Response to Anonymous Referee #1

We thank the reviewer for the valuable and helpful comments. We believe that addressing the issues raised by the reviewer will considerably improve the manuscript. Please see our reply to each comment below.

The paper tackles a timing issue, as efforts are spent in Europe and North America, and now China, to identify and quantify parameters for assessing quality of life in urbanized areas. Air pollution is certainly one major actor. The paper is sound and of interest for the readership of ACP and I suggest the editor to accept it for publication. A few amendments are proposed hereafter.

1. Before any technical comment I invite the authors to a rigorous editing of the manuscript in all of its parts. In general the paper is hard to follow and any effort spent to simplify it would be beneficial. There are several instances where phrasing is confusing and wording un-precise. Most notably, the introduction is a bit dispersive. I think it could be shortened to a half of its current length without loss of information. Please keep only the information that is needed for putting your work into context.

We have intensively modified and shortened the Introduction section to make it more clear and brief. In particular, we substantially shortened the description on how meteorology changes influence air quality, and focused more on urbanization induced air quality changes (see below). There are also other emendations on the phrasing of the text, please refer to the manuscript.

"Based on these urban canopy schemes, a series of modeling studies have investigated the effects of urban land-use changes on regional climate and air quality. Some key climatic effects of urbanization, e.g., an increase in mean surface temperature and PBL height, and decrease in humidity and wind speed, have been captured (e.g. Wang et al., 2012; Wang et al., 2013; Yang et al., 2012; Zhang et al., 2010), which in turn influence the concentrations of pollutants even if the anthropogenic emissions are held constant (Civerolo et al., 2007; Wang et al., 2009; Yu et al., 2012). For instance, Kallos et al. (1993) indicated that land surface conditions play an important role in the development of local circulation and planetary boundary layer (PBL) depth, and could govern the dispersal, transformation, and eventual removal of airborne pollutants. In addition, Ryu et al. (2013) found that the prevailing urban breeze in the afternoon brought O3-rich and biogenic VOC-rich air masses from surrounding mountainous areas to the high-NOx urban regions, resulting in a very high ozone episode in the Seoul metropolitan area..."

2. Further, please try to keep the use of acronyms to a minimum, otherwise the flow of the text is hard to follow and readers are discouraged. If you can't reduce them, consider adding a table.

We have removed the acronyms of "LUF", "LUIND", "YRD", "PBLH", "T2", "RH2" and "W10" throughout the text. We also added a new table in the manuscript to explain all acronyms necessary (see below).

Acronyms	Description
LOCAL cells	the newly urbanized cells in each urban expansion scenario
ADJACENT cells	non-urbanized cells neighboring the LOCAL cells
ADVH	horizontal advection
ADVZ	vertical advection
ADV	the sum of horizontal and vertical advection
EMISS	Emissions
DRYDEP	dry deposition
DIFF	turbulent diffusion
VMIX	the sum of dry deposition and turbulent diffusion
CONV	Convection
CHEM	gas phase chemistry
CLDCHEM	cloud chemistry
AERCHEM	aerosol chemical and microphysical process
WETSCAV	wet scavenging

Table 1. List of acronyms used in this work

3. My major doubt is about the emissions kept constant under expanding urbanization scenario (if I understood it correctly). The finding of the enhanced mixing due to additional turbulence (mechanical and thermal) diluting pollutants more effectively might not hold if the emissions rose according to the urban expansion (more households, more people, more emissions). Having at least one simulation with increased emissions would add robustness to the conclusions which are otherwise confined to the limiting assumption of constant emissions. Please comment on that.

Good suggestion. We agree that the expansion of urban land is necessarily accompanied with the changes of anthropogenic emissions. To understand this emission effect, we conduct 5 additional simulations with anthropogenic emissions in the LOCAL cells of GT0 run amplified by a factor of 1.0, 1.1, 1.3, 1.6 and 2.0, respectively. As shown in Figure R1, surface O₃ concentrations over land in all emission scenarios are larger than the BASE case (Please refer to the perturbation of southerly/southeasterly wind in Figure 9 of the manuscript). However, CO, EC and PM_{2.5} share a different pattern that the diluting effects of urban land could be offset only if the emission augment is high enough. Figure R2 shows the perturbation of surface concentrations increase nearly linearly with increased emissions for CO, EC and PM_{2.5}. Urban land expansion (i.e. GT0) induced CO decrease keeps in both type cells until emission augment factor is larger than 40%. For EC and PM_{2.5}, even

more emission increase (>50%) is needed to compensate the dilution effect of urban land expansion. For O_3 , urbanization induced surface concentration perturbations do not change linearly with emissions, mainly due to the complexity of nonlinear ozone chemistry. Changes in vertical profiles of O_3 , CO, EC and PM_{2.5} concentrations are shown in Figure R3. The main feature is that, as emission increases, all species increase consistently above the near surface layers.

This study mainly focused on understanding the role of urban land forcing in impacting the advection, turbulent mixing and dry/wet removal of pollutants. Emission changes in the newly urbanized areas are subject to large uncertainties in China, since a lot of new buildings in the urban fringe are vacant. Therefore, we summarized above discussion in the supplementary materials. We will discuss this issue in detail (i.e., considering the effects of both the land use and emission changes on air quality) in the follow up studies.



Figure R1. The surface concentration changes (only cells exceeding the 95% significance level are shown) of CO, EC, O_3 and $PM_{2.5}$ in five emission scenarios in which all anthropogenic emissions in LOCAL cells of GTO run are increased by 0%, 10%, 30%, 60% and 100%, respectively, compared with the BASE run in July of 2010. Grey circles indicate urban areas in the BASE run; black crosses indicate LOCAL cells in GTO.



Figure R2. The mean normalized perturbation of surface concentrations of CO, EC, O_3 and PM_{2.5} over domain-wide LOCAL and ADJANCENT cells in five emission scenarios in which all anthropogenic emissions only in LOCAL cells of GT0 run are increased by 0%, 10%, 30%, 60% and 100%, respectively, compared with BASE run in July of 2010.



Figure R3. The mean vertical profile of CO, EC, O_3 and $PM_{2.5}$ over domain-wide LOCAL(top four plots) and ADJACNET(bottom four plots) cells in urbanization scenario of BASE and GT0(all anthropogenic emissions only in LOCAL cells are increased by 0%, 10%, 30%, 60% and 100%, respectively).

4. The authors might consider adding a sentence in the conclusion section conveying the results to a message to urban planner/policy makers so to provide scientific evidence in support of decision making.

We have added a concluding remark in support of decision making for urban planning at the end of Conclusion section:

"Above analysis revealed a nonnegligible and unique role of urban land forcing in the advection, turbulent mixing and dry/wet removal of pollutants, and indicated that dense urbanization has a moderate dilution effect on surface primary airborne contaminants, but may intensify severe haze and ozone pollution if local emissions are not well controlled. Further studies should consider changes in both the land use (using of a more complicated and advanced urban canopy scheme) and emissions simultaneously to better evaluate the potential environmental influence of any urbanization campaign."

Minor editing

1. ABSTRACT Line 15. 'response of meteorology'. Please be more specific Line 20. 'in the square of NULC'. Please clarify and try to avoid acronyms in the abstract. The abstract should be self-explanatory Line 23. 'IPR results', of what? Line 25. Unclear. 'determining the changes of the simulated vertical profiles' is that what you mean?

We have modified the Abstract in the manuscript according to the reviewer's suggestion.

"...Sensitivity tests show that the responses of pollutant concentrations to the spatial extent of urbanization are nearly linear near the surface, but nonlinear at higher altitudes. Over eastern China, each 10% increase in nearby urban land coverage on average leads to a decrease of approximately 2% in surface concentrations for CO, EC, and PM_{2.5}, while for O₃ an increase of about 1% is simulated. At 800 hPa, pollutants' concentrations tend to increase even more rapidly with increase in nearby urban land emerge, the influence of urban expansion on meteorology and air pollution would be significantly amplified. IPR analysis reveals the contribution of individual atmospheric processes to pollutants' concentration changes. It indicates that, for primary pollutants, the enhanced sink (source) caused by turbulent mixing and vertical advection in the lower (upper) atmosphere could be a key factor in changes to simulated vertical profiles...."

2. INTRODUCTION Line 18. Please add 'Britter and Hanna, 2003' to the references there.

Good suggestion. We have added Britter and Hanna (2003) in the revised manuscript.

3. The sentence 'To date there are 4 urban canopy schemes' is too strong and inaccurate. There exist, of course, more schemes (Di Sabatino et al., 2008; Solazzo et al., 2010; Harman et al., 2004; Coceal, O., Belcher 2004; just a few examples). You might say that not all of them have been implemented into regional models (possibly, not sure) or that you want to discuss only four among the most popular ones. Pg 10303, line 1. ' to simulate urban climate' or to account for the effects of urban areas to local climate?

Yes, so far we focused on the four urban canopy schemes that had been implemented in the WRF modeling framework. We have modified related part in Introduction:

"Up to now, a number of urban canopy schemes have been developed (e.g., Coceal and Belcher, 2004; Di Sabatino et al., 2008; Harman et al., 2004; Luhar et al., 2014; Solazzo et al., 2010; Trusilova et al., 2013; Wang et al., 2011). Among which four schemes with different complexities have been implemented in the mesoscale meteorological model (WRF) to account for the effects of urban areas on urban climate, namely Bulk..."

4. Line 14. ' Urban air pollution meteorology'. Please clarify.

We have removed this phrasing (originally referring to the relationship between the meteorology conditions and the evolution of air quality).

5. METHODOLOGY Line 6, pg 10306. '...and other secondary pollutants levels' Line 7. '100 * 100 grid'. you mean cells? Line 7. 'horizontal resolution'. Please change to 'horizontal grid spacing' throughout the text. The model resolution is the scale of the resolved processes.

We have modified Methodology in the manuscript according to the reviewer's suggestion.

"...We focus on summertime air quality because of the high ozone and other secondary pollutants levels. The modeling framework is constructed on a single domain of 100×100 cells with a 10 km horizontal grid spacing..."

6.SECTION 3 Please add a measure of variability, like the ratio of the standard deviation of the measurements to the standard deviation of the model. PM2.5 as simulated by WRF/Chem suffers from too low variability and underestimation (as well as for many other regional air quality transport models) due to

unresolved/missing processes and inaccurate inventories (Im et al., 2014; Solazzo et al., 2012). Please comment on that.

In this study, the daily mean observed and modeled $PM_{2.5}$ concentrations are, respectively, 47.4 (± 22.7) and 51.3 (±29.0) µg/m³ in NJ sites, and are 41.8 (±12.3) and 44.1 (±21.0) µg/m³ in the SH_PD site. Unlike the findings that the modeling of PM_{2.5} suffered from too low variability and underestimation in Europe and North America during AQMEII campaign (Im et al., 2014; Solazzo et al., 2012), the modeling of daily PM_{2.5} in above two sites of NJ and SH overestimated the PM_{2.5} and its variability a bit. We have added this discussion in the beginning of Section 3.

"Recent evaluation of the ensemble of regional air quality models in the Air Quality Model Evaluation International Initiative (AQMEII) indicated that, modeling of PM_{2.5} suffered from too low variability and underestimation (Im et al., 2014; Solazzo et al., 2012). However, in this study the daily mean observed and modeled PM_{2.5} concentrations in NJ sites are 47.4 ±22.7 and $51.3\pm29.0 \ \mu\text{g/m}^3$, while in SH_PD site are 41.8 ± 12.3 and $44.1\pm21.0 \ \mu\text{g/m}^3$, respectively. Both the mean and daily variability (indicated by the ratio of the standard deviation of the measurements to the standard deviation of the model) of PM_{2.5} concentrations are overestimated a bit."

7. TABLE 1 There is something I don't understand with this table. Is it about only one station? What is missing in the header? You use hourly values for the statistics (for the whole month?) and daily for the figures, right? Please specify it in the text.

Yes, the statistics in Table 1 are based on the hourly data for the whole month. Since this table doesn't contain much information, we removed this table and described the evaluation results in the Section 3.

"The modeled and observed hourly concentrations of O_3 , CO, and $PM_{2.5}$ at above five sites are also compared for the whole month. WRF/Chem generally captures the diurnal variation of surface O_3 well, (i.e., R: 0.74, NMB: 6.7%, NME: 34.1 %, and I: 0.86). The model also reproduces the hourly surface burden of $PM_{2.5}$ and CO, with NMEs of 63.4% and 52.6%, respectively."

References

Britter, R., and Hanna, S.: Flow and dispersion in urban areas, Annual Review of Fluid Mechanics, 35, 469-496, 2003.

Im, U., Bianconi, R., Solazzo, E., Kioutsioukis, I., Badia, A., Balzarini, A., Baró, R., Bellasio, R., Brunner, D., Chemel, C., Curci, G., Denier van der Gon, H., Flemming, J., Forkel, R., Giordano, L., Jiménez-Guerrero, P., Hirtl, M., Hodzic, A., Honzak, L., Jorba, O., Knote, C., Makar, P. A., Manders-Groot, A., Neal, L., Pérez, J. L., Pirovano, G., Pouliot, G., San Jose, R., Savage, N., Schroder, W., Sokhi, R. S., Syrakov, D., Torian, A., Tuccella, P., Wang, K., Werhahn, J., Wolke, R., Zabkar, R., Zhang, Y., Zhang, J., Hogrefe, C., and Galmarini, S.: Evaluation of operational online-coupled regional air quality models over Europe and North America in the context of AQMEII phase 2. Part II: Particulate matter, Atmospheric Environment, http://dx.doi.org/10.1016/j.atmosenv.2014.08.072, 2014.

Solazzo, E., Bianconi, R., Pirovano, G., Matthias, V., Vautard, R., Moran, M. D., Wyat Appel, K., Bessagnet, B., Brandt, J., Christensen, J. H., Chemel, C., Coll, I., Ferreira, J., Forkel, R., Francis, X. V., Grell, G., Grossi, P., Hansen, A. B., Miranda, A. I., Nopmongcol, U., Prank, M., Sartelet, K. N., Schaap, M., Silver, J. D., Sokhi, R. S., Vira, J., Werhahn, J., Wolke, R., Yarwood, G., Zhang, J., Rao, S. T., and Galmarini, S.: Operational model evaluation for particulate matter in Europe and North America in the context of AQMEII, Atmospheric Environment, 53, 75-92, <u>http://dx.doi.org/10.1016/j.atmosenv.2012.02.045</u>, 2012.