

Response to Anonymous Referee #3

We thank the reviewer for the valuable and helpful comments. We have extensively revised the manuscript based on these comments and suggestions, which greatly improved the manuscript. Please see our reply to each comment below.

Using WRF-CHEM model, authors of this manuscript assessed quantitatively the effects of urbanization to urban air quality with focus on eastern China where rapid expansion of urban area has been taking place over last several decades. Urbanization can increase anthropogenic emissions of criteria air pollutants and alter dynamics and thermodynamics of air parcels and atmospheric contaminants in the atmosphere. This paper dealt with the response of urban air pollutants to changes in atmospheric turbulence and advection induced by changes in underlying urban surfaces. Results reported in this study fill knowledge gaps in understanding redistribution of air pollutants forced by urbanization from a dynamic perspective. I recommended publication in ACP after following comments are addressed.

1. As the authors mentioned, anthropogenic emission were obtained from the Multi-resolution inventory for China. Given that this model simulation used fixed surface emissions, it is not clear if emissions in 2010 from the MEIC were the fixed emissions used in authors' modeling exercises of 2008 through 2012. In Model evaluation section (3) modeled atmospheric level of O₃, CO, and PM_{2.5} were verified against monitored data in 2008, 2009, and 2012, respectively. Were these modeled concentrations all derived from fixed emissions in 2010 as well?

The MEIC 2010 emissions inventory consists of emissions for each month across the year 2010. In this study, emissions are fixed at the 2010 level though simulations cover 2008-2012. When conducting model evaluation, we used the MEIC 2010 emissions of the corresponding month to match the observational data made in 2008, 2009 and 2012. To make it clear, we have added a description on the utilization of MEIC 2010 in Section 2.1:

“Anthropogenic emission data are from Multi-resolution Emission Inventory for China (MEIC) developed by Tsinghua University for the year 2010, which consists of the emission rates for each month from five sectors (agriculture, industry, power plants, residential and transportation)...We used the MEIC 2010 data of the corresponding month as input for all simulations of 2008 through 2012, ignoring the year-to-year variation in emissions.”

2. Surface wind field perturbations due to urban expansion shown in Fig. 9 seem to suggest that the effect of urbanization on wind field in eastern China is quite

significant. From my view, perturbed southeasterly winds extending from East China Sea to the east seaboard of China are in the regime of east Asian summer monsoon. The top panel of Fig. 9 shows that GT0 scenario yielded strongest perturbation as compared with other two scenarios. Does this suggest that the urban expansion in east China could increase the strength of wind field under the monsoon regime? As authors mentioned (pg 4, line 5-8), both large-scale weather patterns and land surface conditions govern the dispersal, transformation, and eventual removal of airborne pollutants. Does Fig. 9 imply a feedback of change in land use types to large-scale weather pattern?

Good suggestion. As shown in Figure R1, in BASE run, the July mean surface pressure over marine is greater than that over land, and the prevailing surface wind is southeasterly and southerly, exhibiting the feature of monsoon wind during summer. The urban land expansion could strengthen the southeasterly sea breeze over marine and near the east seaboard, due to the increased difference in thermal properties between land and sea (as the urban land is characterized with a greater heat capacity, thermal conductivity and lower albedo than natural land). However, perturbation of wind field in the inner terrestrial is more complicated, generally speaking, the replacement of natural land by urban land would reduce the local pressure and form a cyclonic convergence zone. Nonetheless, our study does not necessarily reveal the feedbacks of land use changes to large-scale weather pattern, as the boundary conditions (B.C.) for each urban land expansion scenario is fixed. The urban land forcing simulated in this study is confined to the regional scale, and how would the perturbation of mesoscale circulation impact the large-scale circulation remain uncertain, and this should be addressed in future studies. Using an integrated general circulation model, Jacobson and Ten Hoeve (2012) found that the simulated UHI effects on global warming would be greater (but of similar order) if the large-scale feedback (e.g. active sea, chemistry-climate interaction) was implemented in the modeling framework. Yet the urban land forcing on the large-scale even planetary-scale needs to be further investigated and characterized, and the mechanisms should be explained in perspective of geophysical fluid dynamics. We have added some discussion of urban land forcing on sea-land breeze in Section 5.1.

“In the GT0 run, as urban land expands, changes in horizontal (vertical) advection tend to increase (reduce) the surface concentration of all four species over the LOCAL cells, whereas the opposite is true for the ADJACENT cells. The associated surface wind field perturbations due to urban land expansion are shown in Figure 9. The urban land expansion could strengthen the southeasterly sea breeze over marine and near the east seaboard, due to the increased difference in thermal properties between land and sea (as the urban land is characterized with a greater heat capacity, thermal conductivity and lower albedo). However, perturbation of wind field in the inner terrestrial is more complicated, generally speaking, the replacement of natural land by urban land would reduce the local pressure (up to 30 Pa) and form a cyclonic

convergence zone...”

Also, the caveat of this study that confines the urban land forcing to the regional scale has been addressed in the last paragraph of Section 5.2.

“The caveats of this study are as follows: 1) The forcing of urban land expansion on the atmospheric environment is confined to the regional scale. Feedback between mesoscale circulation and large-scale circulation, as well as inflows of airborne pollutants from outside the domain of interest, has been ignored...”

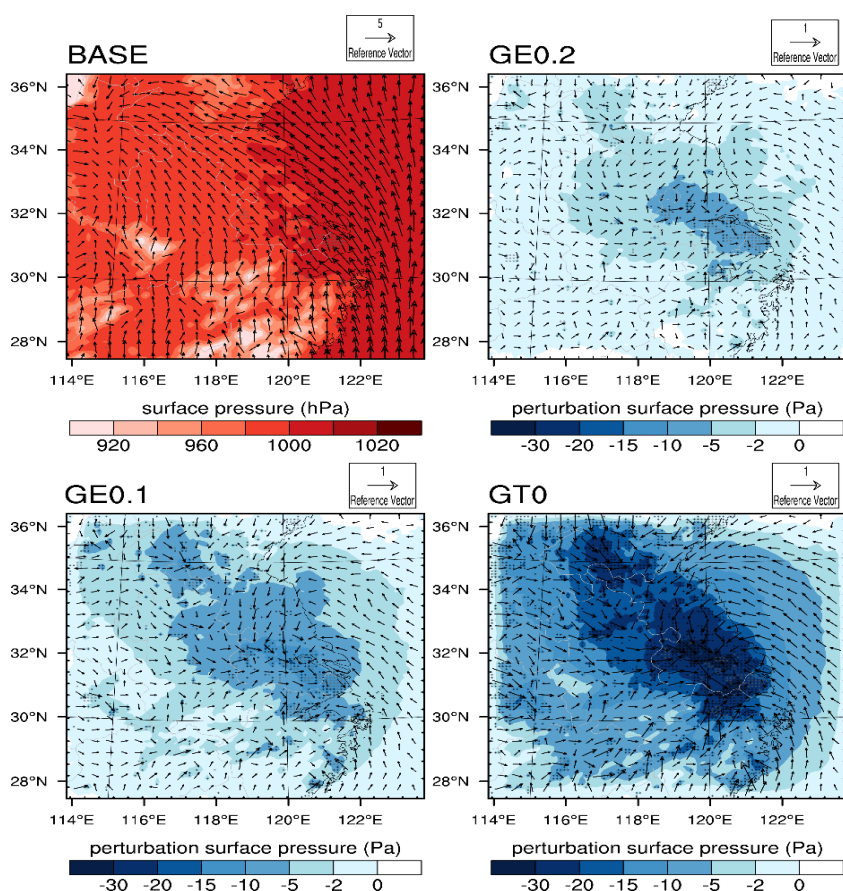


Figure R1. Five-year mean July surface pressure and wind field in BASE run and the perturbation in GE0.2, GE0.1, GT0 runs. Grey circles indicate the locations of urban cells in the BASE run; black crosses indicate the locations of newly urbanized areas in GE0.2, GE0.1, and GT0 runs. Reference vector for BASE run is 5 m/s, while for other runs are 1 m/s.

3. Perhaps I missed the discussions on interactions between targeted species at LOCAL and ADJACENT cells. If urban expansion reduced atmospheric level of these species at LOCAL cells, partly due to urbanization-induced outward horizontal advection, featured by lower concentrations over LOCAL cells than those over the

ADJACENT regions (pg 13, line 10-11), concentrations over the ADJACENT cells might also flow towards LOCAL cells to compensate the lost mass of species at LOCAL cells unless an equilibrium of the species between LOCAL and ADJACENT cells is reached.

We agree with the reviewer that most newly urbanized regions in this study aggregate around the periphery of the original urban districts, and the anthropogenic emissions over LOCAL cells generally exceeded those over ADJACENT cells. As a result, concentrations of EC, CO and PM_{2.5} over LOCAL cells were higher than those over ADJACENT cells (we have corrected this typo in the revised manuscript). Based on the integrated process rate (IPR) analysis (see Figure 8 and 12 in the manuscript), the urban land forcing on LOCAL and ADJACENT cells are not isolated, but are interconnected with each other via urban heat island circulation. For the perturbed surface wind, Figure 9 in the manuscript shows convergence and uplift zones in most LOCAL grids (the direct impacts of urban land), while divergence and downlift zones in most ADJACENT grids (the indirect impacts of urban land, to guarantee the mass balance of ambient air). As indicated by the reviewer, this leads to a tendency for pollutants over the ADJACENT cells flowing towards LOCAL cells to compensate the uplift loss of species at LOCAL cells. The perturbation in advection changes follows exactly with the perturbation in wind field, and also an important factor explaining the concentration changes.

4. Pg 5, line 14, Jing-Jin-Ji, you mean Beijing-Tianjin-Hebei?

Yes, Jing-Jin-Ji does refer to Beijing-Tianjin-Hebei.

5. Pg 10, line 28-29, terrestrial O₃ level (~24-32 ppb) is almost identical to its level (~24-30 ppb) at 800 hPa. You mean 800 hPa over an urban site?

Here 800hPa refers to a much broader region over the eastern China. As shown in Figures 3 and 4, surface O₃ was found to be relatively evenly distributed (about 24-32 ppb) on the mainland of China within the domain compared with the primary pollutants. The distribution pattern for O₃ at 800 hPa is different from that at surface, and O₃ concentrations appear higher over the North China Plain (about 24-30 ppb) than in the southern domain. Unlike surface CO and EC, O₃ concentrations over urban areas do not necessarily exceed those over the surrounding rural areas at both surface and 800 hPa. We have clarified this issue in the revised manuscript:

“At 800 hPa, concentrations of CO (~40–70 ppb), EC (~0.2–0.4 μg m⁻³), O₃ (~24–30 ppb), and PM_{2.5} (~10–20 μg m⁻³) over the North China Plain appear much higher than those in the southern domain...”

6. Pg 17, line 3-12, how about humidity influence on PM_{2.5}? If urban expansion decreases relative humidity, this decline may affect PM_{2.5} formation.

Good suggestion! Yes, the humidity does impact the formation of PM_{2.5} in two ways, as simulated within the framework of aerosol module of MADE (Ackermann et al., 1998) used in this study. Firstly, water molecules act as reactants and solvents in gas phase/particle partitioning, thus impact the formation of both secondary inorganic aerosols, as described by MARS scheme (Saxena et al., 1986) and secondary organic aerosols, as described by SORGAM scheme (Schell et al., 2001). In addition, the cloud chemistry in MADE simulates the formation of aerosols in clouds through a series of aqueous-phase reactions (Baklanov et al., 2008; Mlawer et al., 1997). Figure R2 shows that, besides primary pollutants, the emerged new urban land also relocate the gaseous and liquid water. Over newly urbanized areas, the mixing ratios of water vapor and cloud water decrease near the surface while increase above (about 1.5 km). The production of PM_{2.5} through cloud chemistry increase (decreases) exactly where the humidity increases (decreases). We have added this discussion on how changes in humidity influence PM_{2.5} formation in the revised manuscript (see the fifth paragraph of Section 5.2 or below).

“...The formation of PM_{2.5} through gas phase/particle partitioning and cloud chemistry is also influenced by the relocation of humidity, as simulated by the MADE/SORGAM scheme (Please refer to the supplementary materials for details). As shown in Figure S11 in the supplementary materials, the production of PM_{2.5} through cloud chemistry increase (decreases) exactly where the humidity increases (decreases).”

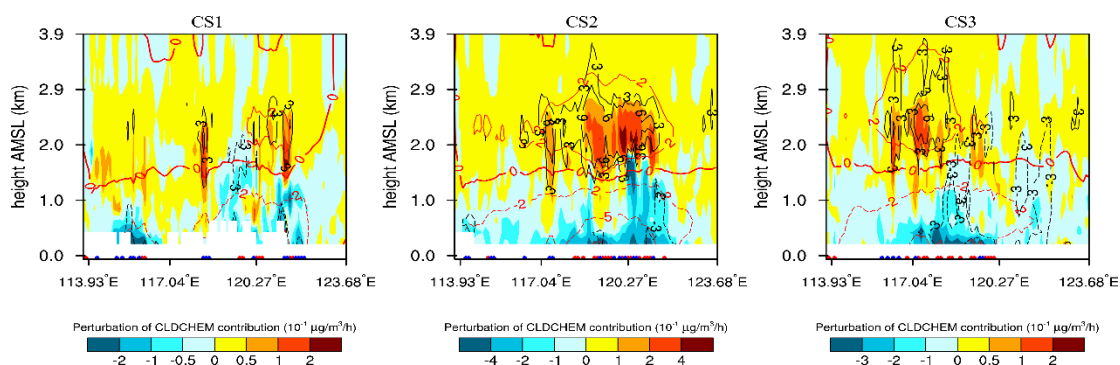


Figure R2. Distribution of 5-year July mean perturbations (GT0 minus BASE) of PM_{2.5} production from cloud processes (color, μg/m³/h), relative humidity (red line, %) and cloud water mixing ratio (black line, mg/kg) in the cross-sections of CS1, CS2, and CS3. Red and blue dots indicate the longitudes of LOCAL cells in the GT0 run along the cross-section lines and adjacent areas, respectively.

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