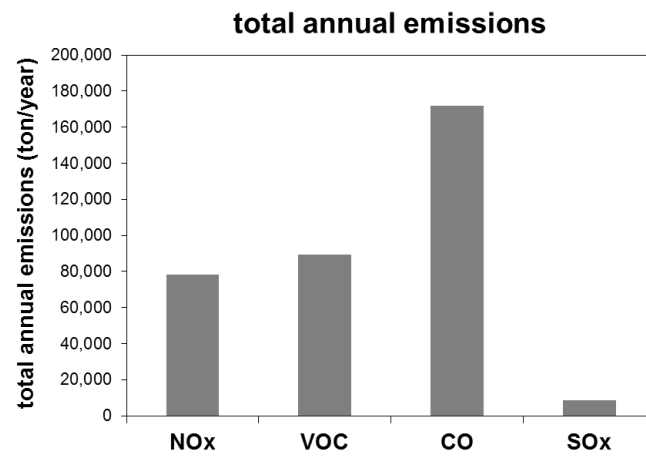


Fig. B2. Total annual emissions of NO<sub>x</sub>, VOC, CO, and SO<sub>x</sub> (= SO<sub>2</sub> + SO<sub>3</sub>) in 2008 in Seoul.