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).

Table 1. List of acronyms used in this work

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

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 O_3 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 averaged over domain-wide LOCAL and ADJACENT cells. Concentration perturbations 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.

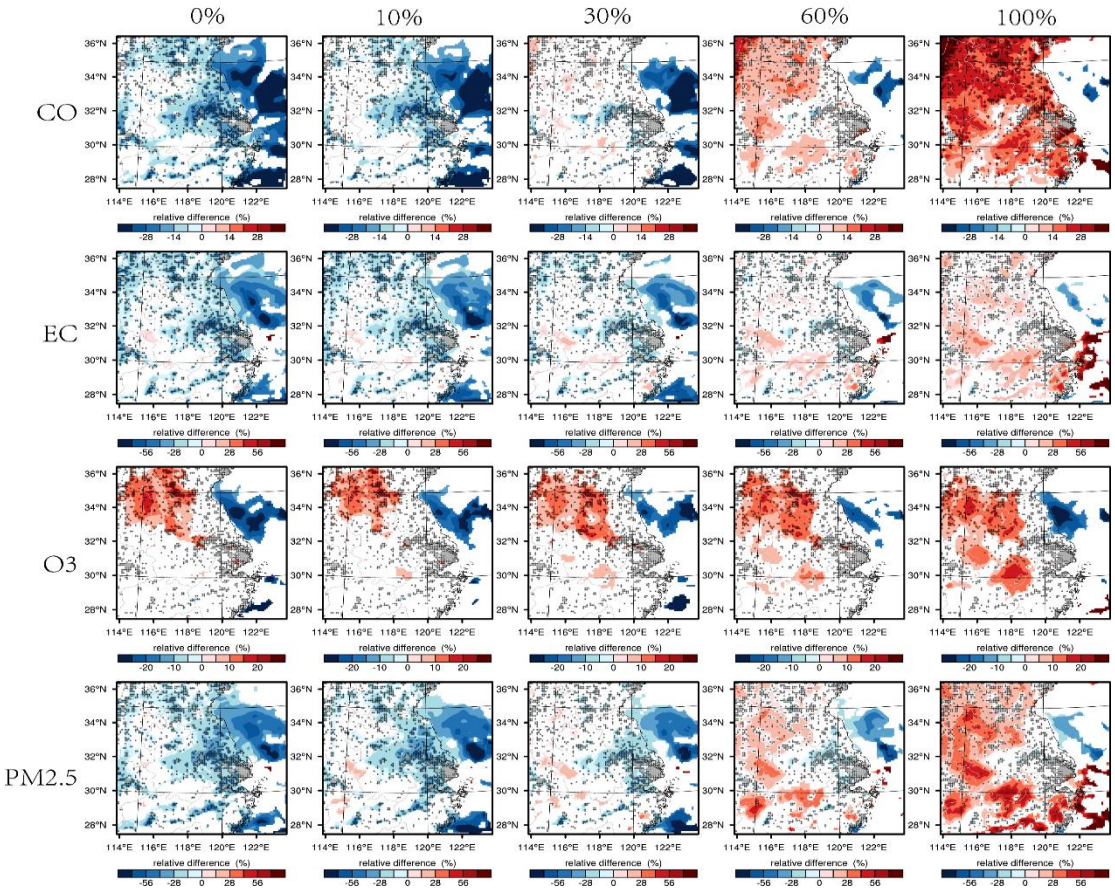

Figure R1. The surface concentration changes (only cells exceeding the 95% significance level are shown) of CO, EC, O_3 and $PM_{2.5}$ in five emission scenarios in which all anthropogenic emissions in LOCAL cells of GT0 run are increased by 0%, 10%, 30%, 60% and 100%, respectively, compared with the BASE run in July of 2010. Grey circles indicate urban areas in the BASE run; black crosses indicate LOCAL cells in GT0.

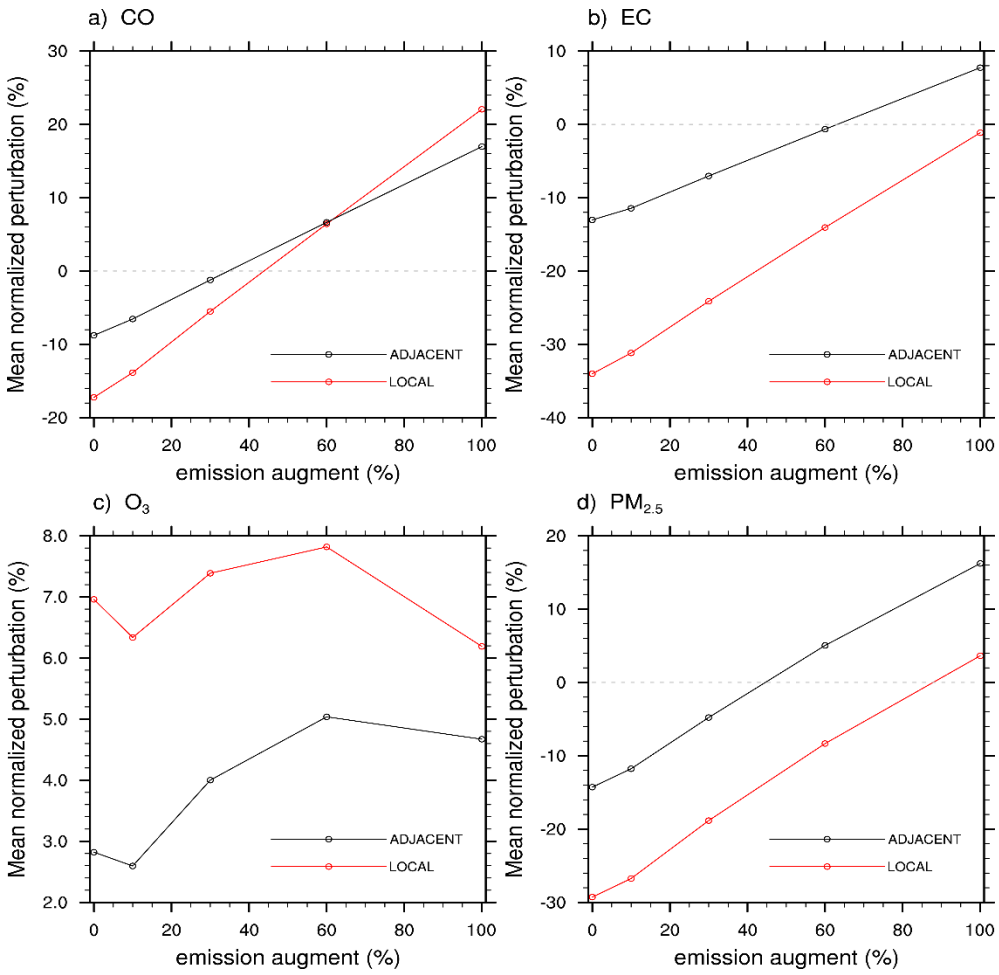

Figure R2. The mean normalized perturbation of surface concentrations of CO, EC, O_3 and $PM_{2.5}$ over domain-wide LOCAL and ADJANCENT cells in five emission scenarios in which all anthropogenic emissions only in LOCAL cells of GT0 run are increased by 0%, 10%, 30%, 60% and 100%, respectively, compared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

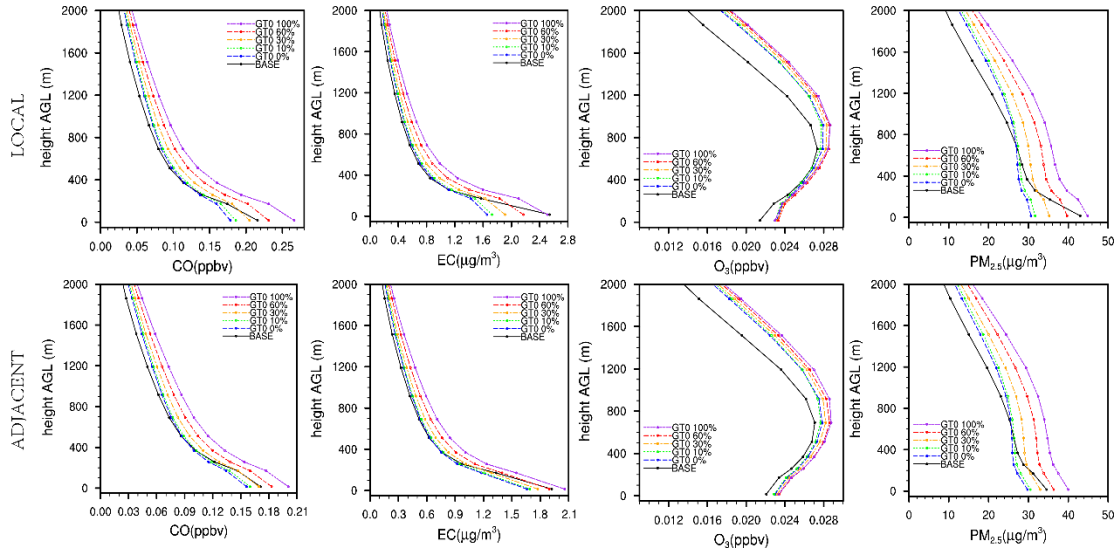

Figure R3. The mean vertical profile of CO, EC, O_3 and $PM_{2.5}$ over domain-wide LOCAL(top four plots) and ADJACNET(bottom four plots) cells in urbanization scenario of BASE and GT0(all anthropogenic emissions only in LOCAL cells are increased by 0%, 10%, 30%, 60% and 100%, respectively).

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 O_3 an increase of about 1% is simulated. At 800 hPa, pollutants' concentrations tend to increase even more rapidly with increase in nearby urban land coverage. This indicates that as large tracts of new 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 ($\pm$ 22.7) and 51.3 ($\pm$ 29.0) $\mu g/m^3$ in NJ sites, and are 41.8 ($\pm$ 12.3) and 44.1 ($\pm$ 21.0) $\mu g/m^3$ 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±29.0 $\mu g/m^3$, while in SH_PD site are 41.8±12.3 and 44.1±21.0 $\mu g/m^3$, 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."

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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 PM2.5 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…"

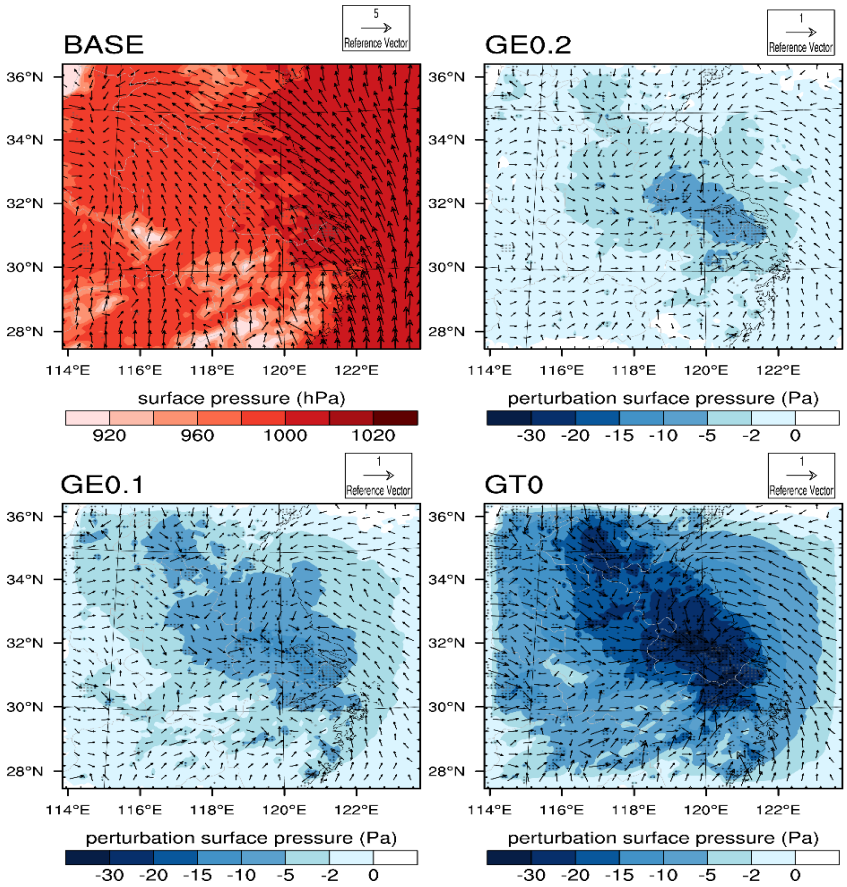

Figure R1. Five-year mean July surface pressure and wind field in BASE run and the perturbation in GE0.2, GE0.1, GT0 runs. Grey circles indicate the locations of urban cells in the BASE run; black crosses indicate the locations of newly urbanized areas in GE0.2, GE0.1, and GT0 runs. Reference vector 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 ADJACNET 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 O_3 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 O_3 at 800 hPa is different from that at surface, and O_3 concentrations appear higher over the North China Plain (about 24-30 ppb) than in the southern domain. Unlike surface CO and EC, O_3 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}), O_3 (~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 PM2.5? If urban expansion decreases relative humidity, this decline may affect PM2.5 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)."

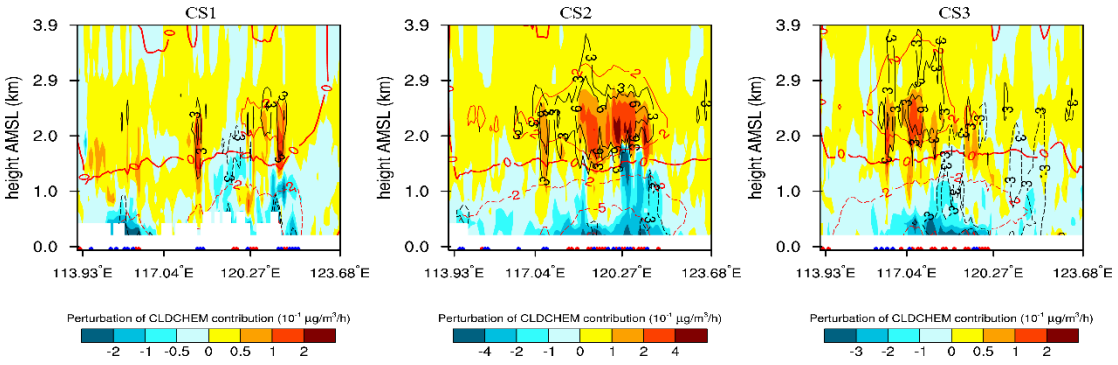

Figure R2. Distribution of 5-year July mean perturbations (GT0 minus BASE) of $PM_{2.5}$ production from cloud processes (color, µg/m^3/h), relative humidity (red line, %) and cloud water mixing ratio (black line, mg/kg) in the cross-sections of CS1, CS2, and CS3. Red and blue dots indicate the longitudes of LOCAL cells in the GT0 run along the cross-section lines and adjacent areas, respectively.

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Effects of Urban Land Expansion on the Regional Meteorology and Air Quality of Eastern China

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Abstract

Rapid urbanization throughout Eastern China is imposing an irreversible effect on local climate and air quality. In this paper, we examine the response of a range of meteorological and air quality indicators to urbanization. Our study uses the Weather Research and Forecasting model coupled with Chemistry (WRF/Chem) to simulate the climate and air quality impacts of four hypothetical urbanization scenarios with fixed surface pollutant emissions during the month of July from 2008 to 2012. An improved integrated process rate (IPR) analysis scheme is implemented in WRF/Chem to investigate the mechanisms behind the forcing–response relationship at the process level. For all years, as urban land area expands, concentrations of CO, elemental carbon (EC), and particulate matter with aerodynamic diameter less than 2.5 microns ($PM_{2.5}$) tend to decrease near the surface (below ~500 m), but increase at higher altitudes (1–3 km), resulting in a reduced vertical concentration gradient. On the other hand, the O_3 burden averaged over all newly urbanized grid cells consistently increases from the surface to a height of about 4 km. Sensitivity tests show that the responses of ~~meteorology factors (e.g. surface temperature, and relative humidity, and planetary boundary layer height) and~~ 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 ~~(NULC)~~ on average leads to a decrease of approximately 2% in surface concentrations for CO, EC, and $PM_{2.5}$, while for O_3 an increase of about 1% is simulated. At 800 hPa, pollutants' concentrations tend to ~~each 10%~~ increase even more rapidly with increase in ~~the square of~~ ~~NULC~~nearby urban land coverage ~~enhances air pollution concentrations by 5–10%, depending on species~~. This indicates that as large tracts of new urban land emerge, the influence of urban expansion on meteorology and air pollution would be significantly amplified. IPR ~~results~~ analysis reveals the contribution~~s~~ of ~~different~~individual atmospheric processes to pollutants' concentration changes~~, and~~. 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. The evolution of secondary pollutants is further influenced by the upward relocation of precursors that impact gas phase chemistry for O_3 and aerosol processes for $PM_{2.5}$. Our study indicates 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.

Keywords: Urbanization, Air pollution, Process analysis, Linearity, Aerosol, Ozone

1 Introduction

Urbanization refers to the growth of urban populations and the vast expansion of urban areas. According to the 2011 Revision of the United Nations (UN) World Urbanization Prospects, the global proportion of the population living in urban areas is likely to increase to 68% (about 6.2 billion) by 2050, and the urban population in less developed regions will almost double from 2.7 billion in 2011 to 5.1 billion in 2050 (Heilig, 2012). The environmental side-effects of urbanization, such as inadvertent climate modification (Changnon, 1992) and air quality degradation (Mage et al., 1996), remain an important research topic with societal relevance.

The radiative, thermal, hydrologic, and aerodynamic properties of urban land surfaces are distinct from those of natural surfaces (e.g., forests, grassland), resulting in unique exchange processes of energy, moisture, and momentum with the ambient atmosphere and thus distinct climatic conditions in urban areas (Oke, 1987). The features of urban climate (e.g. urban heat island (UHI), wind profiles in the urban canopy layer, etc.) have been extensively observed, modeled and comprehensively reviewed (e.g. Arnfield, 2003; Kanda, 2007; Souch and Grimmond, 2006). The urban climate is characterized by multiple scales (Britter and Hanna, 2003; Fisher et al., 2006; Oke, 2006), e.g. flows in the roughness sublayer at micro-scale are not subject to Monin–Obukhov similarity relationships, whereas upper flows in the inertial layer are in equilibrium with the underlying surface, and can be described by meso-scale dynamics. Another feature of urban climatology is, heterogeneity~~,~~, namely~~i.e.~~ the high non-uniformities of roughness elements in urban areas (~~(~~e.g. impervious road and construction, green belt, etc.~~) in urban areas~~), which make it rather complicated to generalize the urban flow details from one landscape to another (Fernando et al., 2001). Factors as anthropogenic heat (Fan and Sailor, 2005), chemistry–climate feedbacks (Rosenfeld, 2000), and topography could alter the characteristics of urban climatic conditions~~,~~ and the intensity of background wind speed~~ or~~. In addition, land-sea breezes could impact the structure of urban boundary layer (Fisher et al., 2006; Rotach et al., 2002) ~~and~~and modify the ventilation conditions~~ as well~~ (Ryu et al., 2013; Yoshikado and Tsuchida, 1996). ~~(Oke, 1987). Anthropogenic heat (Fan and Sailor, 2005), chemistry–climate feedbacks, and topography also contribute to the~~

formation of distinct climatic conditions in urban areas. The features and intrinsic mechanisms of the "urban climate", which are characterized by multiple scales (Britter and Hanna, 2003; Fisher et al., 2006; Oke, 2006) as well as heterogeneity in urban morphology and land cover properties, have been extensively studied. Comprehensive reviews of urban climatology (e.g. Arnfield, 2003; Kanda, 2007; Souch and Grimmond, 2006) have reported advances in recent decades in concepts such as the urban canopy layer (UCL) and roughness sublayer, observations, and numerical modeling.

To dateUp to now, a number of there are four 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 of with different complexities have been implemented in the mesoscale meteorological model (WRF) to account for the effects of urban areas on urban climate,: have been developed: namely Simple Bulk (BULK, Liu et al., 2006), Single-layer Urban Canopy Model (SLUCM, Kusaka and Kimura, 2004), Building Effect Parameterization (BEP, Martilli et al., 2002), and Building Energy Model (coupled to BEP, denoted as BEP+BEM, Salamanca et al., 2010),. These have been implemented in mesoscale meteorological models of WRF to account for the effects of urban areas to local climate simulate urban climate. The BULK scheme parameterizes the urban surface with greater heat capacity, thermal conductivity, roughness length, and lower albedo than those of natural land surfaces, rather than resolving the morphology of the urban canopy, and has been successfully employed in real-time weather forecasts (Liu et al., 2006). It could capture the features of urban synoptic conditions (Liao et al., 2014), and has been widely used for real-time meso-scale weather forecasting over urban areas (Salamanca et al., 2011). The last three schemes represent the urban geometry as street canyons with urban surfaces (i.e., walls, roofs, and roads), and the coupled mesoscale meteorological model of WRF and SLUCM model reportedly has the ability to capture the UHI features of an urban heat island (UHI) in some megacities (Cui and de Foy, 2012; Lin et al., 2011; Miao et al., 2009). However, application of these urban canopy schemes are limited by the difficulty ofrequires specifying a vast number of urban canopy parameters and initial conditions (Chen et al., 2011), which are usually difficult to set accurately and may change rapidly in developing countries. The BULK scheme could capture the features of urban synoptic conditions (Liao et al., 2014), and is still being widely used for real-time meso-scale weather forecasting over urban areas (Salamanca et al., 2011).

Based on these urban canopy schemes, ~~Efforts have been made to find the statistical relationships between Temporal Synoptic Index (TSI) and air pollution loadings (e.g. Greene et al., 1999), and the adverse meteorological conditions of severe air pollution episodes (e.g. Malek et al., 2006; Yegorova et al., 2011), indicating the significant role of meteorology in dominating the evolution of air quality. In terms of urban air pollution meteorology, As Kallos et al. (1993) indicated, large-scale weather patterns and land surface conditions, which play an important role in the development of local circulation and planetary boundary layer (PBL) depth, could govern the dispersal, transformation, and eventual removal of airborne pollutants. It is necessary we investigate However, the issues of how urban land-use changes would impact the local meteorological conditionsy, and thus the evolution of airborne contaminants, remain less understood, and are characterized by multiple scales.. However, understanding this issue is challenged by the complexities of fluid dynamics in urban environment. Air flows (fromof meso- to micro-scale) in different vertical layers above urban areas present different features. For instance, flows in the roughness sublayer are not subject to Monin-Obukhov similarity relationships, whereas upper flows in the inertial layer are in equilibrium with the underlying surface. Also, the background prevailing wind speed (threshold of 3 m s^{-1}) could impacts the structure of the urban boundary layer (Fisher et al., 2006; Rotach et al., 2002),. andFurthermore, land-sea breezes couldcan interact with UHI circulation (UHIC) to impact the ventilation conditions (Ryu et al., 2013; Yoshikado and Tsuchida, 1996). FurthermoreHowever, the high non-uniformities of roughness elements in urban areas make it rather complicated to generalize the flow details from one landscape to another (Fernando et al., 2001). Urban air quality models are increasingly being incorporated into multi-scale integrated atmospheric models (Baklanov et al., 2007; Chen et al., 2011; Zhang et al., 2012). Recently~~ a series of ~~numerical~~modeling studies have ~~utilized mesoscale meteorological models coupled with urban schemes to~~ investigated ~~how~~the effects of urban ~~urbanization induced~~ land-use changes ~~influence the~~on regional climate and air quality. ~~These studies perform sensitivity tests with varying static geographic field inputs, with urbanization represented by increments in the urban land-cover fraction.~~ Some ~~expected~~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)~~. Replacement of natural land by urban land could alter~~, which in turn influence the concentrations of pollutants even if the anthropogenic emissions are held constant (Civerolo et al., 2007; De Meij et al., 2015; Wang et al., 2009; Yu

et al., 2012)~~, but~~. 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, ~~no detailed mechanism for urban land forcing on air quality was analyzed. For instance,~~ Ryu et al. (2013) found that ~~the enhanced turbulence in the deep urban boundary layer diluted NO_x, which in turn reduced O_3 destruction in the NO_x-rich environment; while~~ the prevailing urban breeze in the afternoon ~~then~~ brought O_3-rich and biogenic ~~B~~VOC-rich air masses from surrounding mountainous areas to the high-NO_x urban regions, ~~all contributing to~~resulting in a very high ozone episode in the Seoul metropolitan area.

To date,

~~Framed on these urban canopy schemes, recently A a series of recent numerical studies have utilized mesoscale meteorological models, some of which are coupled to atmospheric chemical transport models, to investigate how urbanization induced land use changes influence the regional climate and air quality. These studies perform sensitivity tests with varying static geographic field inputs, with urbanization represented by increments in the urban land cover fraction. Some expected 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). Replacement of natural land by urban land could alter the concentrations of pollutants even if the anthropogenic emissions are held constant (Civerolo et al., 2007; Wang et al., 2009; Yu et al., 2012), but no detailed mechanism for urban land forcing on air quality was analyzed. Using MM5/CMAQ (the PSU/NCAR MM5 mesoscale meteorological model and the Community Multiscale Air Quality Model), Civerolo et al. (2007) compared the surface ozone concentrations in two land use scenarios (year 1990 and 2050) in the metropolitan region of New York City, and found that when emissions were held constant, ozone levels increased across much of the area. However, when anthropogenic emissions were increased in proportion to the urbanization process, O_3 levels increased in outer counties of the metropolitan region, but decreased in other areas. Based on the Weather Research Forecasting model coupled with Chemistry (WRF/Chem), Wang et al. (2009) simulated two land use scenarios (year 1990 and 2004) in the Pearl River Delta area. They concluded that~~

urbanization (anthropogenic emissions were held constant) decreased surface NO_x and VOC (volatile organic compounds) levels, but increased surface O_3 and NO_3 radical levels. They also noted that SOA (secondary organic compounds) levels increased in some areas, while decreasing elsewhere. Using a similar method, Yu et al. (2012) found that urbanization-induced land surface changes caused daytime CO and O_3 concentrations to increase in the Jing-Jin-Ji (JJJ) and Yangtze River Delta (YRD) areas of China. Replacement of natural land by urban land could alter the concentrations of pollutants even if the anthropogenic emissions are held constant (Civerolo et al., 2007; Wang et al., 2009; Yu et al., 2012), but no detailed mechanism for urban land forcing on air quality was analyzed.Applying the WRF/CMAQ model and a process analysis technique For instance,, Ryu et al. (2013) analyzed the dominant forcing offound that urban land: the enhanced turbulence in the deep urban boundary layer diluted NO_x, which in turn reduced O_3 destruction in the NO_x-rich environment; furthermore, while the prevailing urban breeze in the afternoon then brought O_3-rich and BVOC-rich air masses from surrounding mountainous areas to urban regions, all contributing to a high ozone episode in the Seoul metropolitan area.

To dateStill, the characteristics and intrinsic mechanisms of the forcing exerted by urban land expansion on the atmospheric environment, including the burden of both primary and secondary pollutants,, under current anthropogenic emission levels are still not well understood, particularly throughout Eastern China. Recently, the Chinese government has relaxed its one-child policy to promote the long-term balanced development of the population, and has also launched an ambitious urbanization campaign. Therefore, it is expected that China will undergo continuous urban population growth and rapid urban land expansion in the coming decades. Note that lLand-use changes caused by new urban infrastructure are usually irreversible. If urban land expansion exerts adverse forcing on the ambient environment, mitigation strategies for climate and air quality improvement would be less easily implemented and more costly.

Using WRF/Chem, a mesoscale fully coupled air quality and meteorological model, this study addresses two key questions: 1) How how sensitive are the meteorological conditions and the spatial distribution of airborne contaminants to urban land expansion? 2) What what are the intrinsic mechanisms and dominant processes that drive land-cover change urbanization induced changes in climate and atmospheric chemistry?

In section 2, Wwe present the methodology and describe the methodology and the model used in this studyin section 2, and evaluate the model results . The model evaluation is provided in Section 3. In section 4, we present the impact of urban land expansion on the distribution of key atmospheric species. In section 5, we investigate the individual processes contributing to these changes in atmospheric composition. Conclusions are provided drawn in section 6.

2 Methodology

2.1 Model description and configuration

We use WRF/Chem v3.5 (Grell et al., 2005) to simulate meteorological fields and atmospheric chemistry under four hypothetical urban land surface expansion scenarios in July for the five years from 2008 to 2012. We focus on summertime air quality because of the high ozone and other secondary pollutantsair pollution levels. The modeling framework is constructed on a single domain of 100 × 100 gridcells with a 10 km horizontal resolutionhorizontal grid spacing, and covers nine provinces in eastern and central China (Fig.Figure 1). In this study, the physical options include the Lin microphysics scheme (Lin et al., 1983), RRTM longwave radiation scheme (Mlawer et al., 1997), Goddard shortwave scheme (Kim and Wang, 2011), MM5 M-O surface layer scheme (Chen and Dudhia, 2001), YSU boundary layer scheme (Hong et al., 2006), New Grell cumulus scheme, and Unified Noah land surface model (Chen and Dudhia, 2001). The chemical options include the RADM2 chemical mechanism, MADE/SORGAM aerosol scheme, Madronich F-TUV photolysis scheme, and Megan biogenic emission scheme (Guenther et al., 2006). The 1.0 ° × 1.0 ° NCEP Final Operational Global Analysis data (http://rda.ucar.edu/datasets/ds083.2/) have been processed to provide the meteorological initial conditions (IC) and boundary conditions (BC). We utilize the modified 2008 IGBP (International Geosphere Biosphere Programme) MODIS 20-category 30 s land-use data, which is available from the WRF website (http://www2.mmm.ucar.edu/wrf/users/), to represent current land cover conditions. Anthropogenic emission data are from Multi-resolution Emission Inventory for China (MEIC) developed by Tsinghua University for the year 2010, which consists of including the emission rates for each month from five sectors (agriculture, industry, power plants, residential and transportation). The MEIC is a unit/technology based, bottom-up emission model that covers

~700 anthropogenic emission source categories in China. It is an update of the emission inventory developed by the same group (Lei et al., 2011; Zhang et al., 2009). 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.

Predicted hourly ground level concentrations of CO, O_3, and particulate matter with aerodynamic diameter less than 2.5 microns ($PM_{2.5}$) are examined against observations made at five environmental monitoring sites, namely Nanjing Zhonghuamen Site (NJ_ZHM, 118.78 °E, 32.01 °N), Nanjing Xianlin Site (NJ_XL, 118.91 °E, 32.11 °N), Hangzhou Jiande Site (HZ_JD, 119.28 °E, 29.46 °N), Hangzhou Yuhang Site (HZ_YH, 119.99 °E, 30.26 °N) and Shanghai Pudong Site (SH_PD, 121.55 °E, 31.22 °N), as shown in ~~Fig.~~Figure 1. NJ_ZHM and NJ_XL are located in a mixed residential–educational area of Nanjing City, and the observation data (July 2012) are provided by Nanjing Municipal Environmental Monitoring Center. As NJ_ZHM and NJ_XL are located very close, and cannot be distinguished at the current model resolution, we average the observation data of these two sites and report it as NJ. Both HZ_JD and HZ_YH are located at high schools in Hangzhou City, and the relevant observation data (April 2008) are from Jiang et al. (2012). SH_PD is located in the urban center of Pudong District, Shanghai, and relevant observation data (September 2012) are from Tie et al. (2013). Extra simulations for April 2008 and September 2012 have been conducted for the purpose of model evaluation. The model performance is assessed by computing four conventional statistical metrics: the correlation coefficient (R), normalized mean bias (NMB), normalized mean error (NME), and index of agreement (I), which are defined as follows:

$$R = \frac{\sum_{i=1}^{N}(p_i - \bar{p})(o_i - \bar{o})}{\sqrt{\sum_{i=1}^{N}(p_i - \bar{p})^2 \cdot \sum_{i=1}^{N}(o_i - \bar{o})^2}} \quad (1)$$

$$NMB = \frac{\sum_{i=1}^{N}(p_i - o_i)}{\sum_{i=1}^{N} o_i} \times 100\% \quad (2)$$

$$NME = \frac{\sum_{i=1}^{N} | p_i - o_i |}{\sum_{i=1}^{N} o_i} \quad (3)$$

$$I = 1 - \frac{\sum_{i=1}^{N}(p_i - o_i)^2}{\sum_{i=1}^{N}(|p_i - \bar{o}| + |o_i - \bar{o}|)^2} \quad (4)$$

where p_i, o_i, and N represent model predicted data, observational data, and the number of data pairs, respectively. Eq. 4 indicates that I is a positive value no greater than 1, the larger the value of I, the better the model performs, and a value of 1 indicates a perfect match between the model and observations.

2.2 Scenarios of urban land expansion

This work investigates the sensitivity of climatic conditions and atmospheric chemical fields to changes in urban land cover on a regional scale. Four idealized urban land surface expansion scenarios are designed within the WRF/Chem modeling framework by modifying certain static geographical parameters. The land-use fraction by category ~~(denoted as LUF)~~ describes the percentage coverage of different land-use categories within a given grid cell, and the ~~dominant land-use category~~land-use index ~~(denoted as LUIND)~~ indicates the grid's dominant land-use category. The BASE run uses the prescribed 2008 IGBP MODIS 20-category land-use data. The GE0.2 run converts all cells with an urban land-use fraction~~LUF~~ of 0.2 or more to urban ~~dominant land-use category~~land-use index~~LUIND~~. The GE0.1 (urban land-use fraction~~LUF~~ $\geq$ 0.1) and GT0 (urban land-use fraction~~LUF~~ > 0) urban expansion scenarios are constructed similarly. Note that only the ~~dominant land-use category~~land-use index has been modified since the Noah land surface model considers only ~~dominant land-use category~~land-use index~~LUIND~~ rather than a mosaic of multiple land-use categories with various land-use fractions~~LUF~~. Figure 1 shows a schematic map of the four idealized urban land expansion scenarios (i.e., BASE, GE0.2, GE0.1, and GT0). The urban land surface expands extensively over the ~~YRD~~Yangtze River Delta, and most newly urbanized regions aggregate around the periphery of the original urban districts. Figure S1 (in the supplementary materials) shows the original dominant land-use categories of newly urbanized cells in the GT0 run, as well as a contour map of terrain height in the domain of interest. Most new urban areas emerge to replace cropland, whereas few appear in mountainous areas.

2.3 Integrated process rate analysis

In Eulerian grid air quality models such as WRF/Chem and CMAQ, the numerical technique of operator splitting is used to solve the governing equations for species' concentrations. Operator splitting involves separating the continuity equation for each species into several simpler partial differential equations or ordinary differential equations consisting of one or two individual processes (Gipson, 1999). The technique of integrated process rate (IPR) analysis has been developed to track the accumulated contributions of individual physical and chemical processes to model predictions during runtime. IPR has already been fully implemented in CMAQ, and recent studies have reported its use in investigating a high ozone episode in the ~~YRD~~Yangtze River Delta (Li et al., 2012) and the Pearl River Delta (Wang et al., 2010), China, as well as the fate of major airborne pollutants in the southeastern US (Yang and Shiang-Yuh, 2013).

However, IPR has not yet been officially adopted in the WRF/Chem modeling framework. Jiang et al. (2012) added a simple process analysis scheme to WRF/Chem to calculate the contribution of photochemical and physical processes to O_3 formation. In this paper, we extend this work by implementing an improved online IPR scheme in WRF/Chem to track contributions from 10 processes, namely horizontal and vertical advection (ADVH and ADVZ), emissions (EMISS), dry deposition (DRYDEP), turbulent diffusion (DIFF), convection (CONV), gas phase chemistry (CHEM), cloud chemistry (CLDCHEM), aerosol processes (AERCHEM), and wet scavenging (WETSCAV). The calculation of dry deposition is based on resistance models for gaseous species(Wesely, 1989) and particles (Ackermann et al., 1998). Note Zhang and He (2014) recently developed a new algorithm that linked dry deposition of particles with canopies via leaf area index (LAI), which is not included in the version of WRF/Chem used in this study. Cloud chemistry refers to aqueous-phase processes in different types of clouds, and aerosol processes refer to microphysical nucleation, condensation, and coagulation. Convective scavenging refers to in-cloud rainout within wet convective updrafts (subgrid processes), whereas wet scavenging refers to below-cloud washout during large-scale precipitation. In the modal aerosol scheme MADE/SORGAM (Ackermann et al., 1998; Schell et al., 2001), $PM_{2.5}$ comprises Aitken and accumulation mode particles of sulfate, nitrate, ammonium, organic carbon (including SOA) and black carbon. The process contribution is calculated by subtracting the species burden of each cell before and after each process is simulated. In WRF/Chem, dry deposition is

intermingled with vertical diffusion, so changes in the column burden during vertical mixing can be attributed to dry deposition. The IPR technique is verified by comparing the changes in species burden with the sum of contributions from the 10 processes mentioned above during each model output interval. As shown in Figure~~.~~ S2, the net contribution of these 10 processes broadly matches the species concentration change.

3 Model evaluation

Figure 2 compares simulated versus observed daily mean surface concentrations of O_3, CO, and $PM_{2.5}$ over five monitoring sites: NJ_ZHN (07/2012), NJ_XL (07/2012), SH_PD (09/2009), HZ_YH (04/2008), and HZ_JD (04/2008). The comparison indicates that WRF/Chem is capable of capturing the daily mean concentrations of surface O_3 (R = 0.66; NME = 27%), CO (R = 0.74; NME = 41%), and $PM_{2.5}$ (R = 0.63; NME = 29%). 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 ± 29.0 μg/m^3, while in SH_PD site ~~is~~are 41.8 ± 12.3 and 44.1 ± 21.0 μg/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. ~~Table 1 lists the statistical results for T~~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, ~~with a correlation coefficient~~(i.e., R:~~of~~ 0.74, NMB: ~~of~~ 6.7%, NME: ~~of~~ 34.1 %, and I: ~~value of~~ 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. In addition, WRF/Chem captures the monthly mean surface concentrations of O_3, CO, and $PM_{2.5}$ fairly well (although O_3 in SH is over-predicted by 17% and CO in NJ is under-predicted by 30%). A number of previous air quality studies have evaluated the performance of WRF/Chem in simulating a range of chemical species (e.g., $PM_{2.5}$, CO, and O_3) over China (Li et al., 2012; Tie et al., 2013; Tie et al., 2007), and these have reported similar results.

4 Impacts of urban land expansion on regional atmospheric environment

4.1 Urbanization-induced concentration changes

Urban land expansion significantly alters the local synoptic conditions (see ~~Fig.~~Figure S3 in the supplementary materials), e.g., an increase in 2-meter temperature ~~(T2)~~ and boundary layer height ~~(PBLH)~~, and a decrease in 2-meter relative humidity ~~(RH2)~~ and 10-meter wind speed (~~W10~~, could be different for regions where urban land cover replaces forests). Changes in meteorology impact ambient air quality, even when anthropogenic emissions remain constant. We focus on the response of two gaseous species (i.e., CO and O_3) and two aerosol species (i.e., EC and $PM_{2.5}$). EC and CO are used to study how urban land expansion would impact the dispersion and dilution of primary pollutants. EC includes Aitken-mode EC (ECI) and accumulation-mode EC (ECJ), and the aerosol scheme simulates the aging process by converting ECI to ECJ. O_3 and $PM_{2.5}$ are used to further investigate the effects of urban land expansion on ~~chemical reactions~~secondary pollutants.

The model surface layer (~~Fig.~~Figure 3) and 800 hPa layer (~~Fig.~~Figure 4) are selected to study the 5-year mean concentrations in July of CO, EC, O_3, and $PM_{2.5}$ under the four urbanization scenarios. In the BASE run, high levels of surface CO (~850–1250 ppb), EC (~9–13 μg m^{-3}), and $PM_{2.5}$ (~90–130 μg m^{-3}) are found only in urban areas where anthropogenic emissions are high. In contrast, terrestrial O_3 (~24–32 ppb) is more evenly distributed on the regional scale. At 800 hPa, concentrations of CO (~40–70 ppb), EC (~0.2–0.4 μg m^{-3}), O_3 (~24–30 ppb), and $PM_{2.5}$ (~10–20 μg m^{-3}) over the North China Plain appear much higher ~~over the North China Plain (NCP)~~ than those in the southern domain, consistent with the satellite-observed pollution distribution features over this domain (e.g. Liu et al., 2013). A one-tailed student *t*-test (based on the standard error computed from hourly variability) is used to determine whether the assumed expansion of urban land causes changes in local monthly mean concentrations that are significant at the 95% confidence level. In the surface layer, the change in dominant land-use type to urban generally induces a significant decrease in surface concentrations of CO (up to -44%; domain-wide average of —-11%, or up to 40 ppb decrease in the GT0 run), EC (up to -80%; domain-wide average of —-21%, or -0.3 μg m^{-3} in the GT0 run), and $PM_{2.5}$ (up to -74%; domain-wide average of -21%, or -5.4 μg m^{-3} in the GT0 run) in all urbanization scenarios. However, the changes in surface O_3 are generally insignificant in GE0.2. Urban land expansion leads to a moderate increase in surface O_3 (maximum of 22%; domain-wide average of 0.3%, equivalent to 0.1 ppb in the GT0 run) over the northern

terrestrial domain in the GE0.1 and GT0 runs. At 800 hPa, the expansion of urban land significantly increases the local concentrations of CO (with a domain-wide average of 16%, and up to 5.6 ppb in the GT0 run), EC (domain-wide average of 50%, or ~0.05 μg m^{-3} in the GT0 run), O_3 (domain-wide average of 16%, or ~2.8 ppb in the GT0 run), and $PM_{2.5}$ (domain-wide average of 65%, or ~4.3 μg m^{-3} in the GT0 run) in the different urbanization scenarios. The effect of urban land expansion on CO, EC, O_3 and $PM_{2.5}$ concentrations is consistent in each year, albeit with slight differences in magnitude (not shown).

4.2 Linearity of urbanization-response relationship over East China

The urbanization-response relationship is a complex function of local synoptic conditions, large-scale circulation, land surface type, the physical and chemical properties of an airborne contaminant and its emissions (e.g., release height, frequency, and amount), as well as the time scale being considered. The strength of urban land forcing (e.g., the sensitivity of an atmospheric variable to urban land expansion) can be quantitatively evaluated as the perturbation of this variable from its base condition. Figure 5 shows the response of the 5-year mean concentrations in July of CO, EC, O_3, and $PM_{2.5}$ to changes in domain-wide (i.e., East China) urban land coverage. At both the surface and 800 hPa, the response curves are nonlinear and the rate of domain-wide concentration changes decreases as more urban land emerges. However, as shown in Figures. 3 and 4, the concentration response is nonuniformly distributed, and becomes stronger when large tracts of new urban land appear. This indicates that the aggregation state (further discussed in the next paragraph) of newly urbanized areas would alter the strength of urban land forcing.

The forcing effect of urban land expansion on the spatial distribution of air pollutants is not usually limited to newly urbanized areas, but has a distance of extended influence. To differentiate the shape of urbanization-response curves at different locations, we use LOCAL to denote these newly urbanized cells, and ADJACENT to represent the non-urbanized cells neighboring LOCAL. We further define an aggregation parameter (Agg) for a given LOCAL cell as the number of the surrounding cells that are also LOCAL; we limit the number of surrounding grid cells for this analysis to $5 \times 5 - 1$ or 24 cells (2400 km^2). The "local" and or "adjacent" forcing is defined as:

$$f_j = \frac{\sum_{i=1}^{N_j} \frac{VC_i}{VB_i}}{N_j} \quad (5)$$

where f_1 (i.e., j = 1) denotes local forcing (LF), N_1 is the number of domain-wide LOCAL cells, VC_i and VB_i denote the values of a certain atmospheric variable in cell i of the perturbation run and the BASE run, respectively. f_2 denotes adjacent forcing (AF) over the ADJACENT cells, and N_2 is the number of domain-wide ADJACENT cells.

Figure 6 illustrates the LF and AF induced by urban land expansion (i.e., the perturbation of 5-year mean in July concentrations of CO, EC, O_3, and $PM_{2.5}$ from the BASE situation) in all three idealized urbanization scenarios. At the surface, the relationship between LF and Agg is nearly linear. Each 10% increase in Agg is associated with 1.6%, 1.8%, and 2.2% decreases in LOCAL CO, EC, and $PM_{2.5}$ concentrations, respectively, and a 1% increase in O_3 concentrations. The AF-Agg curves are similar to those for LF-Agg, but have weaker responses. The linearity can be explained as follows: with increasing Agg, there are more LOCAL cells near a given cell, so the same number of adjacent effects are added to the given cell. At 800 hPa, changes in the contaminant burden seem to be more sensitive to the aggregation level of newly urbanized cells than at the surface, and particles seem to be more susceptible than gases. The associated urbanization-response curves become nonlinear. It can be observed that both LF and AF are linearly associated with the square of Agg, which means each 10% increase in the square of Agg may enhance air pollution concentrations by about 5–10% at 800 hPa, with the maximum sensitivity for $PM_{2.5}$ (12%). This indicates that dense urbanization over East China may have a moderate dilution effect on surface air pollution, but could intensify pollutants aloft and therefore severe haze (i.e. visibility degradation) and ozone pollution if local emissions are not reduced in the future.

Besides the response of air pollutants, we found that the perturbations in July-mean ~~PBLH~~boundary layer height, ~~T2~~2-meter temperature, and ~~RH2~~2-meter relative humidity also increase linearly with Agg ($R^2 > 0.96$, shown in ~~Fig.~~Figure S4 in the supplementary materials). These results indicate that when large tracts of new urban land emerge, impacts of land cover change on meteorology and air pollutant concentrations can be magnified.

5 Mechanism governing the urbanization-response relationship

5.1 Process contribution to surface air quality changes

Figure 7 shows the 5-year mean in July diurnal cycles of CO, EC, O_3, and $PM_{2.5}$ surface concentrations averaged over the domain-wide LOCAL cells and the ADJACENT cells. At the surface, CO, EC, and $PM_{2.5}$ share a diurnal variation pattern in which concentrations peak at dawn (~05:00 LST) and reach a trough in the late afternoon (~16:00 LST). Concentrations over LOCAL cells are usually higher than those over the ADJACENT regions, particularly during the night. However, for O_3, the opposite diurnal variation pattern can be observed, and the difference between LOCAL and ADJACENT cells is small. Urban land expansion leads to substantial changes in species concentrations, but has little effect on the shape of the diurnal cycle. As urban land expands, CO, EC, and $PM_{2.5}$ tend to evolve toward lower burden levels. In contrast, when averaged over the domain, the increment in surface O_3 concentrations during most of the day is insignificant. Though the resulting concentration changes in LOCAL and ADJACENT are quite similar for these four species, the IPR analysis suggests that the underlying mechanisms that drive the forcing–response relationship are different.

Figure 8 illustrates the 5-year mean in July diurnal cycles of IPR contributions in the BASE run and their deviations in the GT0 run over the domain-wide surface LOCAL cells and ADJACENT cells. The daytime period is chosen as 07:00–18:00 LST, with the rest of the day considered to be nighttime. In the BASE run, emissions are the dominant source of CO over the LOCAL cells, and the dominant sink is turbulent transport (daytime advection is also a contributing sink). EC follows a similar IPR pattern to CO, except that dry deposition is also a major sink, accounting for about 40% of the total EC removal. Since this study considers constant CO and EC emissions, diurnal variability in concentrations is dominated by variations in the strength of vertical transport. During the daytime, vertical transport is strong, and CO and EC are depleted at the surface. However, during the nighttime, vertical transport becomes weaker, which allows CO and EC to accumulate. The diurnal cycle of IPR for $PM_{2.5}$ is very similar to that of EC, but aerosol processes play an important role. During the daytime, the source of $PM_{2.5}$ is dominated by surface emissions, but the sinks are quite complicated, including turbulent diffusion (~41%), dry deposition (~36%), and aerosol processes (~23%). Note that by conducting IPR analyses within CMAQ, Yang and Shiang-Yuh (2013) found that aerosol processes depletion can act as a sink for $PM_{2.5}$. At night, the production of $PM_{2.5}$

through aerosol processes forms 22% of the source. The IPR diurnal cycle for O_3 is quite different. During the daytime, the major sources for surface O_3 are photochemical production (~37%) and turbulent transport (~63%), and the sink is overwhelmingly dry deposition. At night, removal of O_3 through gas-phase reactions and dry deposition accounts for 40% and 60% of the sink, respectively. The diurnal cycles of IPR contribution over the ADJACENT cells are similar to those of LOCAL cells, except that vertical diffusion becomes a source for $PM_{2.5}$ during the daytime, compensating for the strong loss by aerosol processes and dry deposition.

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 ~~Fig.~~Figure 9. The urban land expansion could strengthen the southeasterly sea breeze over marine and near the east seaboard of China, 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. ~~At the surface, urban land expansion reduces the local pressure (up to 530 Pa) and forms a cyclonic anomaly.~~ The divergence of the perturbed wind field can be calculated by a centered finite difference scheme with a one-sided difference boundary. Values of convergence up to -6×10^{-5} s^{-1} can be observed in most of the newly urbanized areas, similar to the results of Bornstein and Lin (2000), who concluded that UHI would induce a convergence (~-10^{-5} s^{-1}) zone over Atlanta. Figure 10 shows the perturbed wind field in the three vertical–longitudinal cross-sections of CS1, CS2, and CS3 mentioned in ~~Fig.~~Figure 1. The emergence of urban land induces local updrafts of ~1 cm s^{-1}, which enhances the ventilation of primary pollutants to the free troposphere; adjacent downdrafts are also observed. The perturbed wind induced by urban land expansion generally forms convergence zones above the LOCAL cells and divergence zones over the ADJACENT cells (~~Fig.~~Figure 9). UHIC is enhanced, which explains the changes in the contribution of advection.

The effects of urban land expansion are not limited to the UHIC-induced advection changes. In the surface layer, the vertical diffusion of CO is intensified over LOCAL cells (especially

during the night). EC differs from CO in that the loss due to dry deposition markedly increases over the entire day, while the sink due to diffusion is only reduced during the daytime but is increased at night. The sum of dry deposition and turbulent diffusion reflects the role of vertical mixing in relocating the airborne pollutants vertically. The sink due to the vertical mixing of EC is intensified. It can be concluded that the enhanced advection and turbulent mixing in the vertical direction are the key factors in reducing the surface concentrations of CO and EC. For O_3, dry deposition, vertical diffusion, and daytime photochemical production and nighttime chemical depletion are all reduced at the surface, resulting in a net weak enhancement of surface O_3 averaged over domain-wide LOCAL cells. The dry deposition of O_3 is reduced because the canopy resistance increases as vegetation is replaced by urban land; however, the dry deposition of particles is not impacted by canopy resistance, but is rather fostered by the intensified surface turbulence. Vertical diffusion is possibly hindered by convergent updrafts caused by UHIC, and the daytime photochemical production and nighttime chemical depletion are reduced because of the decreased abundance of precursors. For $PM_{2.5}$, daytime loss and nighttime production via aerosol processes are enhanced and hindered, respectively, this may be because the urbanization-induced decrease in precursor concentrations restrains gas-to-particle mass transfer. At the same time, dry deposition and vertical diffusion are also enhanced during the daytime. Therefore, the increased sink resulting from aerosol processes and dry deposition is the key factor reducing $PM_{2.5}$ concentrations at the surface. Over the ADJACENT cells, urbanization-induced outward horizontal advection contributes to the lower burden of CO, EC, and $PM_{2.5}$, and the sink term due to vertical mixing decreases.

5.2 Urbanization-induced process-level vertical profile changes

Figure 11 shows the 5-year mean in July vertical profiles of the four species averaged in the domain-wide LOCAL and ADJACENT cells of the BASE and GT0 runs. In both types of cells, CO, EC, and $PM_{2.5}$ exhibit similar patterns; as urban land expands, the atmospheric burden decreases near the surface (below 1 km), but increases at higher altitudes (1–4 km). On the other hand, the concentration of O_3 increases at most heights from the surface to a height of about 4 km. Unlike near the surface, the magnitude of the concentration perturbations aloft (1–4 km) in both grids is commensurate with the atmospheric burden.

Figure 12 illustrates the vertical profile of 5-year mean in July daytime and nighttime IPR contributions for CO, EC, O_3, and $PM_{2.5}$ in the BASE and GT0 runs, averaged over all

LOCAL cells. Convective scavenging generally plays a minor role in removing these four species. For CO and EC, vertical mixing and advection play key roles in constraining the vertical profiles, and the net diffusion is unidirectional from the ground to higher altitudes. The extent of vertical transport during the daytime is higher than that during nighttime. The forcing of urban land expansion on the transport of primary pollutants is characterized by UHIC-induced advection changes and enhanced vertical mixing, leading to the decreased vertical concentration gradient (as shown in ~~Fig.~~Figure 11). A positive perturbation in the horizontal advection contribution and a negative perturbation in the vertical advection contribution are found in the lower atmosphere (below 500 m), whereas the opposite is true in the upper atmosphere (~0.5–3 km), similar to the Ekman pump. For CO and EC (only during nighttime), the enhanced sink and source due to vertical mixing in lower and upper atmosphere, respectively, could be the key reasons for changes of vertical profile, and advection appears to compensate and balance this effect. However, for EC above 1 km, as surface dry deposition is strengthened markedly in the daytime, whereas diffusion from the lower atmosphere is dampened; this is compensated by the enhanced upward advection.

For O_3, advection, vertical mixing, and gas-phase reactions all play important roles in constraining its vertical profile. Though UHIC-induced horizontal and vertical advection changes cause the IPR to shift significantly across all layers, net advection is not the key process driving the changes in the vertical O_3 profile. Near the ground level, the expansion of urban land fosters upward diffusion and hinders downward diffusion to the surface layer. O_3 production is determined by the availability of precursors, which is increased in the 1–3 km zone by the enhanced uplifting of primary pollutants. The dampened dry deposition and enhanced daytime photochemical production (at around 0.5–3 km) are responsible for a higher O_3 profile.

Besides the transport of precursors, other meteorological factors may also influence O_3 production. The bottom three plots in ~~Fig.~~Figure 10 show the distribution of chemical production (ppb/h), cloud water, and air temperature differences (GT0 minus BASE during the period 12:00–17:00 LST) in the cross-sections of CS1, CS2, and CS3 (defined in ~~Fig.~~Figure 1). As urban land expands, air temperatures increase (up to 1.2 ℃) in the lower layers (below 0.5 km), but decrease slightly above 2 km. However, photochemical production of O_3 is intensified only at altitudes higher than 2 km. This indicates that changes in air temperature may not be the principal factor determining O_3 production above the PBL. As

shown in ~~Fig.~~Figure 10, the locations of newly urbanized cells exactly match the zones where photochemical production of O_3 is reduced in lower layers but cloud water content is significantly increased above 1 km. Low altitude clouds efficiently scatter shortwave radiation, thus hindering photochemical reactions below clouds; thus, urban land forcing indirectly effects the spatial distribution of O_3.

As shown in ~~Fig.~~Figure 12, aerosol chemistry, vertical mixing, and advection all contribute strongly to constraining the $PM_{2.5}$ vertical profile, with wet scavenging and cloud chemistry playing relatively minor roles. During the daytime, the contribution of aerosol chemistry is negative near the surface, but turns positive at higher altitudes. At night, the contribution of aerosol chemistry remains positive in all vertical layers. The net vertical turbulent transport is upward in the surface layer, but reverses to downward above 0.5 km. Similar to O_3, as urban land expands, changes in the vertical profiles of precursors result in enhanced aerosol production at 0.5–3 km, and enhanced loss below 0.5 km (where the downward diffusion is intensified). The perturbation of dry deposition and aerosol processes largely explains the $PM_{2.5}$ vertical profile changes. Alike the case for the photochemical formation of O_3, 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 S5~~11~~ in the supplementary materials, the production of PM2.5 through cloud chemistry increase (decreases) exactly where the humidity increases (decreases).

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. 2) It is important to note that emissions are assumed to remain constant during our study time period. We chose constant emissions to ensure we could tease out the effects of land-cover induced changes on air quality without confounding changes in emissions. Additional s~~S~~ensitivity experiments show that, at the surface, the diluting effects of urban land expansion could be offset~~ed~~ only if the emission ~~augment~~increase is high enough for CO (~40%) and EC (~100%) in LOCAL cells (~~refer to~~see Figures S6-S8 in the supplemental materials for details). 3) The BULK urban canopy scheme used in this work does not resolve the urban morphology, and therefore cannot further investigate how urban canopy parameters, such as the building height and anthropogenic heat,

would impact the climatic conditions and air quality. The urbanization-response relationship unveiled in this work could be urban scheme dependent. In future studies, we will focus on addressing these effects to better quantify the urbanization–response relationship, which could provide support to urban planning.

6 Conclusions

We have used an online coupled mesoscale meteorology-chemistry model (WRF/Chem) with BULK urban scheme embodied in Noah land surface model and an improved integrated process rate (IPR) analysis scheme to study the effects of urban land expansion in eastern China on climate and air quality during the month of July. Urban land expansion could significantly alter local synoptic conditions (e.g., increases in 2-meter air temperature ~~(T2)~~ and boundary layer height ~~(PBLH)~~ and decreases in 2-meter relative humidity ~~(RH2) and 10-meter wind speed (W10)~~). Above the newly urbanized grid cells (referred to as LOCAL cells), horizontal perturbations in wind form cyclonic convergence (~10^{-5} s^{-1}) zones, and vertical perturbations in wind lead to updraft flows (~1 cm/s). This urbanization-induced circulation consequently impacts ambient air quality, even when surface emissions remain constant. For primary pollutants with strong surface emissions (e.g., CO and EC), urban land expansion causes concentrations to decrease below 500 m but increase significantly between 1 and 3 km. On the other hand, the O_3 burden averaged over LOCAL cells consistently increases from the surface to about 4 km. For $PM_{2.5}$, though its source includes both primary emissions and secondary formation, the changes in vertical profile caused by urban land expansion are consistent with those of CO and EC.

The effects of urban land expansion are not localized, but rather its influence extends to neighboring areas. In this study, the local forcing (LF) was found to be significantly larger than adjacent forcing (AF) at the surface, especially for particulate matter. The aggregation state of newly urbanized areas plays an important role in determining the strength of LF and AF. We found that perturbations of CO, EC, O_3, and $PM_{2.5}$ change linearly with aggregation parameter (Agg) at the surface, and with the square of Agg at 800 hPa ($R^2 > 0.94$). In addition, the perturbations of mean July levels of boundary layer height~~PBLH~~, 2-meter temperature~~T2~~, and 2-meter relative humidity~~RH2~~ increase linearly with Agg ($R^2 > 0.96$). This result indicates that when large tracts of new urban land emerge, the effects of urban expansion on atmospheric physics and chemistry are magnified.

IPR was utilized to investigate the forcing mechanisms exerted by urban land expansion on the spatial distribution of CO, EC, O_3, and $PM_{2.5}$ over the LOCAL cells. At the surface, a common feature of all four species governed by the UHIC effect whereby horizontal advection causes increases in concentrations and vertical advection causes decreases in concentrations. Additionally, when natural vegetation was replaced by urban land, the sink term from dry deposition increased for particles but decreased for gaseous species. For primary pollutants CO and EC, enhanced advection and turbulent mixing in the vertical direction are the key factors in reducing the surface concentrations. On the other hand, for $PM_{2.5}$, increased sinks due to aerosol processes and dry deposition are the key factors in reducing surface concentrations. For O_3, the reduced dry deposition and vertical diffusion, as well as the relocation of precursors, played an important role in constraining the surface concentration, resulting in a net enhancement of the surface O_3 averaged over all LOCAL cells.

In contrast to the surface conditions, urban land expansion may induce substantial increases in air pollution at higher altitudes. The positive contribution of vertical advection and the negative contribution of horizontal advection were found to be important in the build-up of air pollution in the upper atmosphere (0.5–3 km). For primary pollutants CO and EC (only during nighttime), the enhanced uplifting caused by strengthened turbulent diffusion (induced by urban land expansion) is the key factor leading to higher burdens in the upper atmosphere. However, in daytime, diffusion of EC from the lower atmosphere is dampened due to intensified dry deposition, which partially counters the concentration increases from enhanced upward advection. However, for secondary species, O_3 and $PM_{2.5}$, the relocation of precursors accelerates daytime chemical production in the upper atmosphere, which is the key factor in the higher burden of secondary pollutants at a height of about 1–4 km.

Above analysis ~~This study mainly has~~ revealed the nonnegligible and unique role of urban land forcing in impacting 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 emission simultaneously to better evaluate the potential environmental influence of any urbanization campaign. ~~The urbanization-response relationship unveiled in this work could be urban scheme dependent,~~

~~and more complicated urban schemes (but with higher levels of uncertainty) should be applied in future studies.~~

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~~Table 1. Statistical results for the modeled and observed hourly concentrations over sites NJ (07/2012), SH_PD (09/2009), HZ_YH (04/2008), and HZ_JD (04/2008) for CO, O_3, and $PM_{2.5}$.~~

~~Species~~	~~Correlation coefficient~~	~~NMB (%)~~	~~NME (%)~~	~~I~~
~~CO~~	~~0.22~~	~~-22.4~~	~~52.6~~	~~0.49~~
~~O_3~~	~~0.74~~	~~6.7~~	~~34.1~~	~~0.86~~
~~$PM_{2.5}$~~	~~0.38~~	~~11.2~~	~~63.4~~	~~0.60~~

Table 1. ~~The explanation of~~List of acronyms used in this work

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

Figures

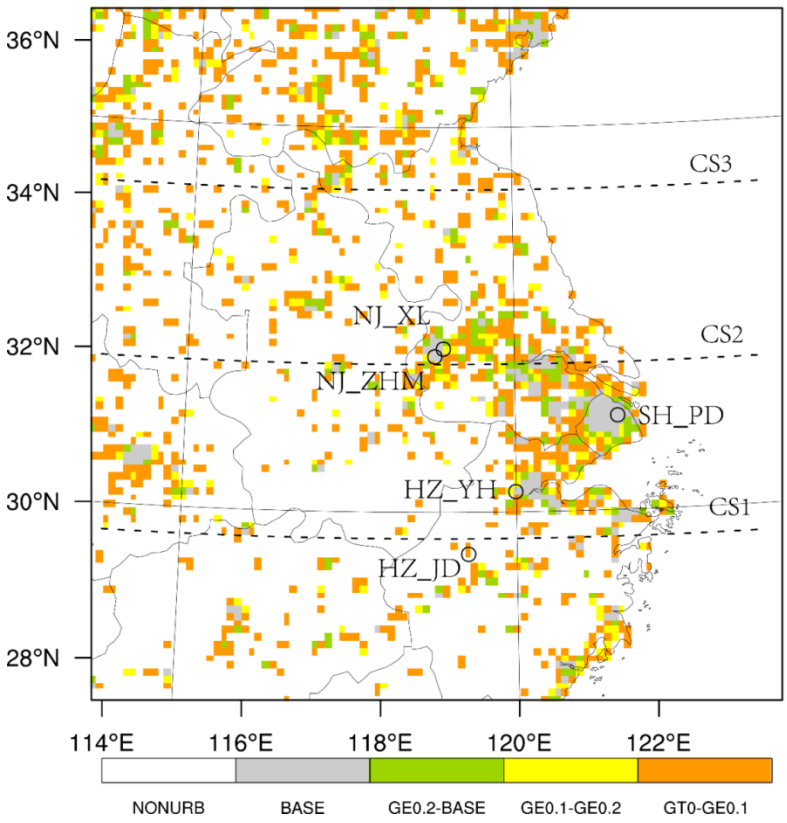


Figure 1. Schematic map of four idealized urban land expansion scenarios (i.e., BASE, GE0.2, GE0.1, and GT0). White denotes non-urban cells, and grey denotes urban cells in the BASE run. Other colors represent additional newly urbanized cells in GE0.2 (green), GE0.1 (yellow), and GT0 (orange) compared to previous urban land expansion scenarios. For example, urban cells in GE0.1 are grey, green, and yellow. Black open circles denote the five air quality monitoring sites. Black dashed lines running west-east demarcate the three vertical-zonal cross-sections CS1, CS2, and CS3.

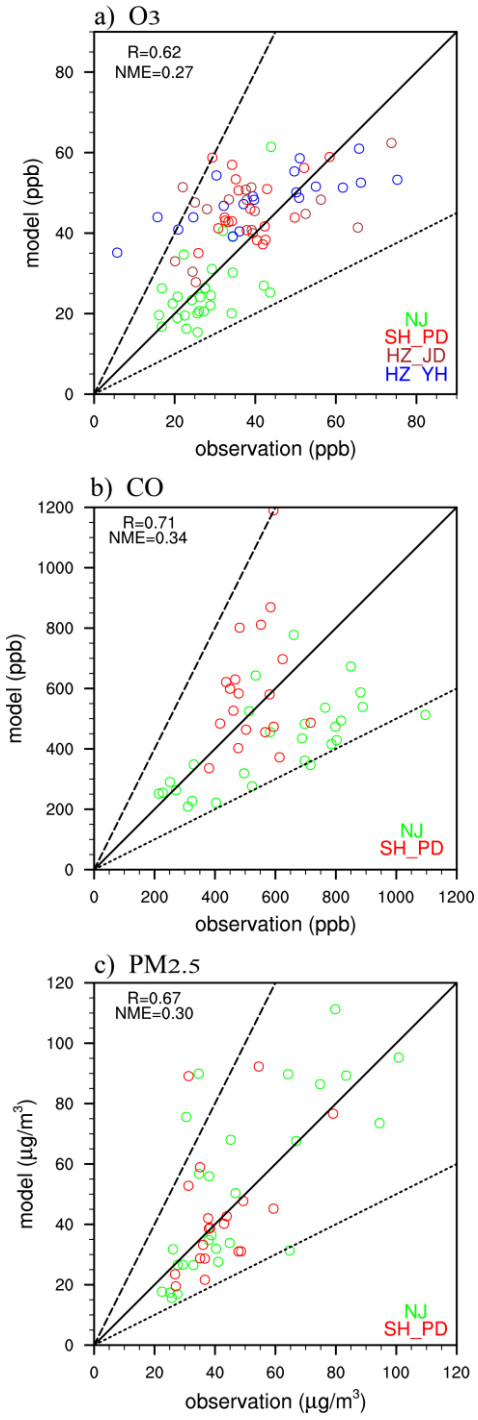


Figure 2. Modeled versus observed daily mean surface concentrations of (a) O_3, (b) CO, and (c) $PM_{2.5}$ at NJ (07/2012), SH_PD (09/2009), HZ_YH (04/2008) and HZ_JD (04/2008). Solid line indicates the 1:1 line; dashed lines indicate the 1:2 and 2:1 lines.

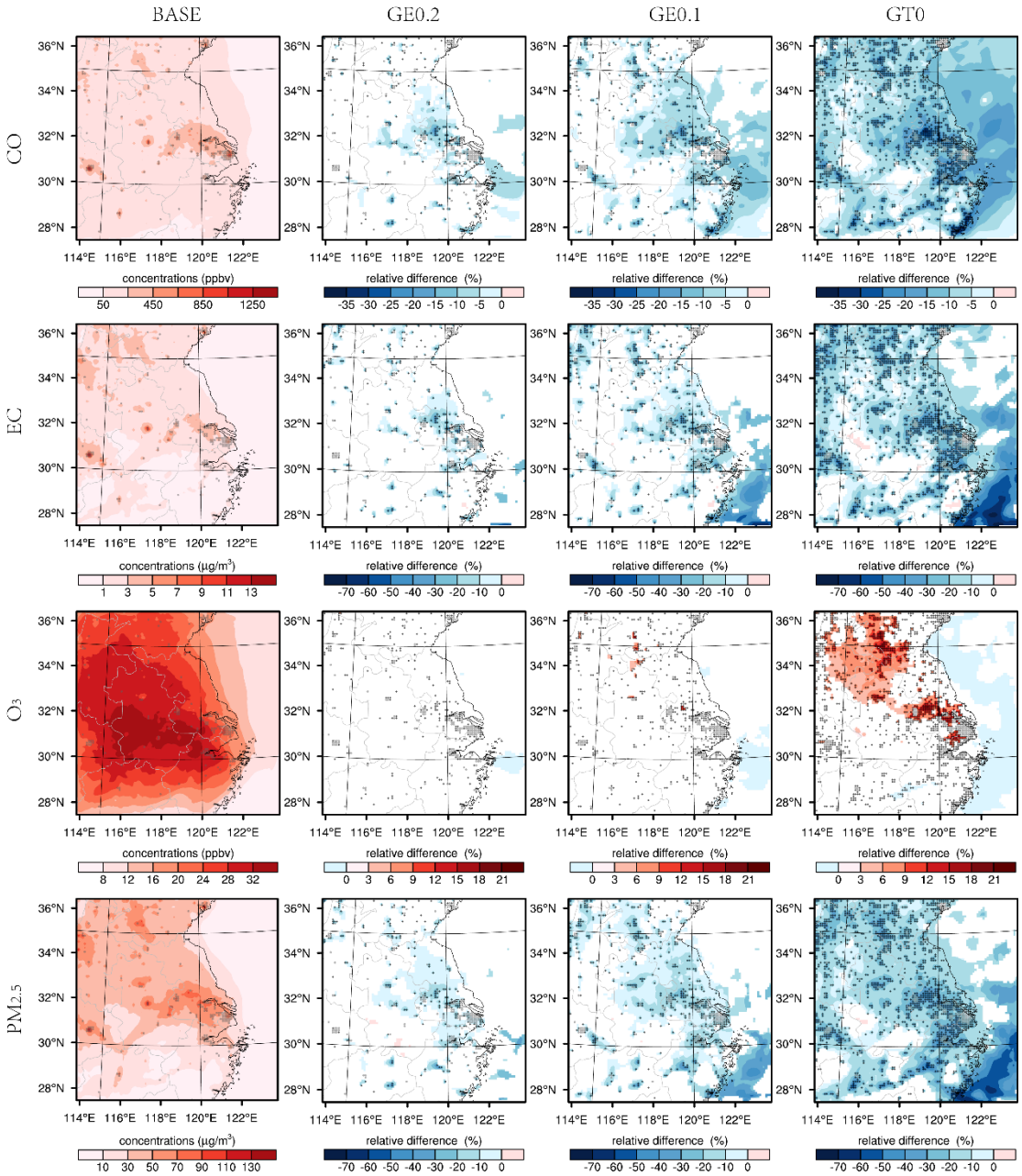


Figure 3. Five-year mean surface concentrations in July of CO, EC, O_3, and $PM_{2.5}$ in the BASE run (left), and the relative difference (only cells exceeding the 95% significance level are shown) of each urban land expansion scenario relative to BASE (right three columns). Grey circles indicate urban areas in the BASE run; black crosses indicate newly urbanized cells in GE0.2, GE0.1, and GT0.

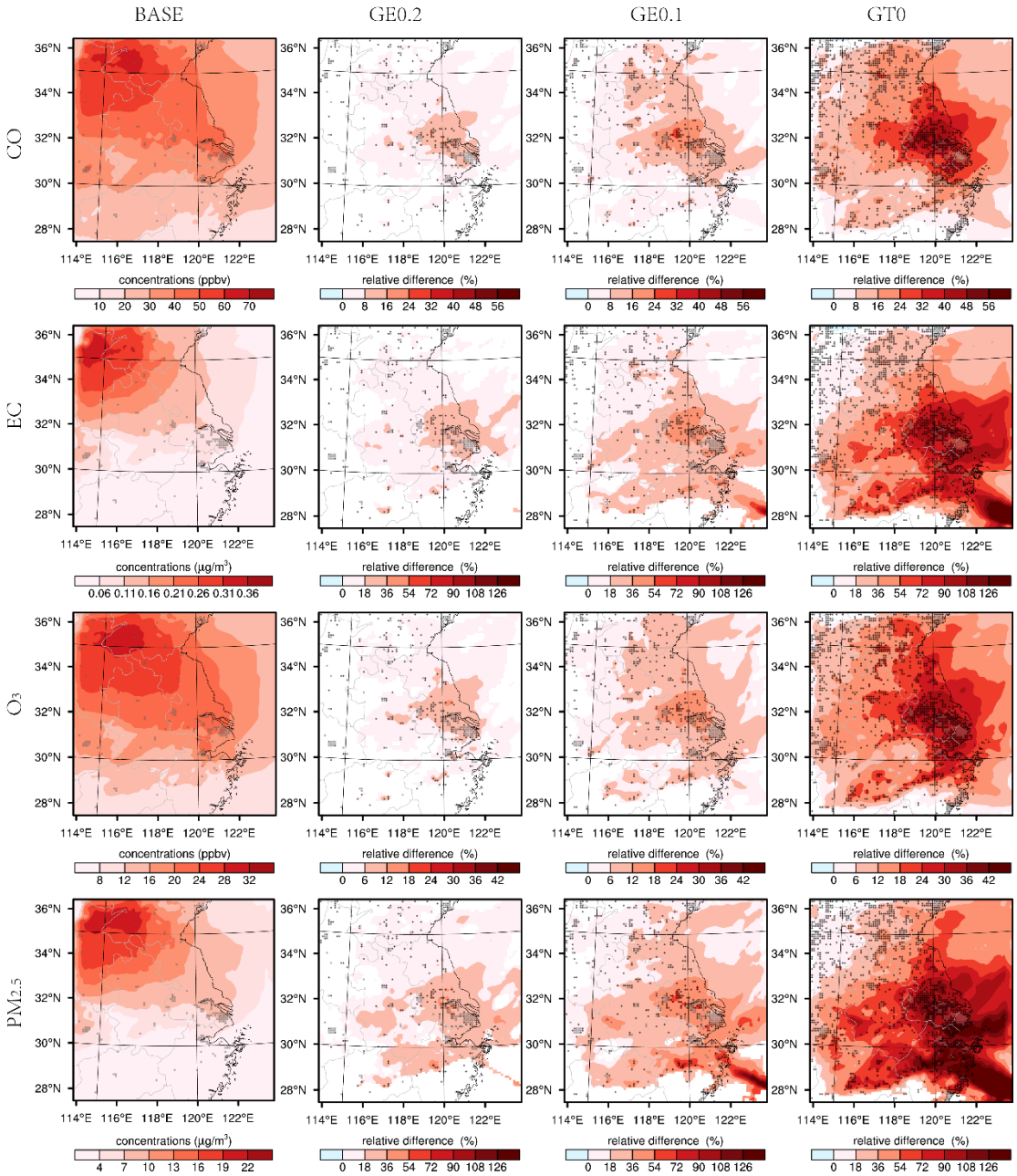


Figure 4. Same as ~~Fig.~~Figure 3, but at 800 hPa.

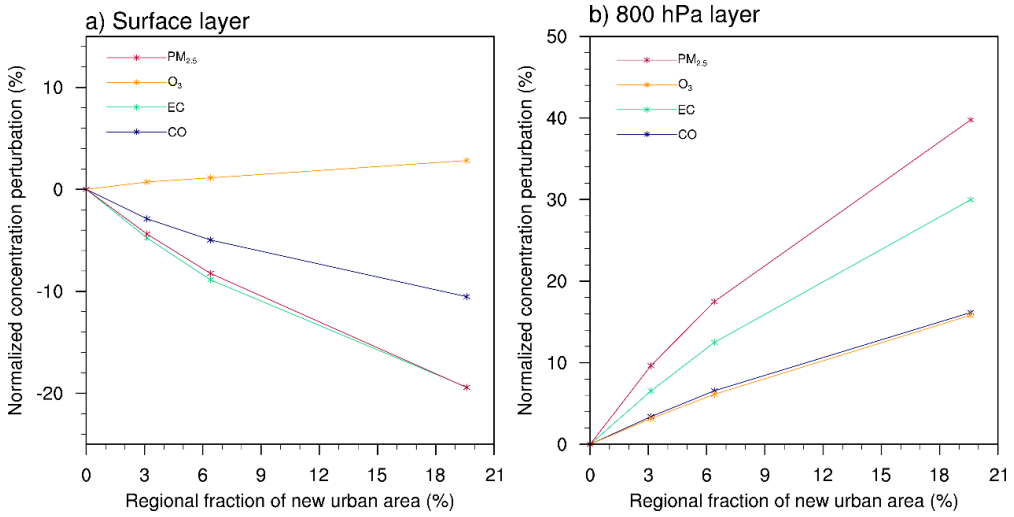


Figure 5. Normalized perturbations (relative to the BASE simulation) for three urbanization scenarios of the 5-year mean concentrations in July of CO, EC, O_3, and $PM_{2.5}$ at the (a) surface and (b) 800 hPa. Values are averaged over land for the entire domain. Corresponding domain-wide land fraction of new urban areas are 3%, 6%, and 20%, respectively, relative to BASE.

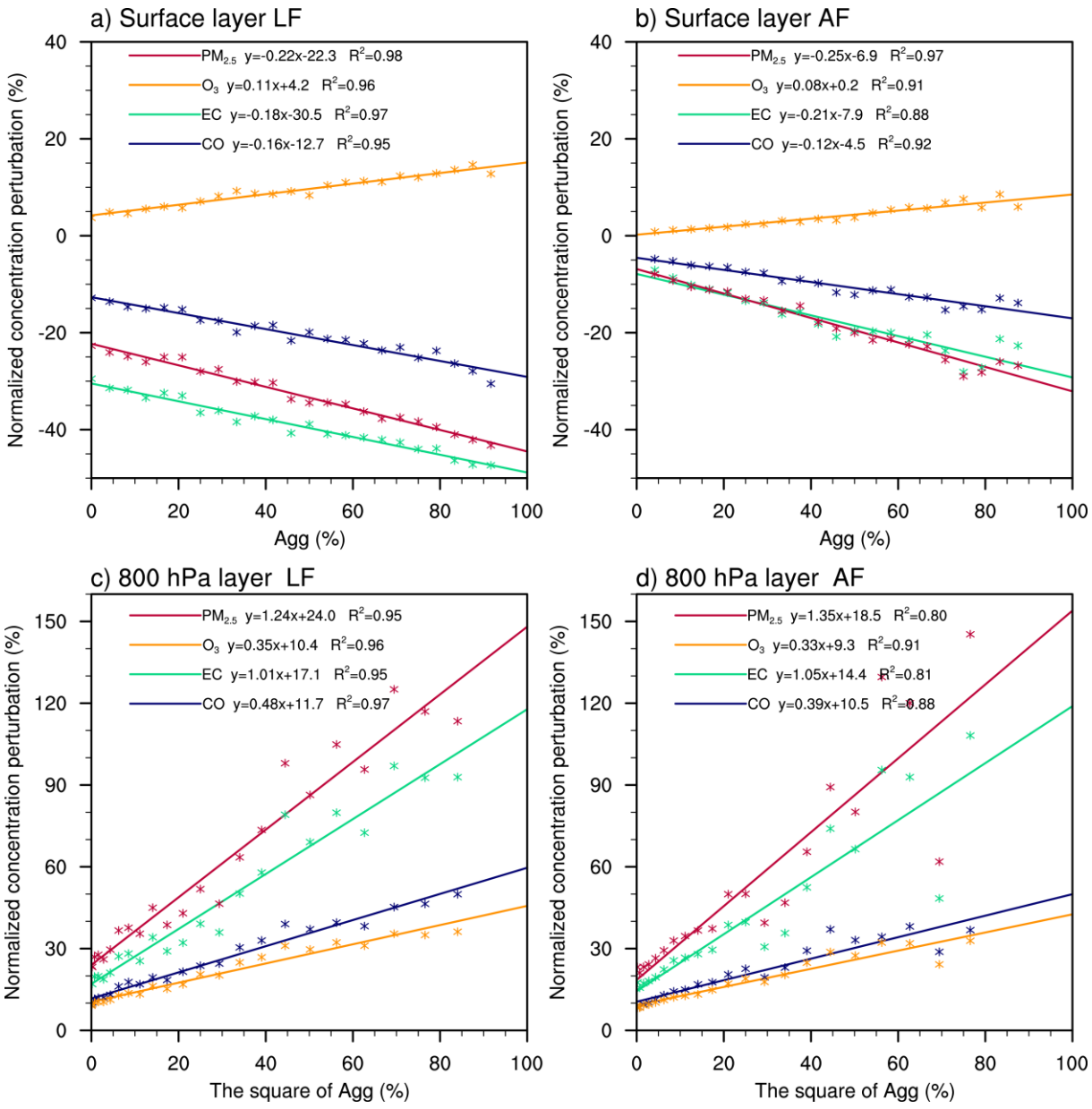


Figure 6. Relationship between normalized perturbations of 5-year mean July concentrations of CO, EC, O_3, and $PM_{2.5}$ and Agg at the surface (top), and the square of Agg at 800 hPa (bottom) for the LOCAL forcing (LF, left) and ADJACENT forcing (AF, right). Agg is defined as the occupation rate of the newly urbanized cells to the surrounding 24 ($5 \times 5 - 1$) cells. Linear regression results are also shown.

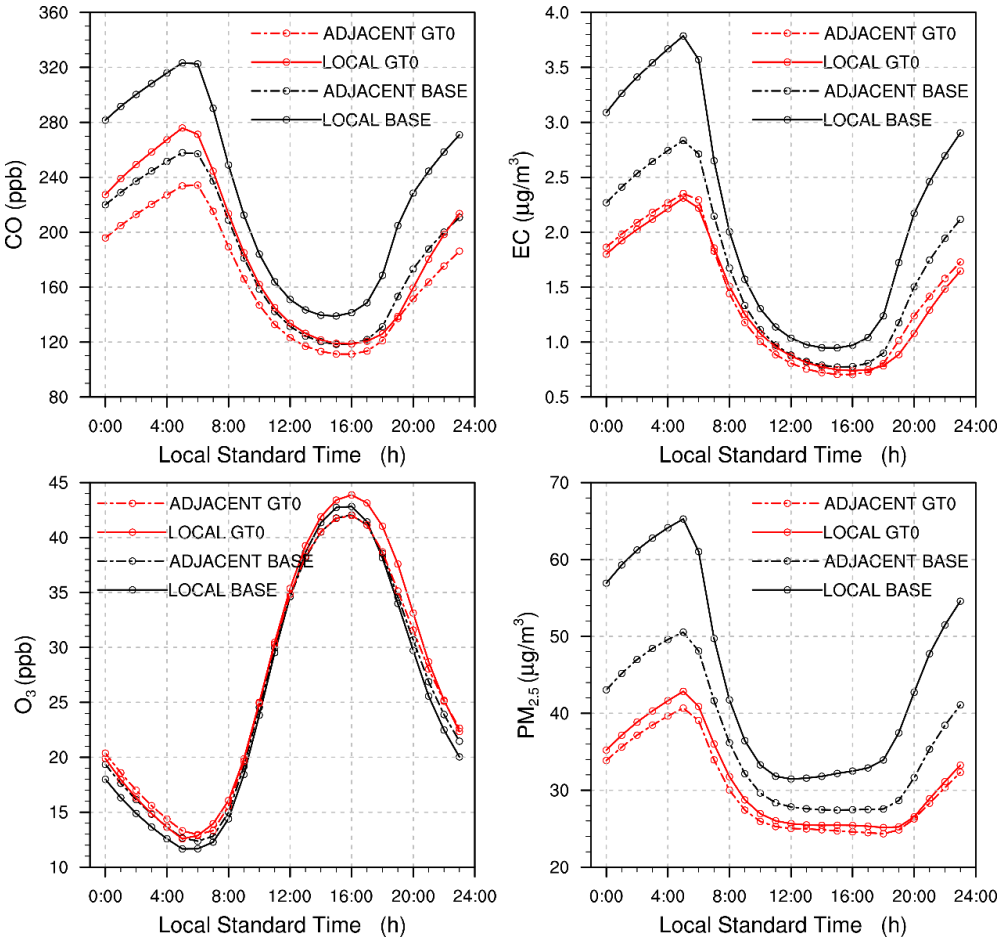


Figure 7. Simulated 5-year mean July diurnal cycle of CO, EC, O_3, and $PM_{2.5}$ at the surface, averaged over domain-wide LOCAL cells (solid lines) and ADJACENT cells (dashed lines).

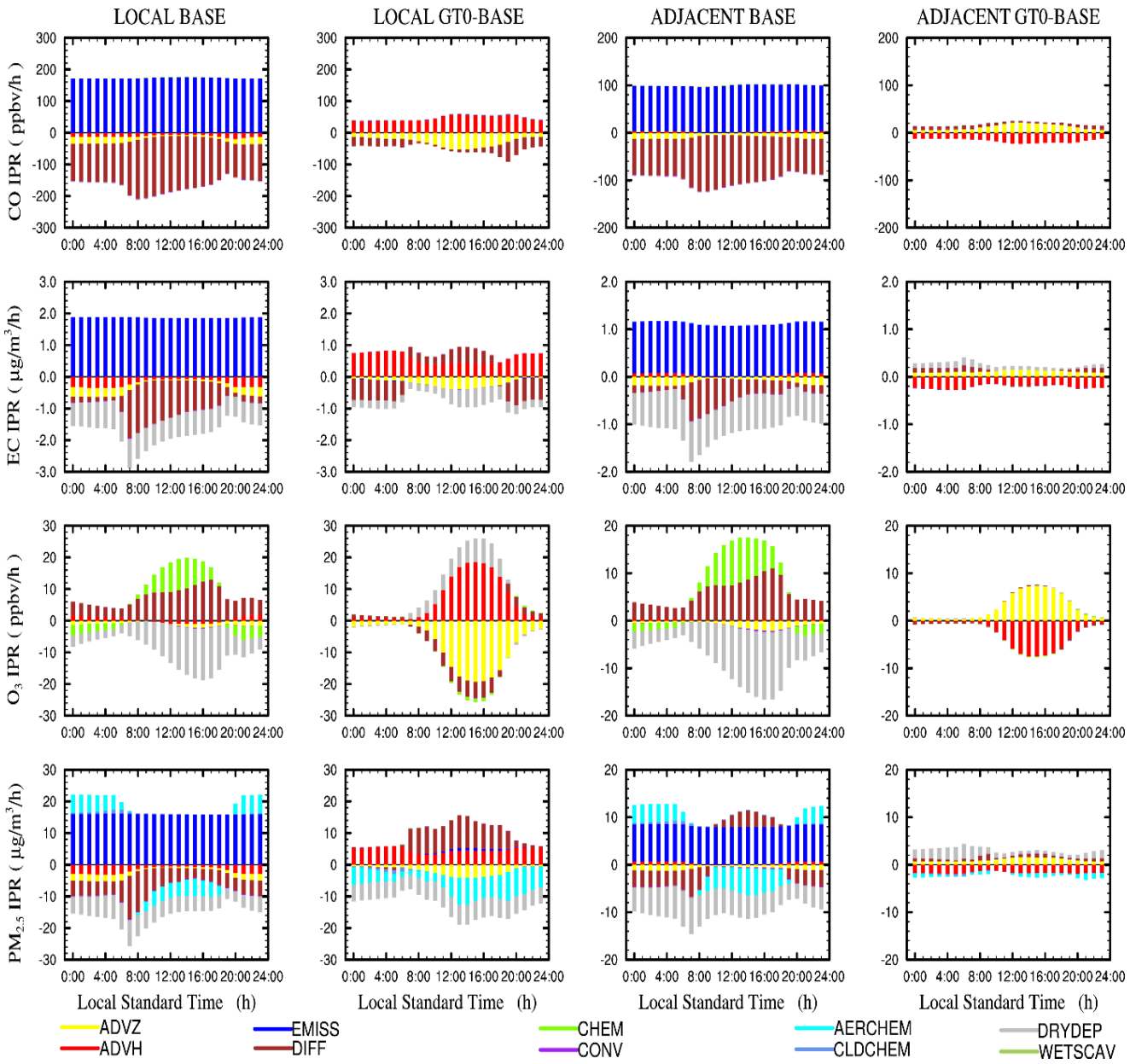
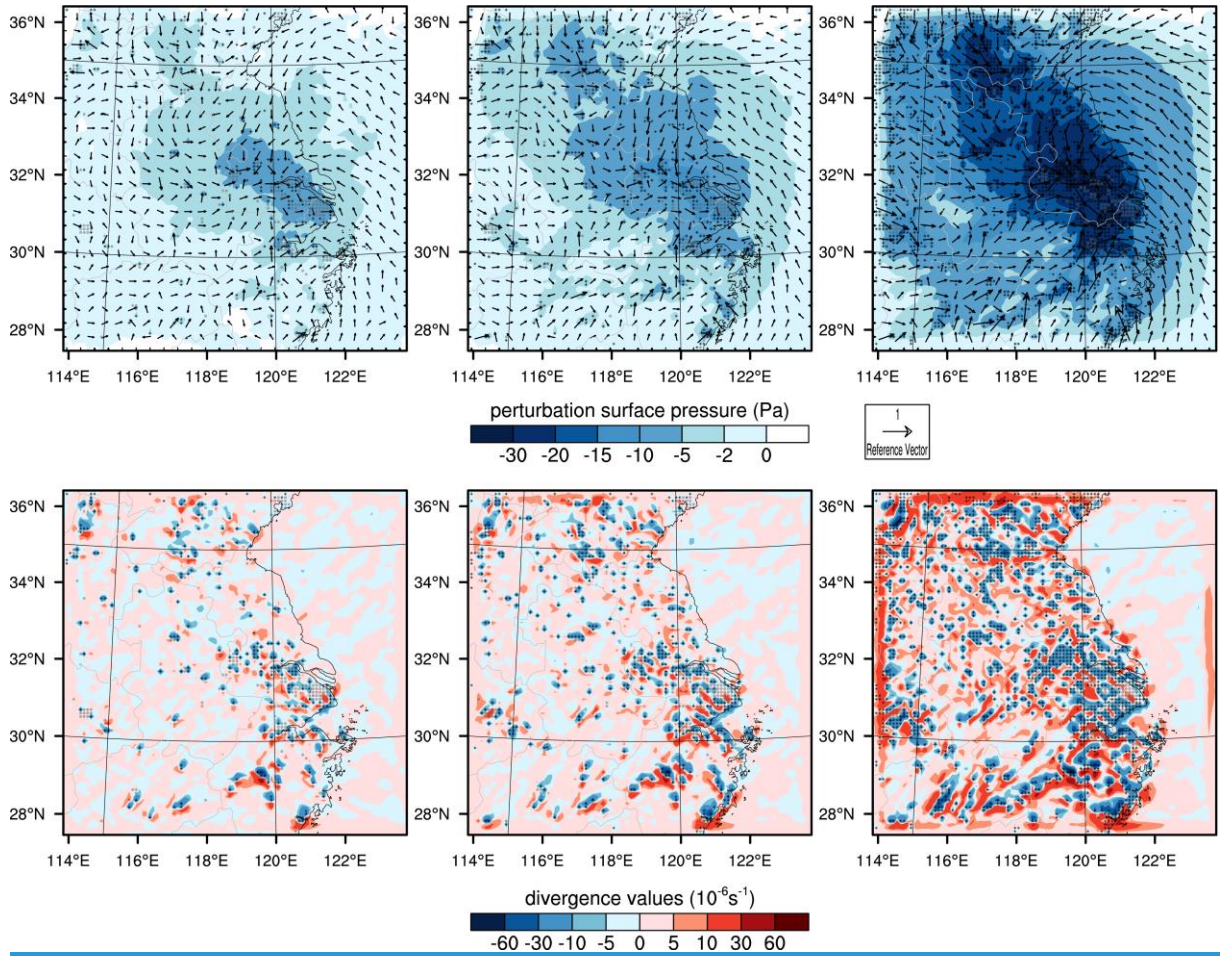


Figure 8. Five-year mean in July diurnal cycles of IPR for surface CO, EC, O_3, and $PM_{2.5}$ concentrations. Values are averaged over all LOCAL (left two columns) and ADJACENT (right two columns) cells. Results are shown for the BASE simulation and for differences between GT0 and BASE.


36°N
34°N
32°N
30°N
28°N
114°E
116°E
118°E
120°E
122°E
perturbation surface pressure (Pa)
-30 -20 -15 -10 -5 -2 0
1
Reference Vector
divergence values ($10^{-6}s^{-1}$)
-60 -30 -10 -5 0 5 10 30 60

1

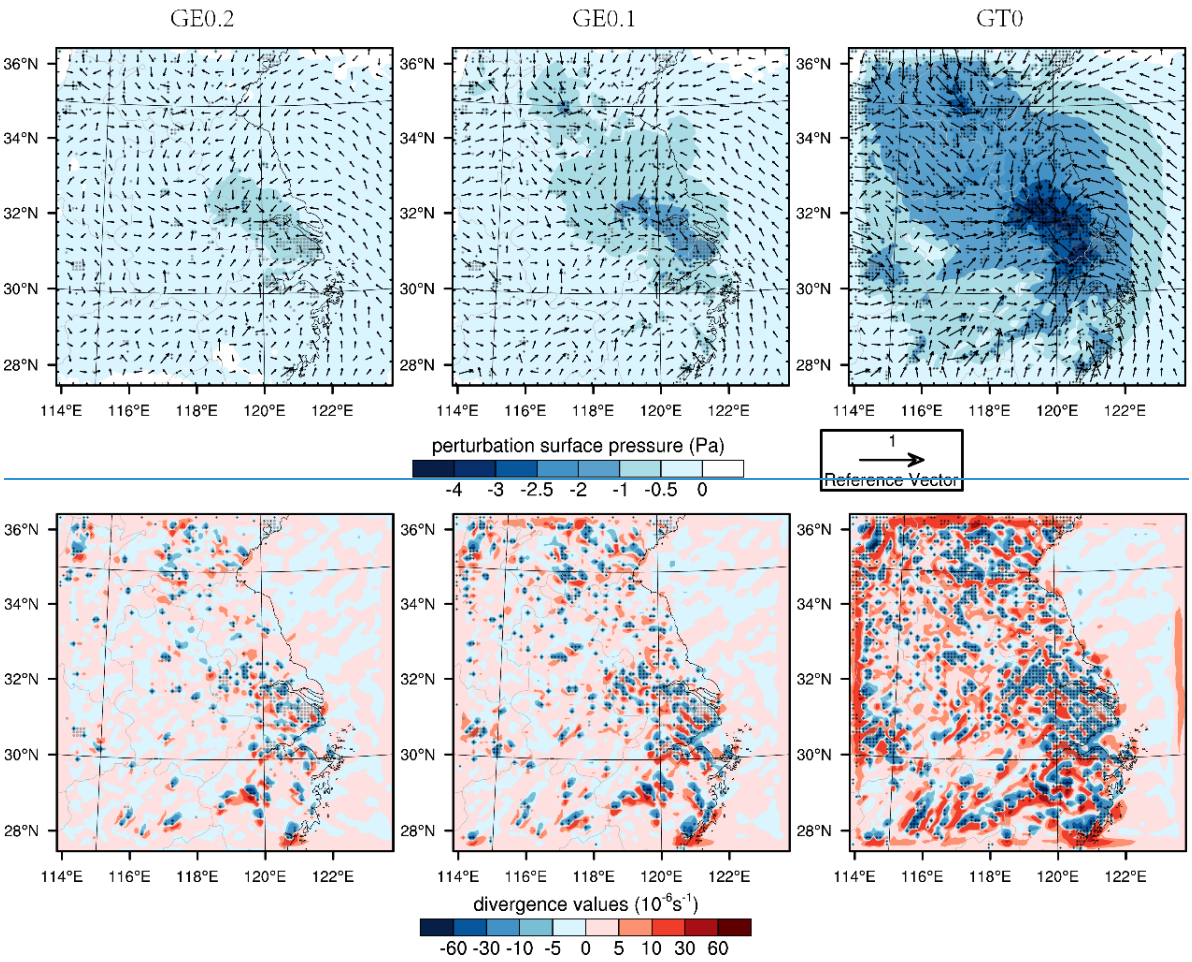


Figure 9. Five-year mean July perturbations of surface pressure and wind field (top three plots, reference velocity is 1 m s^{-1}) and the divergence of surface wind (bottom three plots) in GE0.2, GE0.1, GT0 runs. Grey circles indicate the locations of urban cells in the BASE run; black crosses indicate the locations of newly urbanized areas in GE0.2, GE0.1, and GT0 runs.

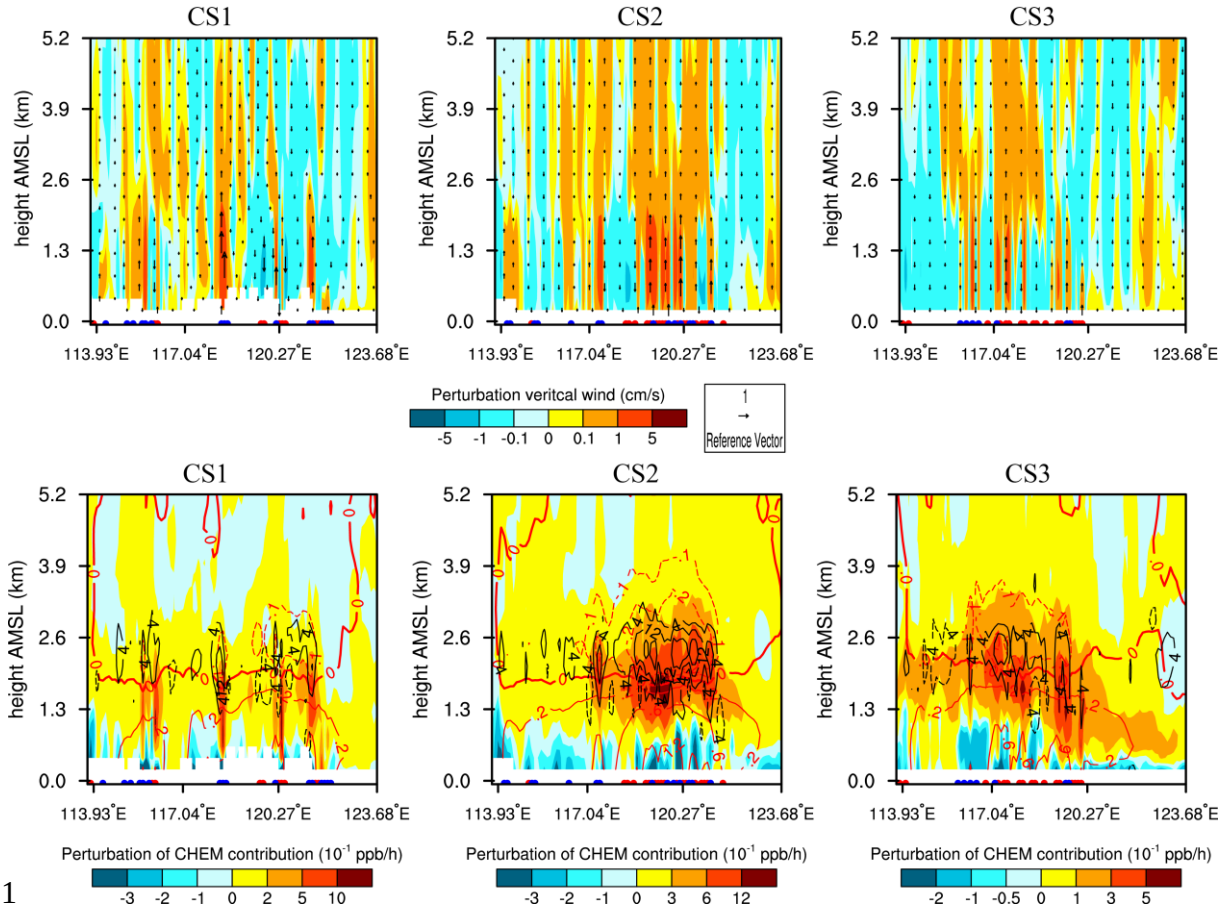


Figure 10. Distribution of 5-year mean July perturbations (GT0 minus BASE) of vertical wind velocities (top three plots, reference wind velocity is 1cm s^{-1}), O_3 production (color, ppb h^{-1}), cloud water content (black line, mg kg^{-1}), and air temperature (red line, K) during 12:00–17:00 LST (bottom three plots) in 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.

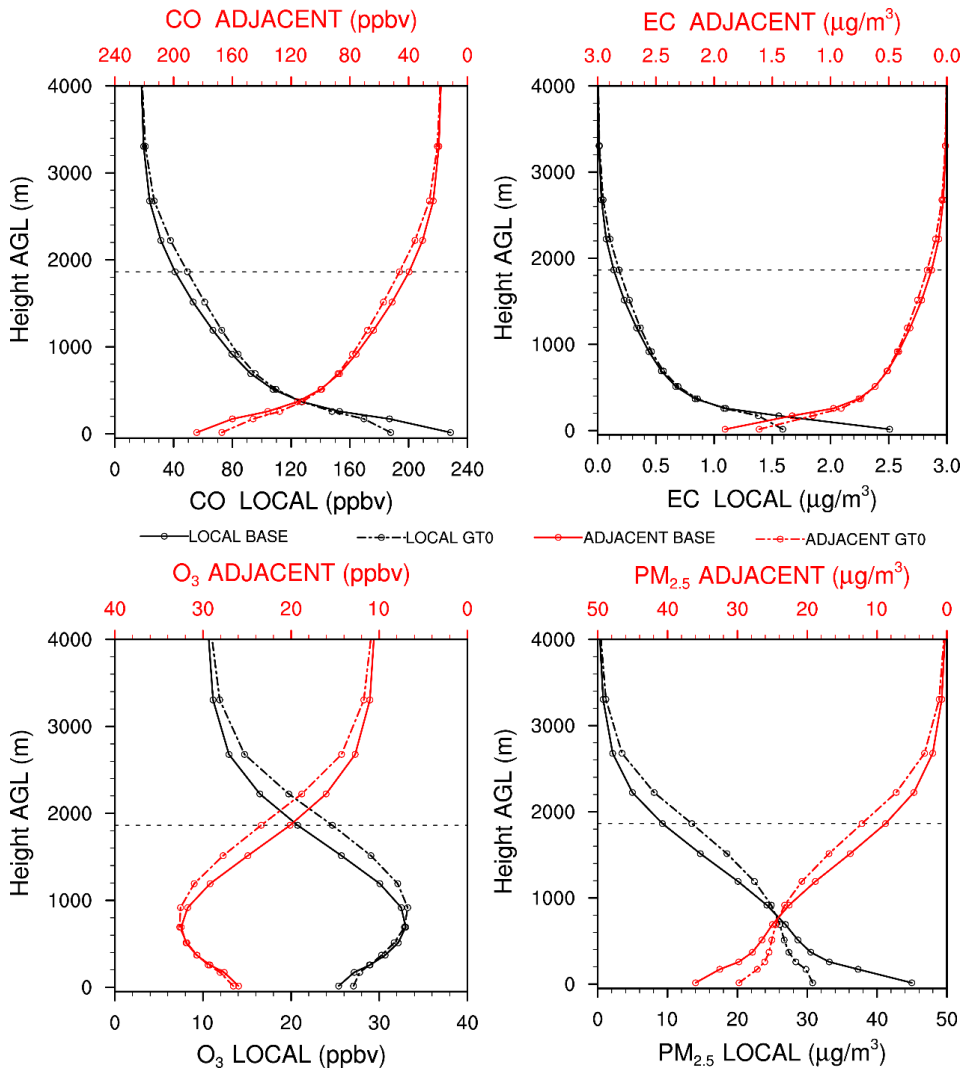


Figure 11. Five-year mean July vertical profiles of CO, EC, O_3, and $PM_{2.5}$ concentrations over the LOCAL cells (black) and ADJACENT cells (red) in the BASE (solid lines) and GT0 run (dashed lines). The horizontal dashed lines indicate the height of 800 hPa.

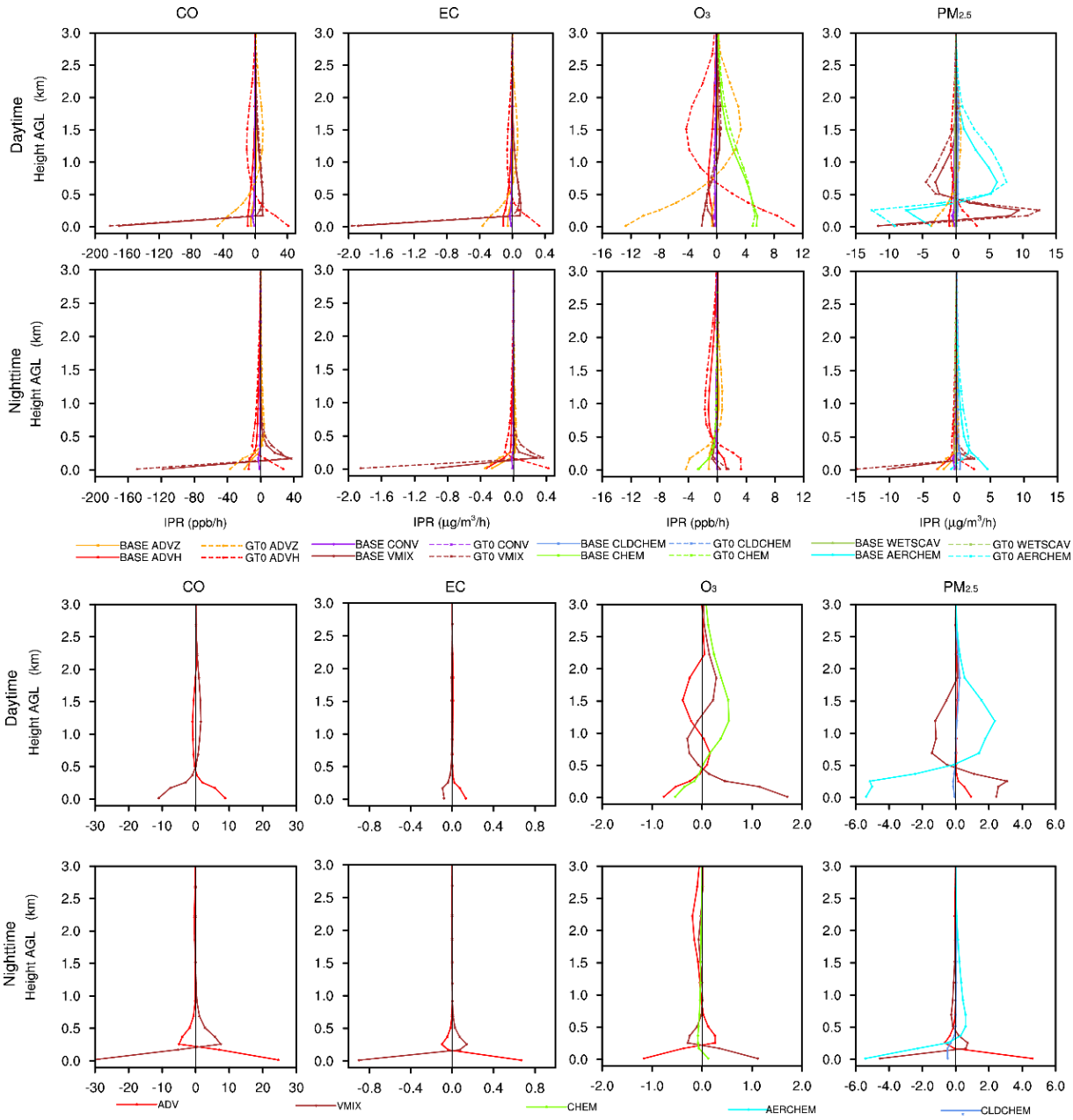


Figure 12. Five-year mean in July vertical profiles of diurnal (07:00–18:00 LST) and nocturnal (19:00–06:00 LST) IPR for CO, EC, O_3, and $PM_{2.5}$ concentrations in the BASE and GT0 run (top eight figures). The bottom eight plots show the difference in IPR between GT0 and the BASE simulation ~~for advection (ADV), vertical mixing (VMIX), gas phase chemistry (CHEM), aerosol processes (AERCHEM), and cloud chemistry (CLDCHEM),~~ averaged over domain-wide LOCAL cells.