Response to Anonymous Referee #1

(Note: Reviewer comments are listed in grey, and responses to reviewer comments are in black. Pasted text from the new version of the paper is in italics.)

The manuscript presents two sets of simulations realized with the model WRF-CHEM coupled with the Single Layer Urban Canopy Model, over the Los Angeles region for a 10 days period at the end of June-beginning of July 2012. One set of simulations is realized with the current land use, including the urban area of Los Angeles. The second set is realized replacing the urban area with shrub, representing the original vegetation (as claimed by the authors). The anthropogenic emissions are the same for both simulations. By comparing the results of the two simulations, authors derive the impact of urbanization on meteorology and air quality in the region.

We greatly appreciate the reviewer's helpful comments. We believe that addressing his/her comments have greatly improved the quality of our paper.

I have two main comments to this manuscript.

a) Authors rely heavily on previous work by the same team (mainly by Vahmani) to justify the set-up used, and the improvements obtained in simulating air temperature (for example due to the inclusion of the irrigation system). However, at lines 358-361, they say that all the previous simulations were performed without accounting for the shadowing effect in the street canyon, and with a different technique to estimate the surface temperature. On the contrary, the simulations presented in the manuscript consider shadowing and use the default formulation to estimate the surface temperature for impervious surfaces. The impact on the results of these different modeling choices seems important to the point that with the new approach urbanization decreases daytime temperature compared to the non-urban case, while with the previous set-up urbanization increased the daytime temperature. While I certainly agree that it is important to account for shadowing, I think that it is necessary to perform a more thorough validation of the simulations to get more confidence in the results, also because the RMSE, presented in table 1, is much larger than the urbanization effect. Therefore, I recommend making a separate analysis of urban and rural stations, and to separate between urban stations based on the different urban morphological characteristics. The validity of this study relies completely on the model capability to reproduce correctly the differences between urban and rural areas, so it is very important to show this comparison. For example, the following questions should be addressed: what are the RMSE and Mean Bias for the urban stations only? And for the rural stations? We have to be sure that the model is simulating correctly the urban areas AND the rural areas (in particular shrubs). Is the model able to capture the maximum and minimum temperature at each station? Is the model able to reproduce the differences between stations, and in particular the differences between the urban and the rural stations? (e.g., if at a certain hour higher temperature is measured in an urban station compared to a rural one, is the model doing the same? If rural stations measured lower minimum (maximum) than urban stations, is the model doing the same qualitatively and quantitatively? etc.).

We thank the author for bringing up these important points. Our detailed response is listed below.

Firstly, after a careful comparison among different model set-ups, we found that the parameterization of surface temperature is the more important factor that affects daytime air temperature compared to whether or not we account for shadow effects in urban canopies (i.e., see Figure S16 in the supplemental information). Therefore, we delete "and shading effects within urban canopies" in the sentence "In addition, increases in thermal inertia caused by use of manmade materials (e.g., pavements and buildings) and shading effects within urban canopies can contribute to simulated temperature reductions during the morning." We also modified the last paragraph in section 3.2.3 as below.

"Note that changes in air temperature during daytime shown here disagree with Vahmani et al. (2016). While our study detects daytime temperature reductions due to urbanization, Vahmani et al. (2016) suggests daytime warming. After detailed comparison of the simulations in our study versus Vahmani et al. (2016), we find that the differences are mainly associated with UCM configuration. First, our study uses model default calculations of surface temperature for the impervious portion of urban grid cells, whereas Vahmani et al. applied the alternative calculation proposed by Li and Bou-Zeid, 2014. Li and Bou-Zeid, 2014 intended the alternate surface temperature calculation to be performed as a post-processing step rather than during runtime. After a careful comparison among different model set-ups, we found that the parameterization of surface temperature is an important factor that affects simulated daytime air temperature (See Figure S16). Second, our study accounts for shadow effects in urban canopies, whereas Vahmani et al. (2016) assumes no shadow effects. (We note here that the default version of the UCM has the shadow model turned off. The boolean SHADOW variables in module_sf_urban.F needs to be manually switched to true to enable the shadow model calculations. With the shadow model turned off, all shortwave radiation within the urban canopy is assumed diffuse.) We suggest that it is important to include the effects of building morphology on shadows within the canopy, and to track direct and diffuse radiation separately, and therefore perform simulations in this study with the shadow model on. Note that the effect of shadowing is not as significant as the parameterization of surface temperature in our study, because the ratio between building height and road width is small."

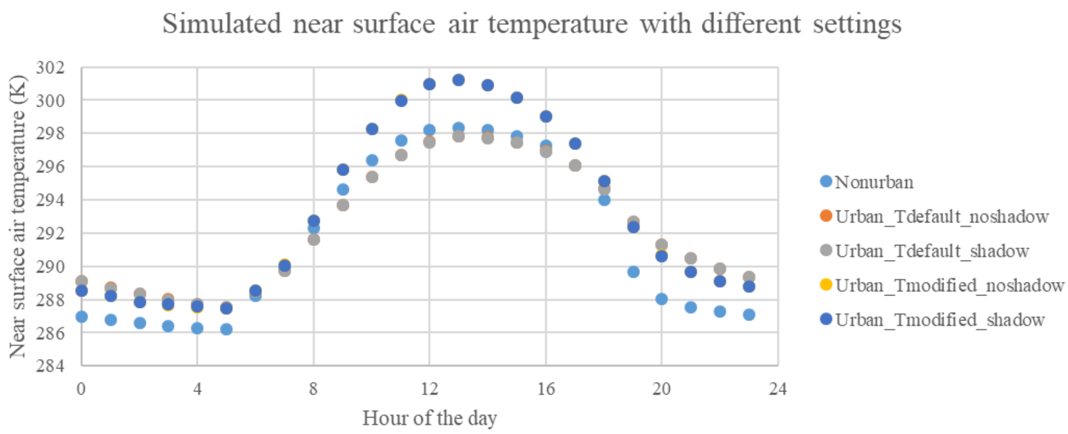

Figure S16. Diurnal cycle of near surface air temperature simulated with different model set-ups. "Tdefault" indicates that the simulation uses the default calculation of surface temperature in WRF, while "Tmodified" indicates that the simulation uses the calculation of surface temperature from Li and Bou-Zeid (2014) (which is also used in (Vahmani el al. (2016)). Dots for "Urban_Tdefault_shadow" and "Urban_Tdefault_noshadow" ("Urban_Tmodified_shadow" and "Urban_Tmodified_noshadow") are overlapping at every hour of the day because the simulation results with shadow on/off are very similar.

Secondly, we suggest that the comparison of urbanization impacts versus RMSE in Table 1 is not a robust comparison. Instead, to assess whether urbanization impacts are statistically distinguishable from zero, we added a new statistical analysis to the paper, using the paired Student's t-test with n = 7 days. We did the test to check 1) whether spatially averaged changes in regional meteorology and air quality are statistically significant within the simulation period, and 2) whether spatially resolved changes in regional meteorology and air quality are significant within the simulation period (i.e., for each urban grid cell). For 1), we edited the relevant sentences in the paper that refer to spatial average changes. For 2), we updated all figures with maps in the paper by adding black dots to grid cells with insignificant changes. Please see section 2.5 and section 3 in the paper for these changes. We haven't pasted the changes here because they are distributed throughout our results section, and would take up over 3 pages in this document.

Thirdly, we agree with the reviewer that the validation of urban sites, nonurban sites (shrubs in particular) and the difference between urban versus nonurban sites is important. In the main paper we only showed the validation of urban sites (i.e., we added a sentence to point this out in section 3.1.). Thus, we added a section in the supplemental information (section S2) discussing this topic. In general, our model can capture observations at nonurban sites, and the difference between urban versus nonurban sites. Thus, we believe the results we obtain based on this model set-up are reliable. The text in the paper are as follows (see below).

In the main paper:
"In addition, we only include observations from monitoring sites that are located in urban grid cells in the Present-day scenario. The validation of near surface air temperatures for both urban and nonurban sites are discussed in section S2 in the supplemental information."

Please see section S2 in the supplemental information for more details on the validation. We again do not paste it here because it is ~3 pages.

b) It must be made clear that the simulation with current anthropogenic emissions, but not the city, is a hypothetical one – there cannot be emissions without a city. In the last sentence of the manuscript (lines 570-574), authors say that their results "can be informative for decision making on sustainable urban planning to achieve a balance between climate mitigation/adaptation and air quality improvements". Honestly, I do not see how. This type of studies may have a scientific value, in the sense that they demonstrate the importance of taking into account the presence of the city in the simulation of air quality and meteorology (it would be interesting to see if the simulation with the city provides better results compared to measurements than the simulation without the city). But I do not see how they can be helpful for urban planning. Replacing the city with shrubs cannot certainly be considered a strategy to manage urban climate or improve air quality. The differences that authors estimated between the urban and the no-urban simulations are not the maximum difference that can be obtained managing the land use. They actually do not give any information about the impact of any realistic mitigation strategy based on land use management. I think it is very important that authors clarify what they have in mind because this is at the basis of the motivation of the whole manuscript.

We thank the reviewer for bringing this up. To emphasize that the "Nonurban" simulation is a hypothetical scenario, we added the following sentences to section 1, and revised sentences in section 2.5 and section 4.

Section 1
"In this paper, we aim to quantify the importance of historical land cover change on air pollutant concentrations, and thus the "Nonurban" scenario assumes current anthropogenic pollutant emissions. This hypothetical scenario cannot exist in reality, since current anthropogenic emissions would not exist without the city, but our intent is to tease out the relative importance of land cover change through urbanization (assuming constant emissions) on air pollutant concentrations."

Section 2.5
"Note that all three aforementioned scenarios adopt identical anthropogenic emission inventories described in Section 2.3. Using current anthropogenic emissions for "Nonurban" is a hypothetical scenario that cannot exist in reality, but allows us to tease out the effects of land surface changes via urbanization on meteorology and air pollutant concentrations."

Section 4
"The two main simulations of focus in this study are the real-world "Present-day" and the hypothetical "Nonurban" scenarios…"

We agree with the reviewer that the last sentence of the manuscript might not be an appropriate implication of the findings in this study. Therefore, we deleted that sentence, and added the following paragraph to the manuscript in section 4.

"This study highlights the role that land cover properties can have on regional meteorology and air quality. We find that increases in evapotranspiration, thermal inertia, and surface roughness due to historical urbanization are the main drivers of regional meteorology and air quality changes in Southern California. …Our findings indicate that air pollutant concentrations have been impacted by land cover changes since pre-settlement times (i.e., urbanization), even assuming constant anthropogenic emissions. These air pollutant changes are driven by urbanization-induced changes in meteorology. This suggests that policies that impact land surface properties (e.g., urban heat mitigations strategies) can have impacts on air pollutant concentrations (in addition to meteorological impacts); to the extent possible, all environmental systems should be taken into account when studying the benefits or potential penalties of policies that impact the land surface in cities."

Detailed comments:
1) Lines 64-66. Urban regions in semi-arid or arid surroundings have a weak (or non- existent) daytime UHI, but they have a very strong nocturnal UHI. I think authors missed the fundamental difference between daytime and nighttime UHI, (being the latter the most frequent).

Thank you for catching this mistake. We modified the sentence as follows.

"In particular, urban regions built in semi-arid or arid surroundings tend to have a weak daytime UHI or even a UCI, whereas those built in moist regions tend to have a larger daytime UHI (Fan et al., 2017; Peng et al., 2012)."

2) Line 168. On which basis authors claim that the period chosen is representative of summer conditions in Southern California?

Typical summer days in Southern California are clear or mostly sunny days without precipitation. The chosen simulation period has these characteristics. We added a figure in the supplemental information (Figure S8) showing the diurnal cycle of averaged (observed) near surface air temperature over JJA (June, July and August) and over our simulation period. We also added a sentence in the main paper.

"This simulation period is chosen as representative of typical summer days in Southern California, which are generally clear or mostly sunny without precipitation. A comparison of observed diurnal cycles for average near surface air temperatures over JJA (June, July and August) versus over our simulation period is shown in Figure S8 in the supplemental information."

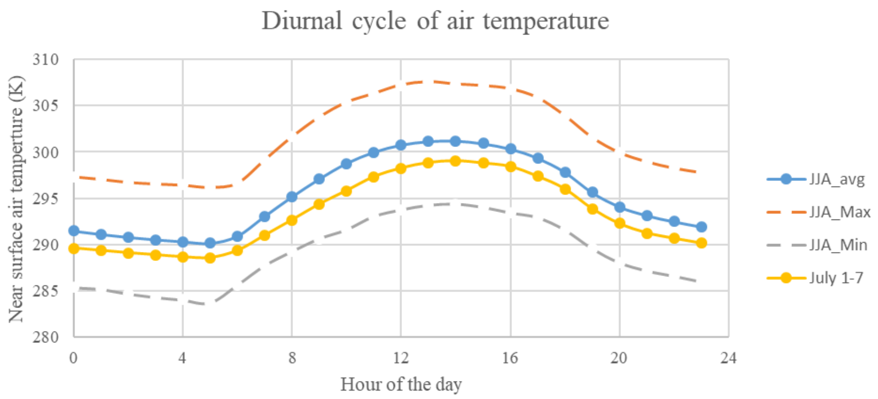

Figure S8. Diurnal cycles for observed near surface air temperature (K) over JJA (June, July and August) in blue, and over our simulation period in yellow. Observations are obtained from MesoWest (https://mesowest.utah.edu/), which are available at Mesonet API (https://developers.synopticdata.com/mesonet/). Mean values are derived by averaging over all observational sites available for the innermost domain and the aforementioned period for each hour of day. Orange and grey curves show the maximum and minimum air temperature at each hour of the day for JJA. Results show that our simulation period (July 1-7) is representative of summertime meteorology for our domain.

3) Line 174. Please provide the value of the depth of the lowest model level.

Thanks for the suggestion. We added it to the manuscript.

"The average depth of the lowest model level is 53 m for all three domains."

4) Line 215. Is the irrigation module implemented just for the pervious fraction of the urban cells, or also for the rural cells (to account for agricultural crops in the region)?

The irrigation module is just for the pervious fraction of the urban grid cells. The rural grid cells surrounding urban regions in Southern California are mostly natural land cover (e.g., shrub lands), and do not need to take irrigation into account. We added this information to the related sentence.

"Here we use an irrigation module developed by Vahmani and Hogue (2014), which assumes irrigation occurs three times a week at 2100 PST on the pervious fraction of urban grid cells."

5) Line 302. I would avoid indicating the percentage for temperature. This would depend on the unit (if you use Celsius or Kelvin). I would just put degrees.

Thanks for the suggestion. We have gone through the manuscript and changed all expressions of temperature in percentage to absolute values in Kelvin.

6) Line 303. On which basis authors claim that this is "acceptable".

We agree with the reviewer that "acceptable" is somewhat vague and unclear, so we modified the sentence as below, and added the comparison between our evaluation results and recommended model performance benchmarks to the supplemental information.

"The statistical results indicate that while model simulations underestimate near-surface air temperature, O_3 and $PM_{2.5}$ concentrations by 1.0 K, 22% and 31%, respectively. The comparison between our evaluation results and recommended model performance benchmarks is presented in supplemental information Table S2."

7) Section 3.2.3. I suggest studying the difference in sea breeze front progression between the two cases (urban and no-urban). This will give a better understanding of what is happening.

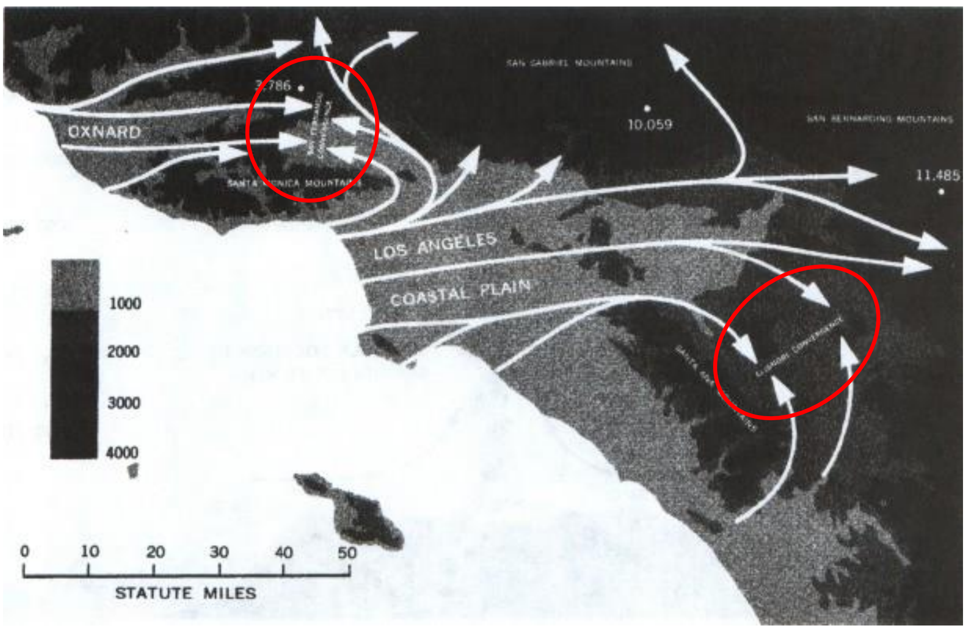

Figure R1.1 Sea breeze flow in the Los Angeles basin. Note the two sea breeze fronts formed at San

Fernando convergence zone (upper left), and the Elsinore convergence zone (lower right). (Figure adopted from: https://www.aviationweather.ws/097_Sea_Breeze_Soaring.php)

Figure R1.1 shows the two major sea breeze convergence phenomena in Los Angeles basin occurring in the San Fernando convergence zone and the Elsinore convergence zone. They are both formed by the meeting of two sea breezes that had flowed around the mountains. The sea breeze that flows across the Los Angeles coastal plain extends into the Mojave Desert. The sea breeze front of this branch of flow is not shown in our innermost domain. Therefore, we present here the difference in the wind vectors in the lowest model layer from 10 am to 5 pm instead of the progression of sea breeze front (Figure R1.2). The wind vectors are pointing towards the sea, which indicates that there is a significant decrease in wind speed in all hours of day presented here.

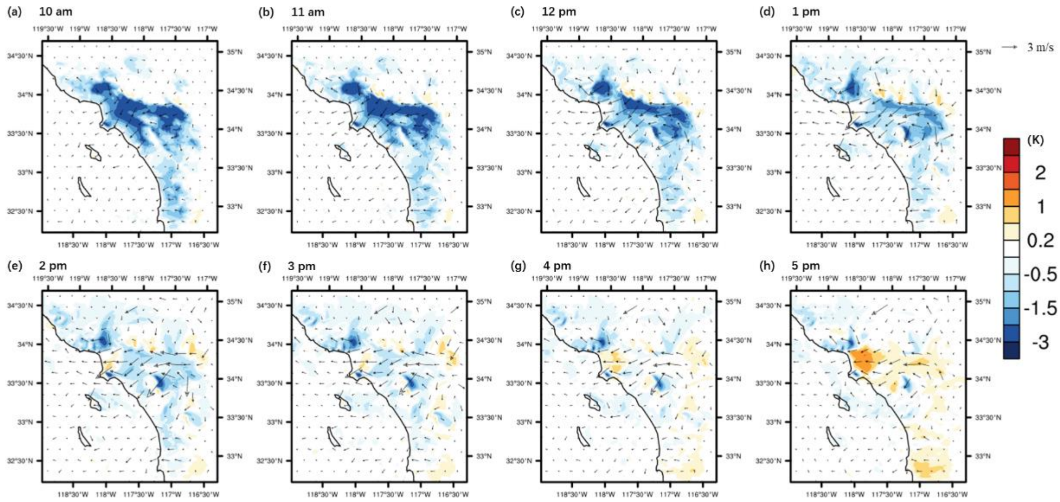

Figure R1.2 Wind vector plots for differences (Present-day – Nonurban) in temporally averaged wind speed in the lowest model layer from 10 am to 5 pm. The background plot shows the differences (Present-day – nonurban) in air temperature.

8) Lines 335-338. This is not clear. Before it is said that urbanization decreases temperatures and not increases.

The averaged temperature in the urban region decreases during the morning and afternoon. However, temperature changes vary spatially. While reductions in air temperature occur across the urban region during the morning, and in the inland urban region during the afternoon, temperatures actually increase in the coastal region during the afternoon. We specified the time when increases in temperature occur in the sentence.

"Note that the onshore sea breeze decreases in strength despite higher temperatures in the coastal region of Los Angeles during the afternoon, which would tend to increase the land-sea temperature contrast and thus be expected to increase the sea breeze strength."

9) Line 370. During nighttime atmosphere cools. The energy stored in the building during daytime (what authors call upward ground heat flux, I suppose) reduces the cooling. The higher PBL in the

urban simulation will reduce the cooling too because the effect of the surface cooling is distributed in a greater depth than in the no-urban case. The two mechanisms (energy stored in buildings, and high PBL), both reduce cooling. They do not compete they go in the same direction.

Thank you for catching this mistake. We revised the related sentences in section 3.2.4.

"The climate response to urbanization during nighttime is driven by the combined effects of (a) temperature increases from increasing upward ground heat fluxes, and (b) temperature increases from increasing PBL heights. Increased soil moisture (from irrigation) and use of man-made materials leads to higher thermal inertia of the ground; this in turn leads to increased heat storage during the day and higher upward ground heat fluxes and thus surface temperatures at night. Increasing PBL heights can also lead to warming because of lower air cooling rates during nighttime. Changes in PBL heights are associated with surface roughness changes since shear production dominants TKE at night. Coastal (inland) regions show larger (smaller) variation in roughness length (Figure 2e), which leads to larger (smaller) increases in PBL heights (Figure S14c). Despite larger increases in PBL heights in coastal versus inland regions, smaller air temperature increases occur in coastal versus inland regions, likely due to accumulative effects from coastal to inland regions with onshore wind flows."

10) Lines 375-376. Same as above, during the night there is not heating, there is cooling.

Please refer to our response to comment 9.

Response to Anonymous Referee #2

(Note: Reviewer comments are listed in grey, and responses to reviewer comments are in black. Pasted text from the new version of the paper is in italics.)

Dear authors, the paper is well written and clearly structured, however I would recommend a number of major changes in order to be suitable for publication. Please find my comments below:

We thank the reviewer for his/her thoughtful and valuable comments. These comments substantially help to improve our manuscript by addressing these issues.

General: I see a general problem in the definition of the scope of the study. A 'before human settlement' scenario should not consider emissions at all and further describes a period about 100-150 years ago which means that you would also have to consider a different climate period, land use etc.. I definitely would recommend to redefine the scope of the study, because in the current state, just distinguishing between 100% urban vs. 0% urban is not sufficient to analyze the above mentioned scenario.

We thank the reviewer for bringing this up. The "Nonurban" simulation in this study is a hypothetical scenario in which we assume current anthropogenic emissions and climate, but natural land cover prior to human perturbation. By doing so, we focus our study on the relative importance of land cover changes via urbanization on regional meteorology and air quality. To make it clearer in the paper that the "Nonurban" simulation is a hypothetical rather than realistic scenario, we added the following sentences to the introduction (section 1), and modified relevant sentences in section 2.5 and the conclusion (section 4).

Section 1

"In this paper, we aim to quantify the importance of historical land cover change on air pollutant concentrations, and thus the "Nonurban" scenario assumes current anthropogenic pollutant emissions. This hypothetical scenario cannot exist in reality, since current anthropogenic emissions would not exist without the city, but our intent is to tease out the relative importance of land cover change through urbanization (assuming constant emissions) on air pollutant concentrations."

Section 2.5

"Note that all three aforementioned scenarios adopt identical anthropogenic emission inventories described in Section 2.3. Using current anthropogenic emissions for "Nonurban" is a hypothetical scenario that cannot exist in reality, but allows us to tease out the effects of land surface changes via urbanization on meteorology and air pollutant concentrations. "

Section 4

"The two main simulations of focus in this study are the real-world "Present-day" and the hypothetical "Nonurban" scenarios"

I am further not fully convinced about the added benefit of this study for sustainable urban planning recommendations. I am aware that these model systems are not suitable for applied urban planning, but however the currently existing urban canopy models in WRF-Chem (and other models), together with high resolution datasets for both emission and urban morphology do offer a framework for a number of different scenarios in the context of climate change/UHI mitigation. Recent studies have been analyzing the impact of highly reflecting building materials, urban greening or varying building density for a number urban areas. These aspects should also be possible with this model system and worth being discussed in order to increase the scientific substance of that work and highlight the new contribution to the field. In light of the scope of the journal, it should also be worked out more detailed what are the implications for atmospheric science in general rather than purely investigating local/regional aspects.

We agree with the reviewer that our description of the implications of this study were somewhat ambiguous in the submitted version of the paper. The main point that we intend to make here is that land surface changes on their own can have a significant influence on regional air quality via altered meteorological conditions. Therefore, we should consider the benefits and penalties of UHI mitigation strategies (i.e., since most of them modify land surface properties) from the viewpoint of both climate and air quality to achieve a comprehensive assessment. We revised the conclusion section (section 4) as follows:

'This study highlights the role that land cover properties can have on regional meteorology and air quality. We find that increases in evapotranspiration, thermal inertia, and surface roughness due to historical urbanization are the main drivers of regional meteorology and air quality changes in Southern California. …Our findings indicate that air pollutant concentrations have been impacted by land cover changes since pre-settlement times (i.e., urbanization), even assuming constant anthropogenic emissions. These air pollutant changes are driven by urbanization-induced changes in meteorology. This suggests that policies that impact land surface properties (e.g., urban heat mitigations strategies) can have impacts on air pollutant concentrations (in addition to meteorological impacts); to the extent possible, all environmental systems should be taken into account when studying the benefits or potential penalties of policies that impact the land surface in cities."

I am convinced, that the model system, combined with the emission and land surface data sets offer a promising tool for discussing air quality/meteorology interactions in large urban areas such as Los Angeles, but however think that the variety of scenarios should be increased in order to allow for a more robust results towards currently relevant issues. The authors rely mostly on previous work with equal model configuration. Therefore, the own contribution to the field and the new development does not come out clearly. The paper however is well written and easy to follow, but crucial points have to be considered in a review before being able undertake a detailed line-by-line evaluation.

The study certainly builds on our prior work, but this paper focuses on air quality impacts, whereas our previous research was on only meteorology. Thus, the most important contribution of this work is that we investigate a totally different environmental system than previous work. In order to do so, we also add a new modeling component (atmospheric chemistry) that is not presented in past work.

Other smaller additions compared to our past work is that we turn on the shadow model and incorporate GIS-based building morphologies, which make the model simulations more representative of current day weather conditions in LA. Moreover, while the influence of land use changes on regional weather has been well studied, its influence on regional air quality has been seldom studied with accurately resolved land surface data, especially in the Southern California region. Therefore, our study fills this research gap. We added several sentences in the last paragraph of introduction section to emphasize this point.

"Note that this paper builds on our prior study Vahmani et al. (2016), but focuses on air quality impacts, whereas our previous research was on meteorological impacts only. While the influence of land surface changes on regional weather has been investigated in numerous past studies, its influence on regional air quality has been seldom studied in past work."

Moreover, the focus of this study is on the impact of land surface changes on regional meteorology and air quality. Thus, the two major scenarios discussed are "Nonurban" and "Present-day" scenarios, which characterized land surface prior to human perturbation and under current conditions respectively. We also included a supplemental scenario "Present-day No Irrigation" that teases out the effects of irrigation.

1. The scope of the study should be defined more clearly in light of the above mentioned points. The experimental design should be expanded, in order to include more own ideas/developments.

As mentioned in our previous responses, we focus this study on the relative importance of land cover changes via urbanization on regional meteorology and air quality, and assume identical climate and anthropogenic emissions in both scenarios. In our simulations, we implemented real-world representation of land surface properties in the "Present-day" scenario, which made it possible to tease out the most important land surface factors. Our results indicate that land surface changes have a significant influence on regional air quality via altered meteorological conditions. This suggests that policies that impact land surface properties should take all environmental systems into account when studying the benefits or potential penalties of the policies. We feel that this is a solid focused story for the paper, and adding additional simulations would only dilute the main points we are trying to make. In other words, adding more complexity to the study would only muddle the story.

2. One interesting and highly relevant point in my opinion is the 'irrigation' module which might offer a nice tool for testing different irrigation scenarios.

We agree that the proposed research idea is an interesting topic. However, it would be more appropriate as an individual study on the influence of irrigation on regional climate and air quality. This isn't the main research question we are trying to answer. In this study, we want to keep the scope well defined in answering the posed research questions on how historical land surface changes have affected regional climate and air pollutant concentrations in Southern California. Thus, investigating the regional influence of irrigation sounds interesting but beyond our motivation and scope.

3. Why did you select a single-layer urban canopy model rather that a more complex multi-layer canopy representation (BEP/BEM)? The latter should deliver higher accuracy close to the ground I guess? What is the depth of the lowest model level?

As suggested by Kusaka et al. (2001) , the model performance of UCM and BEP/BEM with regard to studying mesoscale heat islands are comparable. Chen et al. (2011) also mentions that the UCM may be more suitable than BEP/BEM for weather and air quality prediction. In addition, coupling BEP/BEM to WRF/Chem would be an extremely complex model development exercise, and the resulting model would be prohibitively computationally expensive, but for likely little additional benefit in the quality of simulations. Therefore, we choose to couple the UCM instead of BEP/BEM to WRF/CHEM.

The averaged depth of the lowest model level is 53 m for all three domains. This information has been added to the paper at the last sentence in section 2.1.

"The average depth of the lowest model level is 53 m for all three domains."

4. Where do the input parameters for SLUCM come from?

We use NLCD impervious surface data for impervious fraction of each grid cell. For surface albedo of roof, building wall, and road, we assign the grid cell albedo value derived from MODIS. Building morphologies (including building height, standard deviation of roof height, building width and road width) are from NUDAPT where available. Where NUDAPT data are unavailable, we adopt average building and road morphology from LARIAC. This information is mentioned in section 2.2. For the other parameters in UCM (e.g., anthropogenic latent heat, surface emissivity), we use default WRF settings documented in file URBPARM.TBL. We added this information to section 2.2.

"For the other parameters in the UCM (e.g., anthropogenic latent heat, surface emissivity), we use default WRF settings documented in file URBPARM.TBL."

5. What is the additional gain of a 30 m land surface classification which has to be scaled to 2 km model resolution?

We chose to use 30 m-resolution 33-category NLCD mainly for two reasons. First, urban land use varies at spatial scales on the order of 10s of meters. So it works best to define land use at spatial scales of 10s of m, and then aggregate to the model grid resolution. It would be difficult to detect land use using data at 2km resolution. Second, the 30 m-resolution land use dataset has 33 categories of land use type, which divides urban type into three sub-types: low-intensity residential, high-intensity residential, and industrial/commercial. This allows different parameterizations for different sub- urban types, which better characterize land surface properties.

6. Is there a problem with regard to the discrepancy between emission inventory and model resolution?

No, there should not be a problem. The resulting air quality predictions are simply lower resolution than they would be if they were at 2km. We ensured that the total emissions within in the domain are kept consistent after regriding.

7. How realistic is the surrounding 'non-urban' land use classification for the 'historical' scenario?

The dominant natural land cover type surrounding Los Angeles and San Diego metropolitan areas is shrub. So it is reasonable to assume shrub as the land use type in the "Nonurban" scenario. The land surface properties of these grid cells in the "Nonurban" scenario are derived using the inverse distance weighting approach, which is mentioned in section 2.5, and consistent with our previous publication.

"For the Nonurban scenario, we assume natural land cover prior to human perturbation, and replace all urban grid cells with "shrubs" (Figure 1c). We modify MODIS-retrieved albedo, GVF and LAI in these areas based on properties for shrub lands surrounding urban regions in the Present-day scenario. A detailed explanation on this method (inverse distance weighting approach) can be found in Vahmani et al. (2016)."

8. How well does the model simulate urban AND rural parameters?

In section 3.1, we showed how well the model simulated urban variables (i.e., near surface air temperature, and pollutant concentrations). We agree with the reviewer that it is also important for the model to capture nonurban (especially shrub) air temperatures well, thus we included a discussion on this topic in section S2 in the supplemental information. Note that nonurban observational sites that measure pollutant concentrations are rare. Thus, we decided not the discuss how well the model simulated pollutant concentrations in nonurban area. The new text in the main paper is pasted below. Please see section S2 in the supplemental information for more details on the validation. We did not paste it here because it is ~3 pages.

In the main paper:
"In addition, we only include observations from monitoring sites that are located in urban grid cells in the Present-day scenario. The validation of near surface air temperatures for both urban and nonurban sites are discussed in section S2 in the supplemental information."

9. Please specify how results from this study can serve as contribution for applied urban planning.

As mentioned in our response to your second general concern, we changed the last paragraph in the conclusion section, which explains the implications of our study. Please see that response for more detail.

10. In relation to other chapters, the introduction is slightly too long. Try to focus on the relevant points here and shorten were possible.

We think that the background knowledge, brief literature review, and research gaps described in the introduction section are necessary for a clear explanation of the scope and motivation of this study.

The flow of the introduction section is as follows. First, we point out that urbanization has led to profound modification of the land surface. We then explain how changes in land surface properties can affect regional meteorological fields such as surface and air temperature, wind speed and PBL height. We go on to demonstrate how those changes in meteorology due to land surface modification can in turn affect air pollutant concentrations via different mechanisms. While there are a number of studies that have investigated the impacts of land surface changes on regional meteorology, limited studies have quantified the impact of land surface changes on regional air quality, especially for the Southern California region, which has a history of severe air pollutant problems. In addition, recent studies have made it possible to utilize satellite land surface data in model simulations, which better predict regional weather in urbanized regions, and urban versus nonurban differences. Thus, our study adopts the modified model configuration, and aims to characterize the influence of historical urbanization on urban meteorology and air quality in Southern California.

Please find below comments for specific sections, which partly have been addressed in the main points above.
Ln 11: ventilation not a good expression here

We think that "ventilation" is a proper expression here because it appropriately describes the ability of atmosphere to transport pollutants out of the studied area.

13: 'before human settlement' is a bit misleading here, as it is not entirely captured by your model setup. As mentioned before, some effort has to be put in a clear definition of the scope of your study. What problem should be addressed – also in light of recommendations for real urban planning (Lines 570-573?

The two concerns mentioned in this comment are addressed in the first two general comments respectively.

43: "Differences in surface temperature..." What was the purpose of these studies mentioned here and what do they try to answer? How does this sentence relate to your study and the intention for this work?

The UHI and UCI represents urban versus nonurban difference in surface or air temperature. They are both climate phenomena at urban scale that occur due to variability in land cover changes. Here we summarize possible ways in which land surface modifications can affect surface/air temperature difference between urban and nonurban areas, which is what causes the UHI/UCI. The temperature difference between the "Present-day" scenario and "Nonurban" scenario discussed in our study is analogous to the UHI/UCI. Thus, the background information here is necessary.

47: "UCI": How does this relate to your study?

This is explained in the response to the comment above.

67: What is the role of the atmospheric aerosol burden for UHI formation?

The role of atmospheric aerosol on UHI intensity is an active research topic, and yet no consensus has been reached. For example, Kumar et al. (2017) carried out a Global Climate Model simulation, and suggested that daytime cooling (UCI) can be partially attributed to absorbing aerosols over Indian cities. Cao et al. (2016) used satellite observations, and found positive correlation between urban–rural difference in AOD and nighttime UHI.

73: better "characteristics/shape of the PBL is dependent on..."

Changed. Thanks!

81: better "due to urbanization..."

Changed. Thanks!

86: unclear what is meant by "meteorological changes via altered emissions,..."

We changed the sentence as follows:

"Meteorology can affect emission rates, chemical reaction rates, gas-particle phase partitioning of semi-volatile species, pollutant dispersion, and deposition; thus, it plays an important role in determining air pollutant concentrations."

115: Why do higher PBLs increase $PM_{2.5}$ concentration? Please discuss the related processes here.

In our text, we mentioned that Chen et al. (2018) found that higher PBLs decrease $PM_{2.5}$ concentrations. Please find the original sentence below:

"Chen et al. (2018) studied urbanization in Beijing, and found that modification of rural to urban land surfaces has led to increases in near-surface air temperature and PBL height, which in turn led to increases (+9.5 ppb) in surface O_3 concentrations and decreases (–16.6 $\mu g/m^3$) in $PM_{2.5}$ concentrations."

119: How exactly does your experimental setup treat the "wide heterogeneity of urban land surface processes" compared to existing studies? A large number of studies already exist using model systems (e.g. WRF) which include urban canopy models with varying complexity (SLUCM, BEP), which consider a similar level of heterogeneity than your experiments? Please discuss your statement.

While previous studies have used models with different levels of complexity, most of them failed to incorporate real-world land surface property data as input. They used default WRF settings for land surface properties such as building morphology, albedo, vegetation fraction, which either is out of date, or lacks spatial heterogeneity. By contrast, in this study we use NLCD for land cover type and impervious fraction, satellite-retrieved data for albedo, vegetation fraction and leaf area index, and GIS-based data for building morphology, which resolves spatial heterogeneity of land surface properties, and better predicts regional weather and air quality. The default version of the WRF/UCM assumes that many land cover properties are spatially homogeneous, which is not realistic.

122: unclear expression "amongst"?

We changed the sentence as follows:

"In addition, only few studies investigate interactions between land surface changes and air quality for the Southern California region (Taha, 2015; Epstein et al., 2017; Zhang et al., 2018b), which is one of the most polluted areas in the United States (American Lung Association, 2012)."

134-140: It should be made clear which new aspects you aim to analyze compared to the studies mentioned above. In my opinion simply turning urban on/off does not reveal significantly new insights. Further the term "human disturbance" is unclear, as this would also involve air quality modifications.

As we mentioned in the introduction, there are limited studies on the effect of land surface change via urbanization on regional air quality, most of which do not resolve the real-world spatial heterogeneity. In addition, there are several recent studies by our group, which incorporate satellite data for land surface characterization within Southern California, and quantifies the effect of land surface changes on regional climate including temperature and wind speed. Thus, this study combines the research idea of these two types of studies together, and aims to characterize the influence of land surface changes via historical urbanization on urban meteorology and air quality in Southern California using highly resolved land surface characterization.

We focus on the land surface modifications from human disturbance in this study, and use specific phrasing about this in the paper.

Abstract
"In this study we characterize the influence of land surface changes via historical urbanization from before human settlement to present-day on meteorology and air quality in Southern California using the Weather Research and Forecasting Model coupled to chemistry and the single-layer urban canopy model (WRF/Chem-UCM)."

Last paragraph in introduction section
"Therefore, this study aims to characterize the influence of land surface changes via historical urbanization on urban meteorology and air quality in Southern California by comparing a "Present-day" scenario with current urban land surface properties and land surface processes to a "Nonurban" scenario assuming land surface distributions prior to human perturbation."

Section 2.5

"For the Nonurban scenario, we assume natural land cover prior to human perturbation, and replace all urban grid cells with "shrubs" (Figure 1c)."

First paragraph in conclusion section

"In this study, we have characterized the impact of land surface changes via urbanization on regional meteorology and air quality in Southern California using an enhanced version of WRF/Chem-UCM. ... The two main simulations of focus in this study are the real-world "Present-day" and the hypothetical "Nonurban" scenarios; the former assumes current land cover distributions and irrigation of vegetative areas, while the latter assumes land cover distributions prior to widespread urbanization and no irrigation."

174: Please specify your lowest model level.

Thanks for the suggestion. We added this information to the manuscript.

"The average depth of the lowest model level is 53 m for all three domains."

175: "process parametrization" unclear

We changed the title to:

"Land Surface Property Characterization and Irrigation Parameterization"

180: Please discuss the term "real world representation", answering the question why the WRF default land use classification in WRF is not "real" enough for your case comparing these datasets with your input. What was the idea behind using a 30m dataset? Please briefly discuss the gain of using 30 m land cover data for a maximum resolution of 2 km. How much information "is lost" by the process of "upscaling" the LU data. Would the 2011 NLCD dataset add additional benefit?

By "real world representation", we mean instead of using default land surface properties provided with WRF, we used satellite-retrieved data specifically for the Southern California region. This is beneficial for a better prediction of regional weather.

As we mentioned in response to comment 5, we chose to use 30 m-resolution 33-category NLCD mainly for two reasons. First, urban land use varies at spatial scales on the order of 10s of meters. Second, it separates urban to three sub- urban types, which allows more detailed parameterization.

Also we chose to use the 2006 NLCD dataset in order to keep consistency with previous work from our group.

205: Did you use the additional sub-tiling option in WRF?

No, we didn't. The land surface module (the unified Noah land surface model) we use doesn't have a sub-tiling option. However, the module treats impervious fraction and pervious fraction of the urban grid cell separately.

243: Do you consider daily emission profiles? Meaning, do you find two "peaks" for instance in NOx emission/concentration?

Yes we do. Figure R2.1 shows the diurnal cycle of NOx emissions. The diurnal cycle of NOx concentrations is shown in the paper in Figure 6a. We can see that the emissions of NOx shows two peaks during daytime, and stays high between the two peaks. However, for NOx concentrations, it peaks during morning, and decreases continuously until late afternoon, despite rather high emissions. This indicates that high photolysis rates and high PBL heights due to warm temperatures in the afternoon play an important role on determining NOx concentration during daytime, apart from just emissions.

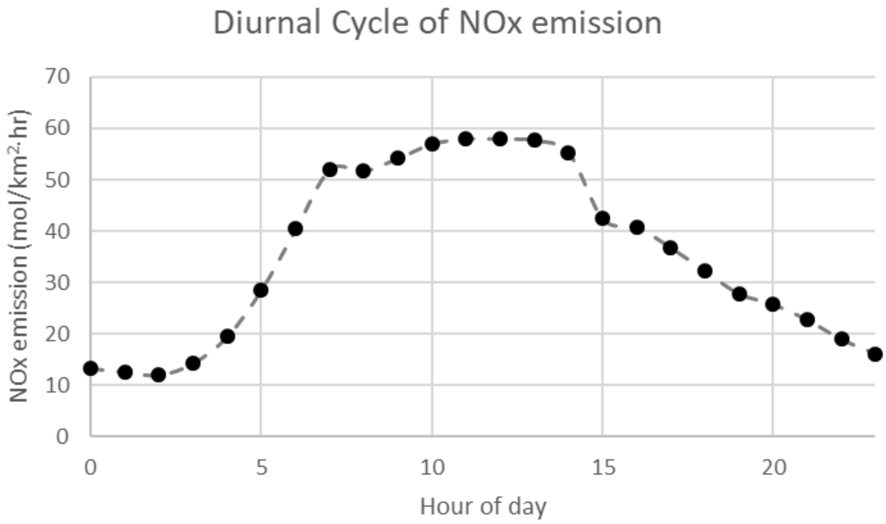

Figure R2.1 Diurnal cycle of NOx emissions. Averaged over urban grid cells within simulation period.

267: How realistic is the conversion to shrub-land for all grid cells? Would you expect different effects for a non-urban, but more heterogeneous "before human" land cover?

Please see the response to comment 7.

294: Please indicate better proof of the "good fit" mentioned here. It is not indicated by Figures S1 and 3 for $PM_{2.5}$. How does the correlation coefficient look like? What are the reasons for the poor correlation especially for the high range of the observed concentrations? How representative are the measurement stations? As the ozone concentration is highly dependent on temperature you find a good fit. Does the poor fit for $PM_{2.5}$ relate to high mixing, chemistry, both? How do correlations look like for NO2, NO, CO? Are the simulated diurnal variations realistic? Please also discuss the values from Table 1? Are they particularly good/bad?

We modified the last sentence in section 3.1 as below, and added the comparison between our evaluation results and recommended model performance benchmarks. The comparison indicates that the evaluation result are close to the ME benchmark for hourly near surface air temperature, and NMB benchmark for hourly Ozone concentrations. For daily $PM_{2.5}$ concentration, the discrepancy between the evaluation and the recommended benchmark is largely due to the underestimation of high observational values. This poor fit at high concentrations is likely occurring due to one or more of the following factors: 1) not including dust emissions in the simulation, which makes up an appreciable fraction of real-world total $PM_{2.5}$, 2) a failure of the emissions inventory to capture high emission rates on particular days, and 3) the chemistry parameterizations in WRF/Chem tending to underestimate $PM_{2.5}$ concentrations at high values, and 4) errors in simulated air pollution meteorology. We also added this information to the main paper.

'The underestimation of $PM_{2.5}$ concentrations may be occurring due to one or more of the following factors: 1) not including dust emissions in the simulation, which makes up an appreciable fraction of real-world total $PM_{2.5}$, 2) a failure of the emissions inventory to capture high emission rates on particular days, 3) the chemistry parameterizations in WRF/Chem tending to underestimate $PM_{2.5}$ concentrations at high values, and 4) errors in simulated air pollution meteorology. Table 1 shows four statistical metrics for model evaluation, including mean bias (MB) and normalized mean bias (NMB) for the quantification of bias, and mean error (ME) and root mean square error (RMSE) for the quantification of error. The statistical results indicate that model simulations underestimate near-surface air temperature, O_3 and $PM_{2.5}$ concentrations by 1.0 K, 22% and 31%, respectively. The comparison between our evaluation results and recommended model performance benchmarks is presented in the supplemental information Table S2."

The correlation coefficients for near surface air temperature, O_3 concentration and $PM_{2.5}$ concentration are 0.92, 0.82, 0.025 respectively. The observation sites for air temperature, O_3 concentration, and $PM_{2.5}$ concentration are shown is Figure S9. The sites are spread across the urban region in the model domain, and should be representative of the urban region in Southern California. On the other hand, point measurements do not capture the same spatial footprint as 2 km model grid cells. Thus, some model versus observational discrepancy is always expected, making interpretation difficult.

Figure S11 and Figure S12 shows the comparison between observed and modeled diurnal cycle for near surface air temperature and O_3 concentrations. Values for each hour are averaged over the whole simulation period for all observation sites. The results indicate that while model underestimates both observed air temperature and O_3 concentrations, it follows the diurnal pattern well. These figures are in the supplemental information, and we added a sentence in the main paper.

"…(Comparisons between observed and modeled diurnal cycles for near surface air temperatures and O3 concentrations are also presented in the supplemental information, Figure S11 and S12.)"

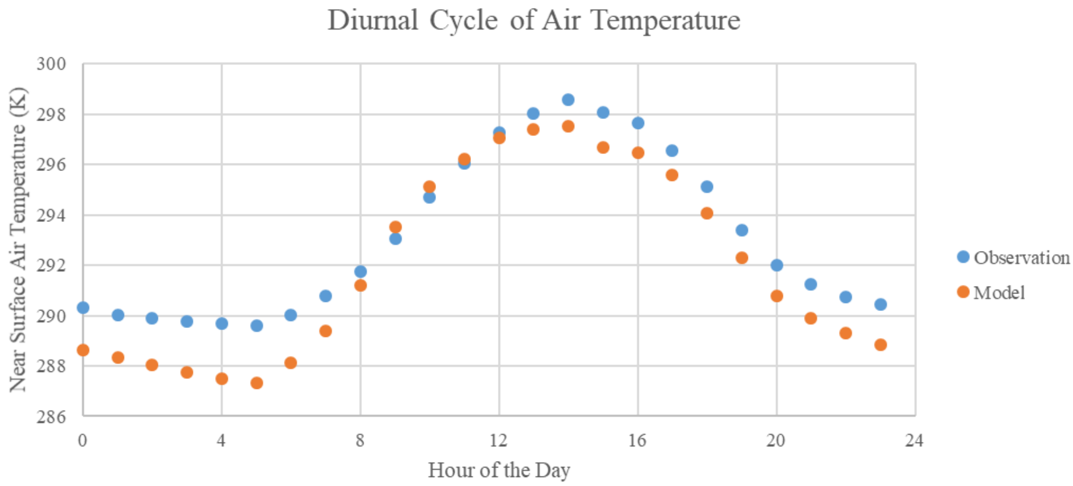

Figure S11. Diurnal cycle of observed and modeled near surface air temperature.

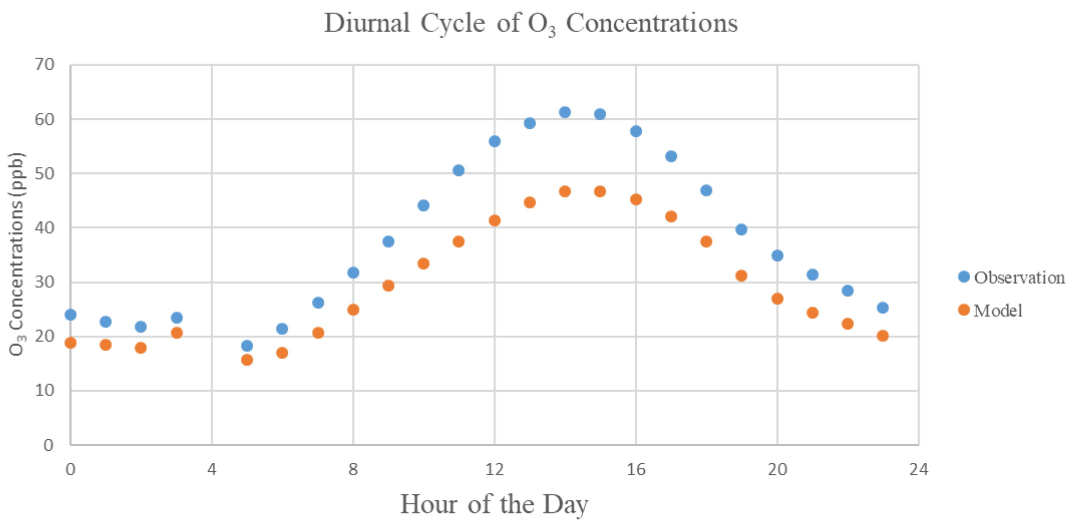

Figure S12. Diurnal cycle of observed and modeled surface O_3 concentrations (ppb).

347: Can you find impacts on the strength of the sea breeze when there is no urban area left?

The Present-day versus Nonurban difference in the strength of sea breeze is shown by Figure S14 in the supplemental information. Land surface changes via urbanization has led to decrease in wind speed throughout the day due to increase in land surface roughness. This weakening is more significant during the day, especially in the afternoon, when the baseline sea breeze is strongest.

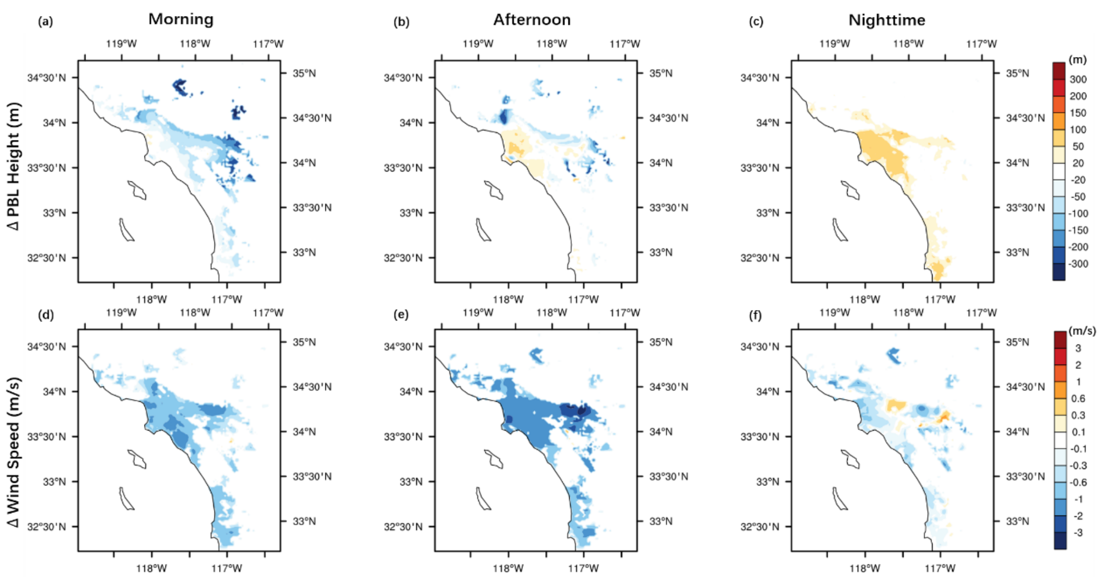

Figure S14. Spatial patterns of differences (Present-day – nonurban) in temporally averaged values during morning, afternoon and nighttime for (a,b,c) PBL heights, and (d,e,f) averaged wind speed under within PBL. Note that values are shown only for urban grid cells. Morning is defined as 7 PST to 12 PST, afternoon as 12 PST to 19 PST, and nighttime as 19 PST to 7 PST. Note that values are shown only for urban grid cells.

363: What is the order of difference between shadow model on/off?

There are two major differences in model configuration between this study and our previous publication (Vahmani and Ban-Weiss, 2016; Vahmani et al., 2016). First, in this study, we turn on the shadow model, while our previous study doesn't account for the shadow effect. Second, we use the model default calculation of surface temperature (which will affect the calculation of air temperature), while our previous study uses an alternative calculation of surface temperature. After a careful comparison among different model set-ups, we found that the parameterization of surface temperature is the more important factor that affects daytime air temperature (Figure S16 in the supplemental information). Therefore, we delete "and shading effects within urban canopies" in the sentence "In addition, increases in thermal inertia caused by use of manmade materials (e.g., pavements and buildings) and shading effects within urban canopies can contribute to simulated temperature reductions during the morning.". We also modified the last paragraph in section 3.2.3 as below.

"Note that changes in air temperature during daytime shown here disagree with Vahmani et al. (2016). While our study detects daytime temperature reductions due to urbanization, Vahmani et al. (2016) suggests daytime warming. After detailed comparison of the simulations in our study versus Vahmani et al. (2016), we find that the differences are mainly associated with UCM configuration. First, our study uses model default calculations of surface temperature for the impervious portion of urban grid cells, whereas Vahmani et al. (2016) applied an alternative calculation proposed by Li and Bou-Zeid, 2014. Li and Bou-Zeid, 2014 intended the alternate surface temperature calculations to be performed as a post-processing step rather than during runtime. After a careful

comparison among different model set-ups, we found that the parameterization of surface temperature is an important factor that affects simulated daytime air temperature (See Figure S16). Second, our study accounts for shadow effects in urban canopies, whereas Vahmani et al. (2016) assumes no shadow effects. (We note here that the default version of the UCM has the shadow model turned off. The boolean SHADOW variable in module_sf_urban.F needs to be manually switched to true to enable the shadow model calculations. With the shadow model turned off, all shortwave radiation within the urban canopy is assumed diffuse.) We suggest that it is important to include the effects of building morphology on shadows within the canopy, and to track direct and diffuse radiation separately, and therefore perform simulations in this study with the shadow model on. Note that the effect of shadows is not as significant as the parameterization of surface temperature for most of the domain in our study because the ratio between building height and road width is small."

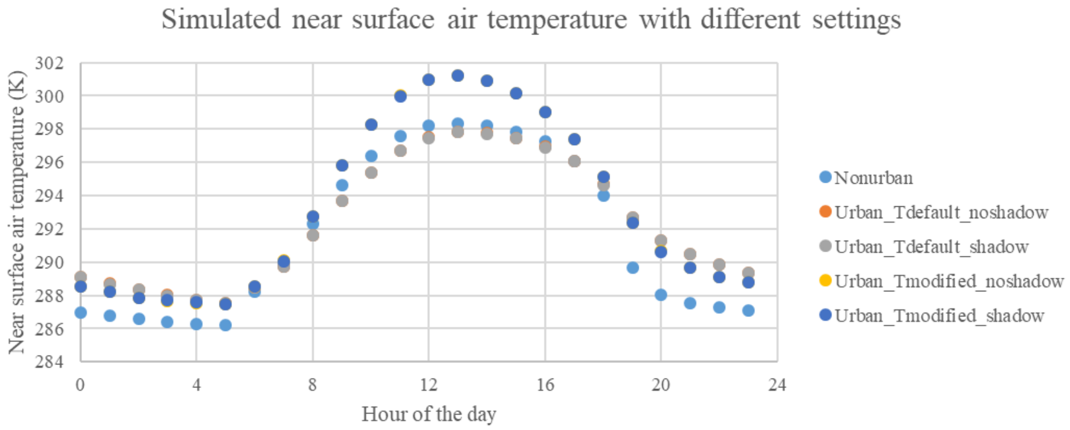

Figure S16. Diurnal cycle of near surface air temperature simulated with different model set-ups. "Tdefault" indicates that the simulation uses the default calculation of surface temperature in WRF, while "Tmodified" indicates that the simulation uses the calculation of surface temperature from Li and Bou-Zeid (2014) (which is also used in (Vahmani el al. (2016)). Dots for "Urban_Tdefault_shadow" and "Urban_Tdefault_noshadow" ("Urban_Tmodified_shadow" and "Urban_Tmodified_noshadow") are overlapping at every hour of the day because the simulation results with shadow on/off are very similar.

367: Origin of the UCM parameters?

Please refer to our response to the fourth comment.

381: Calculation of the ventilation coefficient?

We added the calculation of ventilation coefficient to the main content in section 3.2

"… The integral form of this calculation can be written as (Eq1).

$$Ventilation\ \ Coefficient = \int_0^{PBL\ height} U(z)\ dz \qquad (Eq1)$$

Given that the atmosphere is stratified in models, Eq1 can be discretized as Eq2:

$$Ventilation\ \ Coefficient = \sum_{i=1}^{m} U(z_i) \times \Delta z_i \qquad (Eq2)$$

Where $U(z_i)$ *stands for horizontal wind speed within the* i^{th} *model layer (m/s),* Δz_i *is the depth of*

i^{th} model layer that is within PBL (m), and m is the number of vertical layers up to PBL height.”

386: Please evaluate the quantity values here? Provide relative numbers.

Thanks for the suggestion. We add relative values to the sentence.

“… the spatially averaged decreases are –726 m^2/s (–23%) and –560 m^2/s (–34%), respectively.”

395: What is the relation between PBL height and surface roughness? Please provide more details. Can you find proof for this in your study?

We’ve discussed how surface roughness affects PBL height in the third paragraph in the introduction section. The nighttime PBL height is associated with variations in wind speed, which is related to variations in surface roughness. By changing shrubs (homogeneously throughout the urban region) to buildings (heterogeneously varies according to sub-urban types), the variation in surface roughness is increased. We modified the relevant sentence in section 3.2.4.

“Coastal (inland) regions show larger (smaller) variation in roughness length (Figure 2e), which leads to larger (smaller) increases in PBL heights (Figure S14c).”

490: Can you say something about the change of PBL dynamic comparing urban and non-urban. I suspect concentration of $PM_{2.5}$ is highly dependent on the boundary layer depth. Expecting lower PBLs in “urban-free” areas actually should decrease $PM_{2.5}$ in summer?

As we discussed in section 3.2.5, air temperature (surface roughness) changes are the major driver of PBL changes during the day (night) for urban grid cells. While land surface properties don’t change among nonurban grid cells (i.e., outside the urban domain), changes among urban grid cells will affect nonurban grid cells via transport of moisture, energy and momentum. Thus, most nonurban regions show similar trends for changes in PBL height compared to urban regions (discussed in the response to the next comment).

Responding to your last sentence, lower PBLs would lead to greater $PM_{2.5}$ concentrations, not lower concentrations.

530: What happens to the PBL height in non-urban environment? Even deeper?

Figure R2.2 shows changes in PBL height for Present-day – Nonurban (showing values only for grid cells that are not deemed urban in the Present-day scenario). PBL height decreases in most regions during the day (i.e., morning and afternoon), while changes at night are negligible. The tendency of changes in these grid cells outside the urban region are similar to that in urban grid cells.

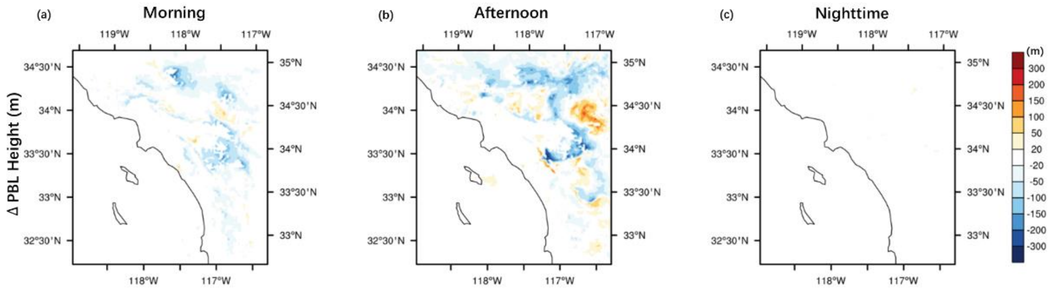

Figure R2.2 Spatial patterns of differences (Present-day – nonurban) in temporally averaged values during morning, afternoon and nighttime for (a,b,c) PBL height. Note that values are shown only for grid cells that are not deemed urban in the Present-day scenario.

535: Specify “enhanced”.

We added the following sentence to the paper to specify “enhanced”.

“We use satellite data for the characterization of land surface properties, and include a Southern California-specific irrigation parameterization.”

541: How confident are you that the land use class in the “before-human” settlement is correct? Or is it just a guess?

Please refer to the response to comment 7.

573: As mentioned earlier I am not entirely convinced, how findings from this study could be used for applied urban planning? You mention ‘mitigation and adaptation’, but a complete ‘removal’ of the urban area should be hard to transfer into an actual applicable strategies. Maybe more ‘moderate’ scenarios would be better. However, avoiding a complete re-doing of model experiments, the scope of the study should be formulated differently

Please refer to our responses to the second general comment, and the first detailed comment.

Reference

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Chen, F., Kusaka, H., Bornstein, R., Ching, J., Grimmond, C. S. B., Grossman-Clarke, S., Loridan, T., Manning, K.W., Martilli, A., Miao, S. and Sailor, D.: The integrated WRF/urban modelling system: development, evaluation, and applications to urban environmental problems, Int. J. Climatol., 31(2), 273–288, 2011.

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Vahmani, P. and Ban-Weiss, G. A.: Impact of remotely sensed albedo and vegetation fraction on simulation of urban climate in WRF-urban canopy model: A case study of the urban heat island in Los Angeles. J. Geophys. Res. Atmos., 121(4), 1511–1531, 2016.

Vahmani, P., Sun, F., Hall, A. and Ban-Weiss, G. Investigating the climate impacts of urbanization and the potential for cool roofs to counter future climate change in Southern California. Environ. Res. Lett., 11(12), 124027, 2016.

Response to Anonymous Referee #3

(Note: Reviewer comments are listed in grey, and responses to reviewer comments are in black. Pasted text from the new version of the paper is in italics.)

This manuscript investigates impacts of urbanization in Southern California on regional meteorology and air quality. Simulations using an innermost domain with 2 km resolution are conducted by WRF-Chem coupled with UCM. The simulations are driven by current climate and anthropogenic emissions with and without urban pixels and are applied to characterize impacts of historical urbanization on regional and temporal distributions of temperature and concentrations of NOx, O_3, and $PM_{2.5}$. The authors conclude that urbanization causes daytime decreases in temperature and increases in O_3 and $PM_{2.5}$. In the nighttime, the simulation results present nighttime increases in temperature and O_3, while the concentrations of NOx and $PM_{2.5}$ show reductions. The authors attribute these changes to urban-induced modifications in various competing drivers including irrigation, thermal properties of building materials and surface roughness.

General comments:

The topic addressed is interesting and relevant to ACP readers. However, I have reservations about the robustness of the conclusions presented. In my opinion, significant revisions with new analysis and more careful model verification of the simulations are required.

We thank the review for his/her helpful comments. We believe that addressing these comments have vastly improved the quality of our paper.

The impact of urbanization is derived from the differences between temperature and concentrations of fields simulated by a WRF-Chem configuration that includes urban pixels and by a scenario where urban pixels were converted to shrub. This methodology has been presented in previous work and the nighttime impact of urbanization has been well documented in the literature. For instance, the paper by Li et al ("Achieving accurate simulations of urban impacts on ozone at high resolution", ERL, 9, 2014) introduced similar configurations (WRF-Chem including anthropogenic emissions, with and without urbanization) and used them to derive impacts of urbanization on air quality by analyzing the differences in the simulated fields between the two scenarios. Although the region and the period of time considered in this manuscript are different, the main idea and the nighttime impact are similar. The daytime impact reported in this manuscript is questionable because its magnitude shows values smaller than the model error (see specific comment 2). Careful analysis of the robustness of the impact is needed, especially given that this impact conflicts with previous results as reported (Line 355). The authors need to emphasize what is new related to this research and how it advances the existing research on the topic.

We thank the reviewer for these comments. The first major idea presented here (robustness of the daytime impact) is also brought up in specific comments 2 and 5. Please see our responses to those comments.

The second major idea is on the novelty of this study. While this study shows some similarity in research idea with previous literature, it extend this research topic in that 1) it includes discussion on the impact of land surface changes on total and speciated $PM_{2.5}$ concentration, which has been seldom studied, 2) it focuses on the Southern California region where such research is limited but necessary given the high pollutant loads, and 3) it incorporates accurately resolved land surface data. We added a few sentences in the last paragraph of introduction section to clarify these points.

"... Note that this paper builds on our prior study Vahmani et al. (2016), but focuses on air quality impacts, whereas our previous research was on meteorological impacts only. While the influence of land surface changes on regional weather has been investigated in numerous past studies, its influence on regional air quality has been seldom studied in past work."

Specific comments:

1. It is unclear why the authors chose a 10-day period of the summer of 2012? And in what basis the period chosen is "representative of typical summer days in Southern California"? Why not using more years?

We chose this 10-day period because the observed meteorology field is representative of typical summer days in Southern California, which are clear (no clouds) and without precipitation. We added a figure in the supplemental information (Figure S8) showing the diurnal cycle of averaged (observed) near surface air temperature over JJA (June, July and August) and over our simulation period. We also added a sentence in the main paper pointing to that figure.

"This simulation period is chosen as representative of typical summer days in Southern California, which are generally clear or mostly sunny without precipitation. A comparison of observed diurnal cycles for average near surface air temperatures over JJA (June, July and August) versus over our simulation period is shown in Figure S8 in the supplemental information."

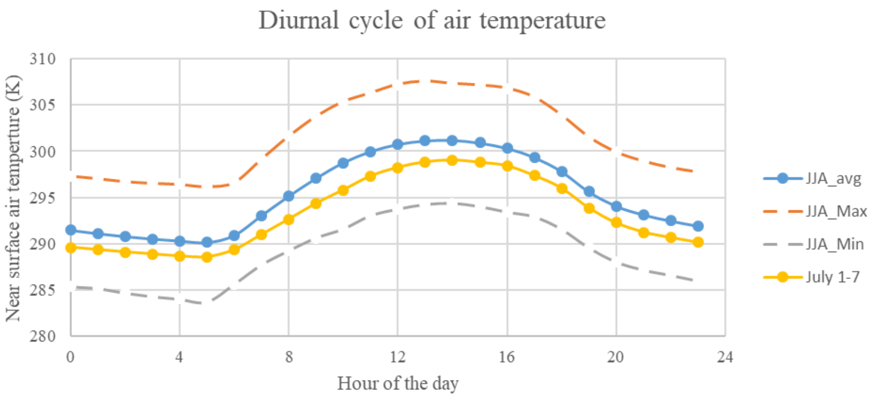

Figure S8. Diurnal cycles for observed near surface air temperature (K) over JJA (June, July and August) in blue, and over our simulation period in yellow. Observations are obtained from MesoWest (https://mesowest.utah.edu/), which are available at Mesonet API (https://developers.synopticdata.com/mesonet/). Mean values are derived by averaging over all observational sites available for the innermost domain and the aforementioned period for each hour of day. Orange and grey curves show the maximum and minimum air temperature at each hour of the day for JJA. Results show that our simulation period (July 1-7) is representative of summertime meteorology for our domain.

We do the simulations for year 2012 because it is the most recent year for which an accurate emissions inventory is available for Southern California.

2. The statistics presented in Table 1 indicate that the impact of urbanization is smaller than the model error for all the fields analyzed. For example, the magnitude of the simulated change in O_3 is less than 5.6 ppb (Line 429). The mean and root mean square errors reported in Table 1 are 11.8 ppb and 14.6 ppb, respectively. Thus, the impact described, which is the main conclusion of the manuscript, is not robust given that it lies within the model error. Perhaps, simulations using other years could increase the statistical significance of the results presented.

We suggest that the comparison of urbanization impacts versus model error in Table 1 is not the right comparison. Instead, to assess whether urbanization impacts are statistically distinguishable from zero, we added a new statistical analysis to the paper, using the paired Student's t-test with n = 7 days. We did the test to check 1) whether spatially averaged changes in regional meteorology and air quality are significant within the simulation period, and 2) whether changes in spatial resolved regional meteorology and air quality are significant within the simulation period (i.e., for each urban grid cell). For 1), we edited the relevant sentences in the paper. For 2), we updated all figures with maps in the paper to mark out the insignificant grid cells with black dots, and edited the relevant description of the spatial patterns. Please see section 2.5 and section 3 for those changes. We haven't pasted the changes here because they are distributed throughout our results section, and would take up over 3 pages.

3. The authors state in the conclusion that ". . .due to historical urbanization are the main drivers of regional meteorology and air quality changes in Southern California" (Line 567). However, the simulations presented in the manuscript cannot be applied to reach such conclusion. There are several critical factors that are not accounted for. For example, the initial and boundary conditions use current atmospheric conditions and therefore do not include the effect of climate change. The amount of the background CO_2 concentration specified in WRF is fixed (assuming that both configurations use the same setup except for urbanization as stated). The anthropogenic emissions did not exist before human settlement. I suggest that the authors rephrase their motivation and conclusion, and simply focus on the impact of urbanization without attributing historical changes solely to urbanization.

We agree with the reviewer that the motivation and conclusions were not sufficiently clear in the original paper. Thus, we modified the last paragraph of the introduction section and conclusion section.

"… In this paper, we aim to quantify the importance of historical land cover change on air pollutant concentrations, and thus the "Nonurban" scenario assumes current anthropogenic pollutant emissions. This hypothetical scenario cannot exist in reality, since current anthropogenic emissions would not exist without the city, but our intent is to tease out the relative importance of land cover change through urbanization (assuming constant emissions) on air pollutant concentrations."

"This study highlights the role that land cover properties can have on regional meteorology and air quality. We find that increases in evapotranspiration, thermal inertia, and surface roughness due to

historical urbanization are the main drivers of regional meteorology and air quality changes in Southern California. …Our findings indicate that air pollutant concentrations have been impacted by land cover changes since pre-settlement times (i.e., urbanization), even assuming constant anthropogenic emissions. These air pollutant changes are driven by urbanization-induced changes in meteorology. This suggests that policies that impact land surface properties (e.g., urban heat mitigations strategies) can have impacts on air pollutant concentrations (in addition to meteorological impacts); to the extent possible, all environmental systems should be taken into account when studying the benefits or potential penalties of policies that impact the land surface in cities."

4. There are some claims that need clarification. For example the authors state in line 152 "In this study, we couple WRF/Chem to the urban canopy model (UCM). . ." However, the WRF/Chem model is already coupled to UCM. I believe what the authors did is activating the option for this coupling. In line 180 "we update the default WRF/Chem to include a real-world representation of land surface physical properties and processes. . ." But again, the options for using NLCD and NUDAPT for land surface representations are available within WRF. Please clarify what is meant by "we update the default WRF/Chem".

Thanks to the reviewer for pointing this out. We modified the sentence "In this study, we couple WRF/Chem to the urban canopy model (UCM). . ." as below.

"In this study, we activate the urban canopy model (UCM) in WRF/Chem that …"

By "update the default WRF/Chem" we mean that we've used GIS-based building morphologies, satellite-retrieved land surface data, and a Southern California specific irrigation module for the simulations, which make the model simulation more representative of current day weather conditions and air quality in Southern California. We also modified gaseous dry deposition in chemistry module based on previous literature so that WRF/Chem can be compatible with 33-category land use types.

5. The ability of WRF-Chem to realistically represent urban processes requires more evaluation to better establish the credibility of the present-day scenario. The comparison between observations and simulations shown in Fig. 3 does not indicate to me a "good fit at lower values" as stated in line 294. The observed low values of temperature are around 290 K, but the simulated temperature shows low values of 287K. The difference between these values is larger that the impact reported. Therefore, better model verification should be considered. I also suggest adding to Fig. 3 panels comparing diurnal variations of observed and simulated temperature, O_3 and $PM_{2.5}$ (similar to Fig. 4a).

For the significance of the reported urbanization impact, please refer to our response to comment 2.

Figures S11 and S12 show the comparison between observed and modeled diurnal variations for near surface air temperature and O_3 concentrations. (For $PM_{2.5}$ concentrations we use only daily values instead of hourly $PM_{2.5}$ concentrations for reasons that are explained in the supplemental information section S1; thus, the diurnal variation of observed and simulated $PM_{2.5}$ is not discussed here.) In Figures S11 and S12, values for each hour are averaged over the whole simulation period for all observation sites. The results indicate that while the model underestimates both observed air temperature and O_3 concentrations, the shape of the diurnal cycle is well modeled. For air temperature, simulation results tend to capture daytime (relatively higher) values better than nighttime (relatively lower) values. For O_3 concentrations, the model predicts lower concentrations better than higher concentrations. Thus, we edited the sentence the reviewer mentioned. And we put these two figures to the supplemental information, and added a sentence in the main paper.

"Figure 3 shows the comparison between observed and modeled hourly near surface air temperature, O_3 concentrations, and daily $PM_{2.5}$ concentrations. (Comparisons between observed and modeled diurnal cycles for near surface air temperatures and O_3 concentrations are also presented in the supplemental information, Figure S11 and S12.) As shown in Figure 3 (and Figure S11), the model simulations better capture higher air temperatures during the daytime relative to lower values during nighttime. By contrast, predictions of O_3 and $PM_{2.5}$ concentrations show good fit with observations at low values that occur with high occurrence frequency. However, observed O_3 and $PM_{2.5}$ concentrations are underestimated by the model at higher values that occur with lower frequency of occurrence."

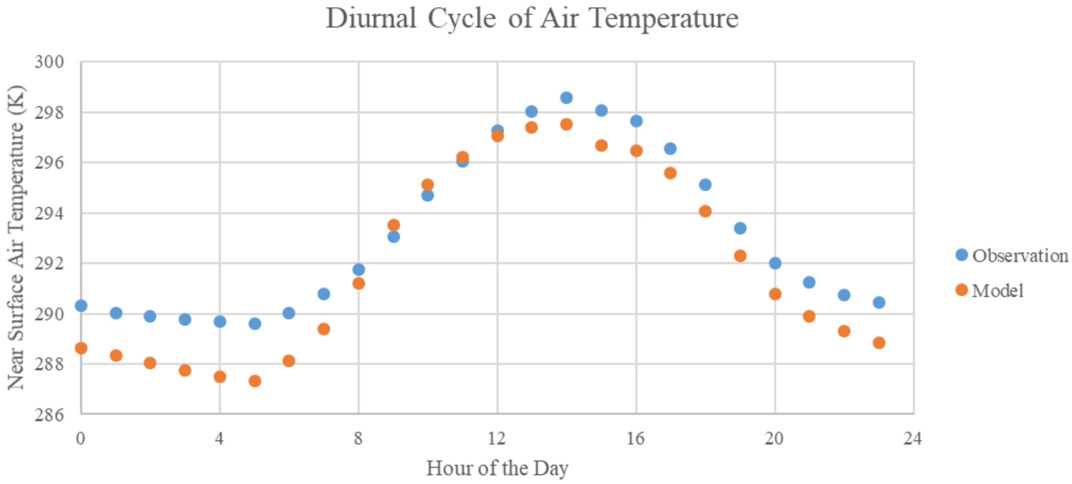

Figure S11 Diurnal cycle of observed and modeled near surface air temperature.

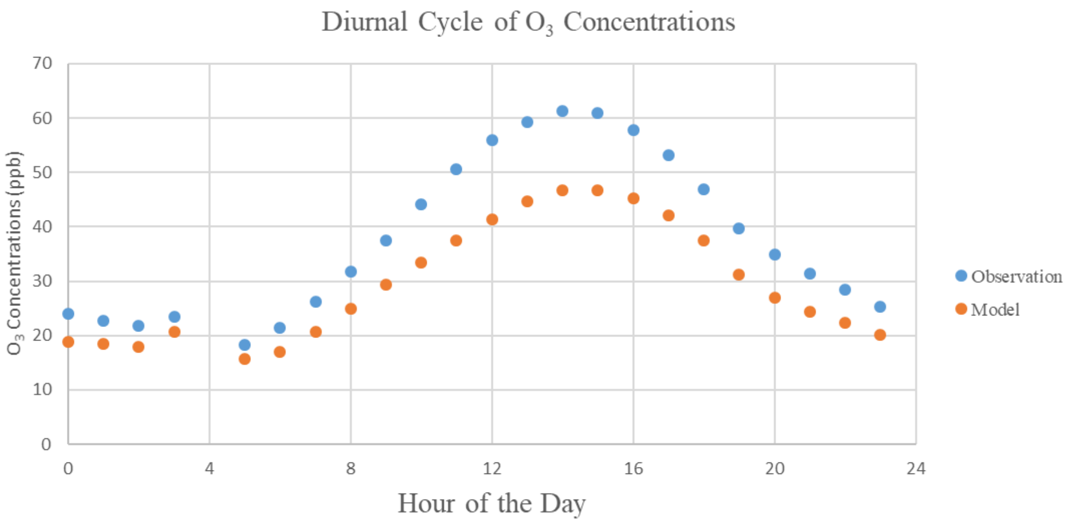

Figure S12 Diurnal cycle of observed and modeled surface O_3 concentrations (ppm).

6. Figs 5, 7 and 9 include values of simulated fields within urban grid cells only. The authors should consider superimposing in these figures values for the entire domain including nonurban grid cells. It would be very helpful to see the differences in the simulated fields within both urbanized pixels and also grid cells that remain natural in both scenarios considered.

This is a good idea. We added new versions of each figure to the supplemental information that include values for non-urban cells (Figures S13, S17 and S18). Please check the supplemental information for these three new figures. In general, the changes in non-urban grid cells are not significantly different from zero at 95% confidence interval for most places. We also added several sentences to the main paper which point to those figures.

Last sentence in section 3.2.2

"A modified version of Figure 5 that includes values for non-urban cells is in the supplemental information Figure S13."

Last sentence in section 3.3.1

"A modified version of Figure 7 that includes values for non-urban cells is in the supplemental information Figure S17."

Last sentence in section 3.4.2

"A modified version of Figure 9 that includes values for non-urban cells is in the supplemental information Figure S18."

Effects of Urbanization on Regional Meteorology and Air Quality in Southern California

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Abstract

Urbanization has a profound influence on regional meteorology and air quality in megapolitan Southern California. The influence of urbanization on meteorology is driven by changes in land surface physical properties and land surface processes. These changes in meteorology in turn influence air quality by changing temperature-dependent chemical reactions and emissions, gas-particle phase partitioning, and ventilation of pollutants. In this study we characterize the influence of land surface changes via historical urbanization from before human settlement to present-day on meteorology and air quality in Southern California using the Weather Research and Forecasting Model coupled to chemistry and the single-layer urban canopy model (WRF/Chem-UCM). We assume identical anthropogenic emissions for the simulations carried out, and thus focus on the effect of changes in land surface physical properties and land surface processes on air quality. Historical urbanization has led to daytime air temperature decreases of up to 1.4 K, and evening temperature increases of up to 1.7 K. Ventilation of air in the LA basin has decreased up to 36.6% during daytime and increased up to 27.0% during nighttime. These changes in meteorology are mainly attributable to higher evaporative fluxes and thermal inertia of soil from irrigation, ~~higher thermal inertia from irrigation and building materials,~~ and increased surface roughness and thermal inertia from buildings. Changes in ventilation drive changes in

hourly NOx concentrations with increases of up to 2.7 ppb during daytime and decreases of up to 4.7 ppb at night. Hourly O_3 concentrations decrease by up to 0.94 ppb in the morning, and increase by up to 5.6 ppb at other times of day. Changes in O_3 concentrations are driven by the competing effects of changes in ventilation and precursor NOx concentrations. $PM_{2.5}$ concentrations show slight increases during the day, and decreases of up to 2.5 $\mu g/m^3$ at night. Processes drivers for changes in $PM_{2.5}$ include modifications to atmospheric ventilation, and temperature, which impacts gas-particle phase partitioning for semi-volatile compounds and chemical reactions. Understanding processes drivers ~~for~~ related to how land surface changes effect regional meteorology and air quality is crucial for decision making on urban planning in megapolitan Southern California to achieve regional climate adaptation and air quality improvements.

1. Introduction

The world has been undergoing accelerated urbanization since the industrial revolution in the 19th Century (Grimm et al., 2008; Seto et al., 2012). Urbanization leads to profound human modification of the land surface and its associated physical properties such as roughness, thermal inertia, and albedo (Fan et al., 2017), and land surface processes like irrigation (Vahmani and Hogue, 2014). These changes in land surface physical properties and processes alter corresponding surface-atmosphere coupling including exchange of water, momentum and energy in urbanized regions (Vahmani and Ban-Weiss, 2016a; Li et al., 2017), which exerts an important influence on regional meteorology and air quality (Vahmani et al., 2016; Civerolo et al., 2007).

Land surface modifications from urbanization drive changes in urban meteorological variables such as temperature, wind speed and planetary boundary layer (PBL) height, which result in urban – rural differences. Differences in surface temperature and near surface air temperature have been widely studied for decades. The urban heat island (UHI) effect, a phenomenon in which temperatures within an urban area are higher than surrounding rural areas (Oke, 1982), has been extensively studied using models and observations for a great number of urban regions (Rizwan et al., 2008; Peng et al., 2012; Stewart and Oke, 2012). A contrary phenomenon, namely the urban cool island (UCI), under which urban temperatures are lower than surrounding rural temperatures, has also been investigated recently in some studies (Carnahan and Larson, 1990; Theeuwes et al., 2015; Kumar et al., 2017). Urban – rural contrast in temperature (i.e. both UHI and UCI) is mainly attributable to differences in thermal properties and energy fluxes due to heterogeneous land surface properties. For urban areas, buildings and roads (i.e., impervious surfaces) are generally made from manufactured materials (e.g., asphalt concrete) with low albedo and thus high solar absorptivity (Wang et al., 2017). These materials also have high thermal inertia, which can lead to reductions in diurnal temperature range due to heat storage and consequent temperature reductions during the day and heat release and consequent temperature increases at night (Hardin and Vanos, 2018). Street canyons, which we refer to as the U-shaped region between buildings, can trap longwave energy fluxes within the canyon because of reductions of sky-view factors (Qiao et al., 2013). On the other hand, shading in street canyons during the day can reduce absorption of shortwave radiation (Carnahan and Larson, 1990; Kusaka et al., 2001). Pervious surfaces within urban areas such as irrigated urban parks and lawns can lead to the urban oasis effect in which evaporative cooling occurs due to increases in evapotranspiration. In addition, soil thermal properties depend on their water content, which ultimately affects ground heat fluxes and thus surface and air temperatures. Land surface properties in surrounding rural areas can also affect urban – rural differences in temperature (Imhoff et al., 2010; Peng et al., 2012; Zhao et al., 2014). In particular, u~~U~~rban regions built in semi-arid or arid surroundings tend to have a weak daytime UHI or even a UCI, whereas those built in moist regions tend to have a larger daytime UHI (Fan et al., 2017; Peng et al., 2012). Lastly, factors such as anthropogenic heat and atmospheric aerosol burdens can play an important role in urban heat/cool island formation in some regions (Oke, 1982; Wang et al., 2017).

Urbanization can also cause differences between urban and rural areas for meteorological variables other than surface and air temperatures. Changes in regional near-surface wind speed and direction can occur in urban areas because of spatially varying modifications in surface roughness (Xu et al., 2006; Vahmani et al., 2016). Changes in near surface winds in coastal urban areas can also be affected by modifying land-sea temperature contrast (Vahmani et al., 2016). The ~~formation~~ characteristics of the PBL ~~is~~ are dependent on the magnitude of turbulent kinetic energy (TKE) (Garratt, 1994). Higher (lower) TKE will lead to deeper (shallower) PBLs. During daytime, the magnitude of TKE is driven by buoyancy production contributed mainly by sensible heat flux (with clear skies); at night, TKE is driven by shear production associated with variance in wind speed. Thus, temperature and surface roughness play an important role on the depth of the PBL during daytime and nighttime, respectively. Lastly, changes in relative humidity, precipitation, and other meteorological variables due to land surface changes can also be significant in some regions (Burian and Shepherd, 2005; Georgescu et al., 2014).

Changes in meteorological conditions due~~from~~ to urbanization can influence concentrations of air pollutants including oxides of nitrogen (NOx), ozone (O_3) and fine particulate matter ($PM_{2.5}$). NOx and O_3 pollution are major public health concerns in megapolitan regions (Lippmann, 1989). $PM_{2.5}$ reduces visibility, ~~exerts~~ causes adverse health effects, and alters regional and global climate via direct and indirect effects (Charlson et al., 1992; Pope and Dockery, 2006; Boucher et al., 2013). Meteorology can affect emission rates, chemical reaction rates, gas-particle phase partitioning of semi-volatile species, pollutant dispersion, and deposition; thus, it plays an important role in determining air pollutant concentrations.~~Changes in air pollutant concentrations due to meteorological changes occur via altered emissions, weather-dependent chemical reactions, gas-particle phase partitioning of semi-volatile species, pollutant dispersion, and deposition.~~ Variations in air temperatures together with vegetation types affect the production of biogenic volatile organic compounds (BVOCs), which are important precursors for ground-level O_3 and secondary organic aerosols (SOA) (Guenther et al., 2006). Gas-phase chemical reactions that form secondary pollutants are also temperature-dependent. Higher (lower) air temperatures in general lead to higher (lower) photolysis reaction rates and atmospheric oxidation rates, which enhance the production of tropospheric O_3, secondary inorganic aerosols (e.g. nitrate, sulfate, and ammonium aerosols) and SOA (Aw and Kleeman, 2003; Hassan et al., 2013). In addition, concentrations of semi-volatile compounds are affected by equilibrium vapor pressure under various temperature conditions (Pankow, 1997; Ackermann et al., 1998). Higher (lower) temperatures favor phase-partitioning to the gas (particle) phase. Ventilation, which is the combined effect of vertical mixing and horizontal dispersion, can also influence pollutant concentrations (Epstein et al., 2017). Higher (lower) ventilation rates lead to lower (higher) pollutant concentrations especially in coastal cities like Los Angeles where upwind air under typical meteorological conditions is clean relative to urban air. Lastly, changes in surface roughness may affect loss of pollutants via surface deposition, which in turn alters air pollutant concentrations (Abdul-Wahab et al., 2005).

A number of previous studies have investigated the impacts of land surface changes on regional meteorology in a variety of urban regions around the world (Kalnay and Cai, 2003; Burian and Shepherd, 2005; Zhang et al., 2010). However, limited studies have quantified the impact of land surface changes on regional air quality, and most of these studies have focused on changes in surface O_3 concentrations. Civerolo et al. (2007) estimated that land-use changes via urban expansion in New York City can cause increases in near-surface air temperature of 0.6 ℃ as well as increases in episode-maximum 8h O_3 concentrations of 6 ppb. Jiang et al. (2008) focused on the Houston, Texas area, and found similar relationships between urban expansion, near-surface air temperatures, and O_3. Nevertheless, only a few studies have included changes in $PM_{2.5}$ concentrations. Tao et al. (2015)

simulated that spatially averaged surface O_3 concentrations slightly increased (+0.1 ppb) in eastern China due to urbanization, whereas $PM_{2.5}$ concentrations decreased by –5.4 μg/m^3 at the near surface. Chen et al. (2018) studied urbanization in Beijing, and found that modification of rural to urban land surfaces has led to increases in near-surface air temperature and PBL height, which in turn led to increases (+9.5 ppb) in surface O_3 concentrations and decreases (–16.6 μg/m^3) in $PM_{2.5}$ concentrations. However, past studies that investigate interactions between land surface changes and changes in meteorology and air quality generally do not identify the major processes driving these interactions. They also do not resolve the wide heterogeneity of urban land surface properties, with most studies assuming that urban properties are homogenous throughout the city. In addition, only few studies investigate interactions between land surface changes and air quality for the Southern California region (Taha, 2015; Epstein et al., 2017; Zhang et al., 2018b), which ~~has among the worst air quality in~~is one of the most polluted areas in the United States (American Lung Association, 2012).

With advances in real-world land surface datasets from satellites, recent modeling studies on land-atmosphere interactions are able to resolve heterogeneous land surface properties and thus better capture urban meteorology, enabling modeling studies that more accurately quantify changes in regional meteorology due to land surface modification. By combining satellite-retrieved high-resolution land surface data with the Weather Research and Forecasting Model coupled to the Single-layer Urban Canopy Model (WRF/UCM), simulations reported in Vahmani and Ban-Weiss (2016a) show improved model performance (i.e. compared to observations) for meteorology in Southern California compared to the default model, which assumes that urban regions have homogeneous urban land cover. A follow-up study, Vahmani et al. (2016), suggested that historical urbanization has altered regional meteorology (e.g., near surface air temperatures and wind flows) in Southern California mainly because of urban irrigation, and changes in land surface thermal properties and roughness. While historical urbanization and its associated impacts on meteorology has the potential to cause important changes in air pollutant concentrations in Southern California, this is never been investigated in past work.

Therefore, this study aims to characterize the influence of land surface changes via historical urbanization on urban meteorology and air quality in Southern California by comparing a "Present-day" scenario with current urban land surface properties and land surface processes to a "Nonurban" scenario assuming land surface distributions prior to human perturbation. To achieve this goal, we adopt a state-of-the-science regional climate-air quality model, the Weather Research and Forecasting Model coupled to chemistry and the Single-layer Urban Canopy Model (WRF/Chem-UCM), and incorporate high-resolution heterogeneity in urban surface properties and processes to predict regional weather and pollutant concentrations. We assess the response of regional meteorology and air quality to individual changes in land surface properties and processes to determine driving factors on atmospheric changes. Note that this paper builds on our prior study Vahmani et al. (2016), but focuses on air quality impacts, whereas our previous research was on meteorological impacts only. While the influence of land surface changes on regional weather has been investigated in numerous past studies, its influence on regional air quality has been seldom studied in past work. In this paper, we aim to quantify the importance of historical land cover change on air pollutant concentrations, and thus the "Nonurban" scenario assumes current anthropogenic pollutant emissions. This hypothetical scenario cannot exist in reality, since current anthropogenic emissions would not exist without the~~a~~ city, but our intent is to tease out the relative importance of land cover change through urbanization (assuming constant emissions) on air pollutant concentrations.

2. Methodology and Data

2.1 Model Description and Configuration

WRF/Chem v3.7 is used in this study to simulate meteorological fields and atmospheric chemistry. WRF/Chem is a state-of-the-science nonhydrostatic mesoscale numerical meteorological model that facilitates "online" simulation of processes relevant to atmospheric chemistry including pollutant emissions, gas and particle phase chemistry, transport and mixing, and deposition (Grell et al., 2005). In this study, we ~~couple~~ activate ~~WRF/Chem to~~ the urban canopy model (UCM) in WRF/Chem that resolves land-atmosphere exchange of water, momentum, and energy for impervious surfaces in urban areas (Kusaka et al., 2001; Chen et al., 2011; Yang et al., 2015). The UCM parameterizes the effects of urban geometry on energy fluxes from urban facets (i.e., roofs, walls, and roads) and wind profiles within canyons (Kusaka et al., 2012). We account for the effect of anthropogenic heat on urban climate by adopting the default diurnal profile in the UCM. Physics schemes included in our model configuration are the Lin cloud microphysics scheme (Lin et al., 1983), the RRTM longwave radiation scheme (Mlawer et al., 1997), the Goddard shortwave radiation scheme (Chou and Suarez, 1999), the YSU boundary layer scheme (Hong et al., 2006), the MM5 similarity surface layer scheme (Dyer and Hicks, 1970; Paulson, 1970), the Grell 3D ensemble cumulus cloud scheme (Grell & Dévényi, 2002), and the unified Noah land surface model (Chen et al, 2001). Chemistry schemes include the TUV photolysis scheme (Madronich, 1987), RACM-ESRL gas phase chemistry (Kim et al., 2009; Stockwell et al., 1997), and MADE/VBS aerosols scheme (Ackermann et al., 1998; Ahmadov et al., 2012).

All model simulations are carried out from June 28th, 2100 UTC (June 28th, 1300 PST) to July 8th, 0700 UTC (July 7th, 2300 PST), 2012 using the North American Regional Reanalysis (NARR) dataset as initial and boundary meteorological conditions (Mesinger et al., 2006). This simulation period is chosen as representative of typical summer days in Southern California, which are generally clear or mostly sunny without precipitation. A comparison of observed diurnal cycles for average near surface air temperatures over JJA (June, July and August) versus over our simulation period is shown in Figure S8 in the supplemental information. Hourly model output from July 1st, 0000 PST to 7th, 2300 PST is used for analysis, and simulation results prior to July 1st, 0000 PST are discarded as spin up. Figure 1a shows the three two-way nested domains with horizontal resolutions of 18 km, 6 km and 2 km, respectively, centered at 33.9°N, 118.14°W. Only the innermost domain (141 ×129 grid cells), which encapsulates the Los Angeles and San Diego metropolitan regions, is used for analysis. All three domains consist of 29 unequally spaced layers in the vertical from the ground to 100 hPa. The average depth of the lowest model level is 53 m for all three domains.

2.2 Land Surface Property Characterization and Irrigation~~Process~~ Parameterization

One important aspect of accurately simulating meteorology and air quality is to properly characterize land surface – atmosphere interactions (Vahmani and Ban-Weiss, 2016a; Li et al., 2017). In addition, accurately quantifying the climate and air quality impacts of historical urbanization requires a realistic portrayal of current land cover in the urban area (Vahmani et al., 2016). For both of these reasons, we update the default WRF/Chem to include a real-world representation of land surface physical properties and processes.

In this study, we use the (30 m resolution) 33-category National Land Cover Database (NLCD) for the year 2006 for all three model domains. NLCD differentiates three urban types including low-intensity residential, high-intensity residential, and industrial/commercial (shown in Figure 1b) (Fry et al., 2011). In the model (UCM), each of these three types can have unique urban physical properties such as building morphology, albedo, and thermal properties for each facet. We adopt the grid-cell specific National Urban Database and Access Portal Tool (NUDAPT) where available in the innermost domain for building morphology including average building heights, road widths, and roof widths

(Ching et al., 2009). Where NUDAPT data are unavailable, we use average building and road morphology for three urban categories from the Los Angeles Region Imagery Acquisition Consortium (LARIAC). Details on the generation of averaged urban morphology parameters from real-world GIS datasets can be found in Zhang et al. (2018a). For the other parameters in the UCM (e.g., anthropogenic latent heat, surface emissivity), we use default WRF settings documented in file URBPARM.TBL. Note that the original gaseous dry deposition code based on Wesely (1989) is only compatible with the default 24-category U.S. Geological Survey (USGS) global land cover map. We therefore modify the code according to Fallmann et al. (2016), which assumes that the three urban types in the 33-category system have input resistances that are the same as the urban type for the 24-category system. In addition, impervious fractions (i.e. the fraction of each cell covered by impervious surfaces) for each of the three urban categories in the innermost domain are from the NLCD impervious surface data (Wickham et al., 2013).

Land surface properties including albedo, green vegetation fraction (GVF), and leaf area index (LAI) are important for accurately predicting absorption and reflection of solar radiation and evaporative fluxes in urban areas (Vahmani and Ban-Weiss, 2016a). To resolve high-resolution real-world heterogeneity in these land surface properties, the simulations performed in this study use satellite-retrieved real-time albedo, GVF, and LAI for the innermost domain. Input data compatible with WRF are regridded horizontally using albedo, GVF, and LAI maps generated based on MODIS reflectance (MCD43A4), vegetation indices (MOD13A3), and fraction of photosynthetically active radiation (MCD15A3) products, respectively. Raw data are available from the USGS National Center for Earth Resource Observations and Science website at http://earthexplorer.usgs.gov. A detailed description on the implementation of MODIS-retrieved land surface properties for WRF can be found in Vahmani and Ban-Weiss (2016a). Our previous research has shown that the model enhancements described here reduce model biases in surface and near-surface air temperatures (relative to ground and satellite observations) for urban regions in southern California. In particular, the root-mean-square-error for nighttime near-surface air temperature has been narrowed from 3.8 to 1.9 °C.

Resolving urban irrigation is also of great significance for accurately predicting latent heat fluxes and temperatures within Los Angeles. Here we use an irrigation module developed by Vahmani and Hogue (2014), which assumes irrigation occurs three times a week at 2100 PST on the pervious fraction of urban grid cells. This model was tuned to match observations of evapotranspiration in the Los Angeles area. Details on the implementation of this irrigation module and its evaluation with observations can be found in Vahmani and Hogue (2014). Note that we do not use the default irrigation module available in the single layer canopy model in WRF/UCM v3.7, which assumes daily irrigation at 2100 PST in summertime, because (1) the irrigation module of Vahmani and Hogue (2014) was already evaluated and tuned for Southern California, and (2) we strive to maintain consistency with our previous related studies.

2.3 Emission Inventories

Producing accurate air quality predictions also relies on using emission inventories that capture real-world emissions. We adopt year 2012 anthropogenic emissions from the California Air Resource Board (CARB) for the two outer domains (CARB, 2017) where data are available (i.e. within California), and from South Coast Air Quality Management District (SCAQMD) for the innermost domain (SCAQMD, 2017). For areas within the two outer domains that are outside California, we use the U.S. Environmental Protection Agency (EPA) National Emissions Inventory (NEI) for 2011 that is available with the standard WRF/Chem model (U.S. EPA, 2014). CARB and SCAQMD emission inventories as provided have 4 km spatial resolution, with 18 and 11 layers in the vertical from the ground to 100 hPa, respectively. We regridded these inventories in the horizontal and vertical to match the grids of our modeling domains. Note that the aforementioned emission inventories use chemical speciation from the SAPRC chemical mechanism (Carter, 2003), and thus we have converted species to align with the RACM-ESRL and MADE/VBS mechanisms, both of which use RADM2 (Regional Acid Deposition Model) speciation (Stockwell et al., 1990). The conversion uses species and weighting factors from the emiss_v04.F script that is distributed with NEI emissions for WRF/Chem modeling. (The original script is available at: ftp://aftp.fsl.noaa.gov/divisions/taq.) More details on re-speciating the emissions datasets are presented in the supplemental information (Table S1). For online calculation of biogenic volatile organic emissions we adopt the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006). The default LAI in MEGAN is substituted with the satellite-retrieved LAI for better quantification of biogenic emissions. Note that we have turned on online calculation of sea salt emissions, but turned off that of dust emissions (both available with default WRF).

2.4 Meteorology and Air Pollutant Observations

To facilitate model evaluation, we obtain hourly near-surface air temperature observations, hourly ground-level O_3 and daily $PM_{2.5}$ observations within our simulation period. Near-surface air temperature data are gathered from 12 stations from the California Irrigation Management Information System (CIMIS). Air pollutant observations are from the Air Quality System (AQS), which is maintained by the U.S. EPA. Ozone ($PM_{2.5}$) data from 33 (27) air quality monitoring stations are collected representing Los Angeles, Orange, Riverside and San Bernardino Counties. The locations of monitoring stations are shown in Figure S95. Among the 27 monitoring stations where $PM_{2.5}$ observations are available, daily $PM_{2.5}$ concentrations from gravimetric analysis can be directly obtained from 20 stations, while hourly observations acquired using a Beta Attenuation Monitoring (BAM) are obtained from 15 stations. Hourly $PM_{2.5}$ observations at each station are temporally averaged to obtain daily $PM_{2.5}$ values.

2.5 Simulation Scenarios

To investigate the effects of land surface changes via historical urbanization on regional meteorology and air quality in Southern California, we carry out two simulations, which we refer to as the "Present-day" scenario and "Nonurban" scenario. The two scenarios differ only by the assumed land surface properties and processes, which are shown in Figure 2. The Present-day scenario assumes the land cover (Figure 1b) and irrigation of current for Southern California (described in Section 2.2). Urban morphology from NUDAPT and LARIAC, and MODIS-retrieved albedo, GVF and LAI are used in this scenario. To help explain the impact of urbanization without the addition of irrigation, a supplemental simulation, which we refer to as "Present-day No-irrigation", is also carried out; this simulation is identical to "Present-day" but assumes that there is no irrigation. For the Nonurban scenario, we assume natural land cover prior to human perturbation, and replace all urban grid cells with "shrubs" (Figure 1c). We modify MODIS-retrieved albedo, GVF and LAI in these areas based on properties for shrub lands surrounding urban regions in the Present-day scenario. A detailed explanation on this method (inverse distance weighting approach) can be found in Vahmani et al. (2016). The spatial pattern of land surface properties in both "Present-day" and "Nonurban" scenarios are shown in Figure S10~~6~~. Note that all three aforementioned scenarios adopt identical anthropogenic emission inventories described in Section 2.3. Using current anthropogenic emissions for "Nonurban" is a hypothetical scenario that cannot exist in reality, but allows us to tease out the effects of land surface changes via urbanization on meteorology and air pollutant concentrations. (Biogenic emissions do change for the scenarios due to changes in land surface properties (e.g., vegetation type and LAI) and meteorology (e.g., temperature).) To check whether the influence on regional meteorology and air quality due to land surface changes are distinguishable from zero, statistical significance at 95% confidence interval is tested using the paired Student's t-test with n = 7 days

2.6 Uncertainties

Note that the results reported in this paper are based on model simulations and are thus dependent on how accurately the regional climate/chemistry model characterizes the climate/chemistry system (e.g., meteorology, surface-atmosphere coupling, and atmospheric chemical reactions). Results may be dependent on model configuration (e.g., physical and chemical schemes), land surface characterizations (e.g., satellite data from MODIS, or default dataset available in WRF) and emission inventories (e.g., anthropogenic emission inventories from CARB, SCAQMD or NEI). In addition, since irrigation is not included in the Nonurban scenario, simulated meteorology in the Nonurban scenario are dependent on assumed soil moisture initial conditions. In this study, we adopt the initial soil moisture conditions from Vahmani et al. (2016) for consistency with our previous work. Soil moisture initial conditions are based on values from six-month simulations without irrigation (Vahmani and Ban-Weiss, 2016b).

3. Results and Discussion

3.1 Evaluation of Simulated Meteorology and Air Pollutant Concentrations

In this section, we focus on the predicative capability of the model for simulated near-surface air temperature, O_3 and total $PM_{2.5}$ concentrations (including sea salt, but excluding dust) for the Present-day scenario. Note that for the evaluation of $PM_{2.5}$ concentrations we include only observations from daily (gravimetric) measurements in this section. The comparison between modeled $PM_{2.5}$ concentrations versus daily averaged observations derived from hourly BAM measurements is discussed in the supplemental information section S1. In addition, we only include observations from monitoring sites that are located in urban grid cells in the Present-day scenario. The validation of near surface air temperatures for both urban and nonurban sites are discussed in section S2 in the supplemental information. Figure 3 shows the comparison between observed and modeled hourly near surface air temperature, O_3 concentrations, and daily $PM_{2.5}$ concentrations. (Comparisons between observed and modeled diurnal cycles for near surface air temperatures and O_3 concentrations are also presented in the supplemental information, Figure S11 and S12.) As shown in Figure 3 (and Figure S11), the model simulations better capture higher air temperatures during the daytime relative to lower values during nighttime. By contrast, predictions of O_3 and $PM_{2.5}$ concentrations show good fit with observations at low values that occur with high occurrence frequency. However, observed O_3 and $PM_{2.5}$ concentrations are underestimated by the model at higher values that occur with lower frequency of occurrence. The underestimation of $PM_{2.5}$ concentrations may be occurring due to one or more of the following factors: 1) not including dust emissions in the simulation, which makes up an appreciable fraction of real-world total $PM_{2.5}$, 2) a failure of the emissions inventory to capture high emission rates on particular days, 3) the chemistry parameterizations in WRF/Chem tending to underestimate $PM_{2.5}$ concentrations at high values, and 4) errors in simulated air pollution meteorology. Table 1 shows four statistical metrics for model evaluation, including mean bias (MB) and normalized mean bias (NMB) for the quantification of bias, and mean error (ME) and root mean square error (RMSE) for the quantification of error. The statistical results indicate that ~~while~~ model simulations underestimate near-surface air temperature, O_3 and $PM_{2.5}$ concentrations by 1.0 K~~0.3%~~, 22% and 31%, respectively.~~,~~ ~~their performance on capturing observations are acceptable.~~ The comparison between our evaluation results and recommended model performance benchmarks is presented in the supplemental information Table S2.

3.2 Effects of Urbanization on Air Temperature and Ventilation Coefficient

The effects of land surface changes via urbanization in Southern California on air temperature and ventilation coefficient are discussed in this section. Air temperatures are reported for the lowest atmosphere model layer rather than the default diagnostic 2m (near-surface) air temperature variable to be consistent with reported air pollutant concentrations shown in later sections. (The chemistry code makes use of grid cell air temperature and does not use 2m air temperature.) Ventilation coefficient is calculated as the product of PBL height and the average wind speed within the PBL, and thus considers the combined effects of vertical and horizontal mixing, and indicates the ability of the atmosphere to disperse air pollutants (Ashrafi et al., 2009). The integral form of this calculation can be written as (Eq1).

$$\text{Ventilation Coefficient} = \int_0^{\text{PBL height}} U(z)\ dz \qquad \text{(Eq1)}$$

Given that the atmosphere is stratified in models, Eq1 can be discretized as Eq2:

$$\text{Ventilation Coefficient} = \sum_{i=1}^{m} U(z_i) \times \Delta z_i \qquad \text{(Eq2)}$$

where $U(z_i)$ stands for horizontal wind speed within the i^{th} model layer (m/s), Δz_i is the depth of the i^{th} model layer that is within the PBL (m), and m is the number of vertical layers up to PBL height.

3.2.1 Spatial average temperature change

As shown in Figure 4a, urbanization in Southern California has in general led to urban temperature reductions during daytime from 7 PST to 16 PST, and urban temperature increases during other times of day. The largest spatially averaged temperature reduction occurs at 10 PST (ΔT = –1.4 K), whereas the largest temperature increase occurs at 20 PST (+1.7 K). Additionally, urbanization led to spatially averaged reduction in diurnal temperature range by 1.5 K. Spatially averaged urban temperature reductions during morning (i.e., defined here and in the following sections as 7:00 – 12:00 PST) and afternoon (i.e., 12:00 – 19:00 PST) are –0.9 K and –0.3 K, respectively. At nighttime (i.e., 19:00 – 7:00

PST), the spatially averaged temperature increase is +1.1 K. The spatially averaged changes significantly differs from zero at the 95% confidence level for all three times of the day using the paired Student's t-test with n=7 days.

3.2.2 Spatial distributions of temperature change

During the morning, temperature reductions are larger in regions further away from the sea (e.g., San Fernando Valley and Riverside County) than coastal regions (e.g., west Los Angeles and Orange County) (Figure 5a). (Note that regions that are frequently mentioned in this study are in Figure 2a.) Spatial patterns in the afternoon are similar to morning, with the exception that coastal regions experience temperature increases (as opposed to decreases) of up to +0.82 K (Figure 5b). During nighttime, temperature increases spread throughout urban regions, and are generally larger in the inland regions of the basin relative to coastal regions (Figure 5c). A modified version Figure 5 that includes values for non-urban cells is in the supplemental information Figure S13.

3.2.3 Processes driving daytime changes

The temporal and spatial patterns of air temperature changes suggest that the climate response to urbanization during daytime is mainly associated with the competition between (a) temperature reductions from increased evapotranspiration and thermal inertia from urban irrigation, and (b) temperature increases from decreased onshore sea breezes (Figure S147a, bd, e). Decreases in the onshore sea breeze are primarily caused by increased roughness lengths from urbanization. (Note that the onshore sea breeze decreases in strength despite higher temperatures in the coastal region of Los Angeles during the afternoon, which would tend to increase the land-sea temperature contrast and thus be expected to increase the sea breeze strength.) Inland regions show larger temperature reductions relative to coastal because they have lower urban fractions (Figure S106a), and thus higher pervious fractions. Since irrigation increases soil moisture in the pervious fraction of the grid cell in this model, irrigation will have a larger influence on grid cell averaged latent heat fluxes (Figure S158) and thermal inertia when pervious fractions are higher. The inland regions are also less affected by changes in the sea breeze relative to coastal regions since they are (a) farther from the ocean, and (b) experience smaller increases in roughness length. Roughness length effects on the sea breeze are especially important in the afternoon when baseline wind speeds are generally highest in the Los Angeles basin. Thus, the afternoon temperature increases simulated in the coastal region occur because temperature increases from reductions in the afternoon onshore flows dominate over temperature decreases from increased evapotranspiration. In addition, increases in thermal inertia caused by use of manmade materials (e.g., pavements and buildings) and shading effects within urban canopies can contribute to simulated temperature reductions during the morning. Please see the supplemental information section S32 for the additional simulation (Present-day No-irrigation scenario) carried out to identify the influence of urbanization but without changing irrigation relative to the Nonurban scenario (i.e., with no irrigation).

Note that changes in air temperature during daytime shown here disagree with Vahmani et al. (2016). While our study detects daytime temperature reductions due to urbanization, Vahmani et al. (2016) suggests daytime warming. After detailed comparison of the simulations in our study versus Vahmani et al. (2016), we find that the differences are mainly associated with UCM configuration. First, our study uses model default calculations of surface temperature for the impervious portion of urban grid cells, whereas Vahmani et al. (2016) applied thean alternative calculation proposed by Li and Bou-Zeid, 2014. Li and Bou-Zeid, 2014 intended the alternate surface temperature calculations to be performed as a post-processing step rather than during runtime.our study accounts for shadow effects in urban canopies, whereas Vahmani et al. (2016) assumes no shadow effects. (We note here that the

~~default version of the UCM has the shadow model turned off. The boolean SHADOW variables in module_sf_urban.F needs to be manually switched to true to enable the shadow model calculations. With the shadow model turned off, all shortwave radiation within the urban canopy is assumed diffuse.) We suggest that it is important to include the effects of building morphology on shadows within the canopy, and to track direct and diffuse radiation separately, and therefore perform simulations in this study with the shadow model on.~~ After a careful comparison among different model set-ups, we found that the parameterization of surface temperature is an important factor that affects simulated daytime air temperature (See Figure S16). Second, our study accounts for shadow effects in urban canopies, whereas Vahmani et al. (2016) assumes no shadow effects. (We note here that the default version of the UCM has the shadow model turned off. The boolean SHADOW variable~~s~~ in module_sf_urban.F needs to be manually switched to true to enable the shadow model calculations. With the shadow model turned off, all shortwave radiation within the urban canopy is assumed diffuse.) We suggest that it is important to include the effects of building morphology on shadows within the canopy, and to track direct and diffuse radiation separately, and therefore perform simulations in this study with the shadow model on. Note that the effect of shadows is not as significant as the parameterization of surface temperature for most of the domain in our study because the ratio between building height and road width is small. ~~our study uses model default calculations of surface temperature for the impervious portion of urban grid cells, whereas Vahmani et al. applied the alternative calculation proposed by Li and Bou-Zeid, 2014. Li and Bou-Zeid, 2014 intended the alternate surface temperature calculation to be performed as a post-processing step rather than during runtime.~~

3.2.4 Processes driving nighttime changes

The ~~temporal and spatial patterns of air temperature changes suggest that the~~ climate response to urbanization during nighttime is driven by the ~~competition~~ combined effects of~~between~~ (a) temperature increases from increasing upward ground heat fluxes, and (b) temperature ~~reductions~~ increases from increasing PBL heights. Increased soil moisture (from irrigation) and use of man-made materials leads to higher thermal inertia of the ground; this in turn leads to increased heat storage during the day and higher upward ground heat fluxes and thus surface temperatures at night. ~~The magnitude of air temperature change during nighttime is also related to the magnitude of PBL height changes; i~~Increasing PBL heights can ~~counteract~~ also lead to warming because of lower air cooling rates during nighttime.~~driven by higher upward ground heat flux. Greater increases in PBL heights will lead to increasingly diminished air heating rates, and thus smaller air temperature increases.~~ Changes in PBL heights are associated with surface roughness changes since shear production dominants TKE at night. Coastal (inland) regions ~~have~~ show larger (smaller) ~~increase in~~ variation in roughness length (Figure 2e), which leads to larger (smaller) increases in PBL heights (Figure S14c)~~, and thus smaller (greater) increase in air temperature~~. Despite larger increases in PBL heights in coastal versus inland regions, smaller air temperature increases occur in coastal versus inland regions, likely due to accumulative effects from coastal to inland regions with onshore wind flows.

3.2.5 Temporal and spatial patterns of ventilation changes and process drivers

Changes in ventilation coefficient show a similar temporal pattern as air temperature (Figure 4b); values decrease by up to –36.6% (equivalent to –826 m^2/s, at 10 PST) during daytime, and increase up to +27.0% (equivalent to +77 m^2/s, at 23 PST) during nighttime, due to urbanization. Absolute reductions in ventilation coefficient are more noticeable in the afternoon than in the morning; the spatially averaged decreases are –726 m^2/s (–23%) and –560 m^2/s (–34%), respectively. These reductions significantly differ from zero at 95% confidence level using the paired Student's t-test with n=7 days. Reductions during daytime are also generally greater in inland regions than in coastal regions as shown is Figure 5d and 5e. Daytime reductions in ventilation occur due to the combined effect of weakened wind speeds due to higher surface roughness and changes (mostly decreases) in PBL heights (Figure S14~~7~~). Changes in PBL heights during daytime are mainly associated with air temperature changes because buoyancy production dominants TKE during the day. Where there are larger air temperature decreases (increases), there is reduced (increased) buoyancy production of TKE, which results in shallower (deeper) PBLs.

At night, spatially averaged ventilation coefficient increases by +8.2% (+24.3 m^2/s). This increase significantly differs from zero at 95% confidence level. As shown in Figure 5f, ~~greater~~ statistically significant ventilation growth occurs in most parts of coastal Los Angeles and Orange County, likely due to higher PBL height increases (i.e., stemming from higher surface roughness increases from urbanization). By contrast, in Riverside County, the effect of reductions in wind speed surpasses ~~slight increases~~changes in PBL heights, leading to slight but not statistically significant reductions in atmospheric ventilation (Figure S14~~7~~).

3.3 Effects of Urbanization on NOx and O_3 Concentrations due to Meteorological Changes

Concentrations of pollutants are profoundly impacted by meteorological conditions including air temperature and the ventilation capability of the atmosphere (Aw and Kleeman, 2003; Rao et al., 2003). This section discusses how meteorological changes due to land surface changes via urbanization in Southern California affect gaseous pollutant concentrations (i.e., NOx and O_3).

3.3.1 Temporal and spatial patterns of NOx concentration changes and process drivers

As shown in Figure 6a, changes in meteorological fields due to urbanization have led to increases in hourly NOx concentrations during the day (7 PST to 18 PST) and decreases at all other times of day. Peak increases in NOx of +2.7 ppb occur at 10 PST (i.e., for spatial mean values), while peak decreases of –4.7 ppb occur at 21 PST. Spatial mean changes in NOx concentrations are +2.1 ppb and +1.2 ppb in the morning and afternoon, respectively, and –2.8 ppb at night. The spatially averaged changes are significantly different from zero at 95% confidence level for all three times of the day. In addition, daily 1-hour maximum NOx concentrations change only slightly: from 17.8 ppb at 6 PST in the Nonurban scenario to 17.9 ppb at 7 PST in the Present-day scenario.

Figures 7a,b,c show the spatial patterns of NOx concentration changes due to urbanization. In the morning (afternoon), most inland urban regions show statistically significant increases in NOx concentrations (Figure 7a, b), with larger NOx concentration increases of up to +13.8 ppb (+5.5 ppb) occurring in inland regions compared to coastal regions. By contrast, NOx concentrations decrease at night across the region, with the largest decreases reaching –20.8 ppb. In general, greater decreases are shown in inland regions compared to coastal regions.

The spatial patterns of changes in NOx concentrations are similar to those for CO concentrations (Figure 7d,e,f). CO is an inert species and can be used as a tracer for determining the effect of ventilation on air pollutant dispersion since it includes accumulation effects of ventilation changes both spatially and temporally. Thus, the similarity in changes to NOx and CO spatial patterns suggests that NOx changes are driven by ventilation changes. For example, at night, Riverside County shows decreases of up to –20.8 ppb in NOx concentrations (with corresponding decreases in CO of –119 ppb) despite suppressed ventilation at this location because of accumulative effects from coastal to inland regions. A modified version of Figure 7 that includes values for non-urban cells is in the supplemental information Figure S17.

3.3.2 Temporal and spatial patterns of O_3 concentration changes

As indicated by Figure 6b, O_3 concentrations in the lowest atmospheric layer decrease from 7 PST to 11 PST, and increase during other times of day. The largest decrease of –0.94 ppb occurs at 10 PST, while the largest increase of +5.6 ppb occurs at 19 PST. Spatially averaged hourly O_3 concentrations undergo a –0.6 ppb decrease, +1.7 ppb increase, and +2.1 ppb increase in the morning, afternoon, and night, respectively. The spatially averaged changes significantly differ~~s~~ from zero at 95% confidence level for all three times of the day. Additionally, daily 1-hour maximum O_3 concentrations, which occurs at 14 PST in both scenarios, increases by +3.4%, from 41.3 ppb in the Nonurban scenario to 42.7 ppb in the Present-day scenario. The daily 8-hour maximum O_3 concentration increases from 38.0 ppb to 39.3 ppb (averaged over 11 PST to 19 PST in both scenarios).

Figure 7g,h,i show the spatial patterns of surface O_3 concentration changes. In the morning (Figure 7g), while most regions show reductions in O_3 concentrations, the reductions are in general statistically insignificant.~~with the largest decrease of up to –5.4 ppb in the inland regions. By contrast,~~ During the afternoon, most inland urban regions show increases in O_3 concentrations~~ during the afternoon~~ (Figure 7h), with the largest increase of +5.7 ppb occurring in Riverside County. Increases in O_3 concentrations are larger during night than the afternoon (Figure 7i), especially in the Riverside County, with the largest increase in O_3 concentrations reaching +12.8 ppb.

3.3.3 Processes driving daytime and nighttime changes in O_3

The temporal and spatial patterns of changes in O_3 concentrations during the day suggest that these changes are mainly driven by the competition between (a) decreases in ventilation, which would tend to cause increases in O_3, and (b) the nonlinear response of O_3 to NOx changes. In the VOC-limited regime, increases in NOx tend to decrease O_3 concentrations, and vice versa. (This explains why decreases in NOx emissions over weekends can cause increases in O_3 concentrations, a phenomenon termed the “weekend effect” (Marr & Harley, 2002).) The underlying cause of the weekend effect has to do with titration of O_3 by NO, as shown in R1.

$$NO + O_3 \rightarrow NO_2 + O_2 \quad \text{(R1)}$$

When NOx is high relative to VOC, R1 dominates NO to NO_2 conversion, which involves consuming O_3. In addition, increases in NO_2 can reduce OH lifetime due to increased rates of the OH + NO_2 reaction (R2), which is chain terminating.

$$NO_2 + OH + M \rightarrow HNO_3 + M \quad \text{(R2)}$$

In addition to these two aforementioned processes, changes in air temperature can also affect the production rate of O_3, with higher temperatures generally leading to higher O_3 (Steiner et al., 2010).

In the morning when ventilation is relatively weak (shallow PBL and weak sea breeze), changes in NOx concentrations play an important role in driving surface O_3 concentrations. Regions with greater increases in NOx concentrations in general show greater decreases in O_3 concentrations (Figure 7g). Decreases in air temperature would also contribute to decreases in O_3 concentrations due to reductions in O_3 production rates. In the afternoon when ventilation is strengthened (deep PBL, and stronger sea breeze), changes in both NOx concentrations and ventilation play important roles in determining O_3 concentrations (Figure 7h). Regions with higher increases in NOx concentrations tend to have lower increases in O_3 concentrations; this indicates that NOx increases (that would tend to decrease O_3) are counteracting decreases in ventilation (that would tend to increase O_3). In regions with relatively lower increases in NOx concentrations and greater decreases in ventilation, such as Riverside County, increases in O_3 concentrations are larger.

At night, changes in O_3 concentrations are dominated by its titration by NO_2 as shown in (R3).

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \quad \text{(R3)}$$

Where there are larger decreases in NOx concentrations (Figure 7c), there are greater increases in O_3

concentrations (Figure 7i), regardless of the magnitude of increases in atmospheric dilution (Figure 5f).

3.4 Effects of Urbanization on Total and Speciated $PM_{2.5}$ Concentrations due to Meteorological Changes

In this section, we discuss changes in total and speciated $PM_{2.5}$ mass concentrations due to urbanization. Total mass concentrations reported here only consider $PM_{2.5}$ generated from anthropogenic and biogenic sources mentioned in section 2.3, and exclude sea salt and dust. Speciated $PM_{2.5}$ is classified into three categories: (secondary) inorganic aerosols including nitrate (NO_3^-), sulfate (SO_4^{2-}) and ammonium (NH_4^+); primary carbonaceous aerosols including elemental carbon (EC), and primary organic carbon (POC); and secondary organic aerosol (SOA) including SOA formed from anthropogenic VOC precursors (ASOA) and biogenic VOC precursors (BSOA).

3.4.1 Temporal patterns of total and speciated $PM_{2.5}$ concentration changes

Figure 8 illustrates diurnal changes in total and speciated $PM_{2.5}$ concentrations due to meteorological changes attributable to urbanization. As suggested by Figure 8a, urbanization is simulated to cause slight spatially averaged increases in total $PM_{2.5}$ concentrations from 9 PST to 16 PST (up to +0.62 μg/m^3 occurring at 12 PST), and decreases during other times of day (up to –3.1 μg/m^3 at 0 PST). Increases in total $PM_{2.5}$ during 9 PST to 16 PST come from increases in primary carbonaceous aerosols, and nitrate; these species show hourly averaged concentration increases of up to +0.21, +0.14 μg/m^3, respectively. By contrast, BSOA decreases slightly during these hours. During other times of day, concentrations of all $PM_{2.5}$ species decrease dramatically. Inorganic aerosols, primary carbonaceous aerosols, and SOA show decreases of up to –1.7, –0.5 and –0.3 μg/m^3, respectively.

During morning hours, averaged hourly total $PM_{2.5}$ concentrations decrease by –0.20 μg/m^3 but are not statistically significant., with 74% and 73% of the decrease contributed by changes in inorganic aerosols and SOA, respectively (Figure 8). In the afternoon, spatially averaged total $PM_{2.5}$ concentrations increase by +0.24 $\mu g/m^3$. Primary carbonaceous aerosols contribute to half of the increase (+0.12 $\mu g/m^3$). For nighttime, total $PM_{2.5}$ concentrations undergo a decrease of –2.5 $\mu g/m^3$, with 54% of the decrease attributed to changes in inorganic aerosols and 17% by primary carbonaceous aerosols. Both afternoon and nighttime changes are significantly different from zero at 95% confidence interval.

3.4.2 Spatial patterns of total and speciated $PM_{2.5}$ concentration changes

Figure 9 presents spatial patterns of changes in total and speciated $PM_{2.5}$ due to urbanization. Decreases in concentrations prevail in urban regions during morning and night, whereas increases in concentrations are dominant during the afternoon.

In the morning, the spatial pattern of changes in total $PM_{2.5}$ concentrations (Figure 9a) is similar to that of inorganic aerosols (Figure 9d). The west coastal region shows increases of up to +0.59 $\mu g/m^3$ in total $PM_{2.5}$ and +0.44 $\mu g/m^3$ in inorganic aerosols. In addition, increases occurring in some inland regions are caused by changes in primary carbonaceous aerosols. Other inland regions show decreases of up to –4.2 $\mu g/m^3$ in inorganic aerosol and –2.7 $\mu g/m^3$ in total $PM_{2.5}$ concentrations.changes in total $PM_{2.5}$ and speciated $PM_{2.5}$ are not statistically distinguishable from zero at 95% confidence level.

In the afternoon, increases in total $PM_{2.5}$ are statistically significant in only some inland regions, driven mostly by (up to +1.4 $\mu g/m^3$, Figure 9b) increases in include contributions from 1) inorganic aerosols (up to +0.8 $\mu g/m^3$, Figure 9e), 2) primary carbonaceous aerosols (up to +0.5 $\mu g/m^3$, Figure 9h), and 3) SOA (up to +0.3 $\mu g/m^3$, Figure 9.k).

At night, most regions within the Los Angeles metropolitan area show decreases in total $PM_{2.5}$ of –

3.0 to –6.0 $\mu g/m^3$ (Figure 9c) with contributions from all three categories of speciated $PM_{2.5}$.

A modified version of Figure 9 that includes values for non-urban cells is in the supplemental information Figure S18.

3.4.3 Processes driving daytime and nighttime changes in $PM_{2.5}$

During the day, changes in speciated $PM_{2.5}$ concentrations are dictated by the relative importance of various competing pathways, including (a) reductions in ventilation causing increases in $PM_{2.5}$, (b) changes in gas-particle phase partitioning causing increases (decreases) in $PM_{2.5}$ from decreases (increases) in temperature, and (c) increases (decreases) in atmospheric oxidation from increases (decreases) in temperature. Changes in ventilation appear to dominate the changes in primary carbonaceous aerosols, as indicated by the similarity in spatial pattern to changes in CO, which can be considered a conservative tracer (Figure 7d and 7e). As for semi-volatile compounds such as nitrate aerosols (red dotted curve in Figure 8b) and some SOA species, concentrations increase during daytime hours. This is because both decreased ventilation and gas-particle phase partitioning effects favoring the particle phase (from temperature decreases) outweigh reductions in atmospheric oxidation. Concentrations of sulfate and ammonium slightly increase due to urbanization (blue dotted curve in Figure 8b). Since sulfate is nonvolatile, gas-particle phase partitioning does not affect sulfate concentrations; lowered atmospheric oxidation rates due to reduced temperatures (which would tend to decrease sulfate) nearly offset the effect of weakened ventilation (which would tend to increase sulfate). In addition, BSOA concentrations are simulated to decrease (blue dotted curve in Figure 8d) due to reduced biogenic VOC emissions, which occur due to reductions in both vegetation coverage and air temperature from urbanization.

At night, decreases in $PM_{2.5}$ across urban regions are due to (1) enhanced ventilation owing to deeper PBLs (relevant for all PM species), and (2) gas-particle phase partitioning effects that favor the gas phase for semi-volatile compounds (i.e., nitrate aerosols and some SOA species) because of higher air temperatures.

4. Conclusion

In this study, we have characterized the impact of land surface changes via urbanization on regional meteorology and air quality in Southern California using an enhanced version of WRF/Chem-UCM. We use satellite data for the characterization of land surface properties, and include a Southern California-specific irrigation parameterization. The two main simulations of focus in this study are the real-world "Present-day" and the hypothetical "Nonurban" scenarios; the former assumes current land cover distributions and irrigation of vegetative areas, while the latter assumes land cover distributions prior to widespread urbanization and no irrigation. We assume identical anthropogenic emissions in these two simulations to allow for focusing on the effects of land cover change on air pollutant concentrations.

Our results indicate that land surface modifications from historical urbanization have had a profound influence on regional meteorology. Urbanization has led to daytime reductions in air temperature for the lowest model layer and reductions in ventilation within urban areas. The impact of urbanization at nighttime shows the opposite effect, with air temperatures and ventilation coefficients increasing. Spatially averaged reductions in air temperature and ventilation during the day are –0.6 K and –650 m^2/s respectively, whereas increases at night are +1.1 K and 24.3 m^2/s respectively. Changes in meteorology are spatially heterogeneous; greater changes are simulated in inland regions for (a) air temperatures decreases during day and increases during night, and (b) ventilation reductions during daytime. Ventilation at night shows increases in coastal areas and decreases in inland areas. Changes in meteorology are mainly attributable to (a) increased surface roughness from buildings, (b) higher evaporative fluxes from irrigation, and (c) higher thermal inertia from building materials and increased soil moisture (from irrigation).

Changes in regional meteorology in turn affect concentrations of gaseous and particulate pollutants. NOx concentrations in the lowest model layer increase by +1.6 ppb during the day, and decrease by –2.8 ppb at night, due to changes in atmospheric ventilation. O_3 concentrations decrease by –0.6 ppb in the morning, and increase by +1.7 (2.2) ppb in the afternoon (night). Decreases in the morning and increases during other times of day are more noticeable in inland regions. Changes in O_3 concentrations are mainly attributable to the competition between (a) changes in atmospheric ventilation, and (b) changes in NOx concentrations that alter O_3 titration. Note that while changes in air temperature can also influence O_3 concentrations during the day, this effect is overwhelmed by changes in ventilation and concentrations of NOx in our study. As for $PM_{2.5}$, total mass concentrations increase by +0.24 μg/m^3 in the afternoon, and decrease by –0.20 (–2.5) μg/m^3 atin the morning (night). Changes during the morning are not statistically significant. The major driving processes of changes in $PM_{2.5}$ concentrations are (a) changes in atmospheric ventilation, (b) changes in gas-particle phase partitioning for semi-volatile compounds due to air temperature changes, and (c) changes in atmospheric chemical reaction rates from air temperature changes.

Our findings suggestThis study highlights the role that land cover properties can have on regional meteorology and air quality. We find that increases in evapotranspiration, thermal inertia, and surface roughness due to historical urbanization are the main drivers of regional meteorology and air quality changes in Southern California. During the day, our simulations suggest that urbanization has led to regional air temperature reductions but increased ozone and $PM_{2.5}$ concentrations. During nighttime, urbanization has led to increases in regional air temperatures and O_3 concentrations, but decreases in

NOx and $PM_{2.5}$ concentrations. ~~Our study provides insight into the potential impacts of land surface modifications via urbanization on regional meteorology and air quality, and can be informative for decision making on sustainable urban planning to achieve a balance between climate mitigation/adaptation and air quality improvements.~~ Our findings indicate that air pollutant concentrations have been impacted by land cover changes since pre-settlement times (i.e., urbanization), even assuming constant anthropogenic emissions. These air pollutant changes are driven by urbanization-induced changes in meteorology. This suggests that policies that impact land surface properties (e.g., urban heat mitigations strategies) can have impacts on air pollutant concentrations (in addition to meteorological impacts); to the extent possible, all environmental systems should be taken into account when studying the benefits or potential penalties of policies that impact the land surface in cities.

Author Contributions

GBW designed the study. YL performed the model simulations, carried out data analysis, and wrote the manuscript. GBW and DS mentored YL. JZ contributed to the setup of WRF/Chem-UCM. All authors contributed to editing the paper.

Competing Interests

The authors declare that they have no conflict of interest.

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Figures and Tables

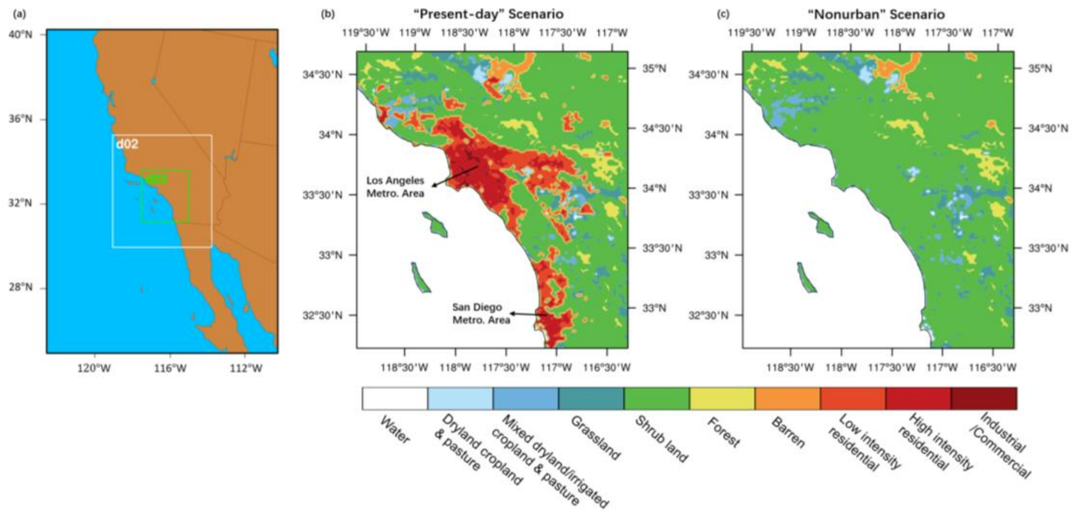


Figure 1. Maps of (a) the three nested WRF/Chem-UCM domains, and (b,c) land cover types for the innermost domain (d03) for the (b) Present-day and (c) Nonurban scenarios.

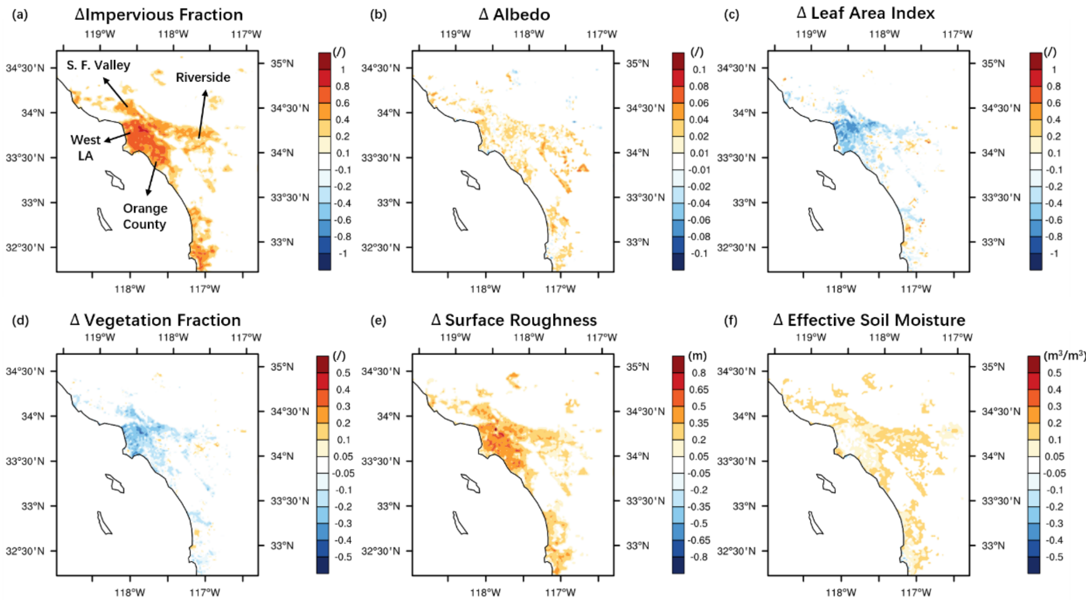


Figure 2. Spatial patterns of differences (Present-day – Nonurban) in land surface properties for urban grid cells. Panels (a) to (f) are changes in impervious fraction, albedo, leaf area index (LAI), vegetation fraction (VEGFRA), surface roughness, and effective soil moisture, respectively. Effective soil moisture is calculated as the product of pervious fraction for urban grid cells (1 – impervious fraction) and soil moisture for the pervious portion of the grid cell.

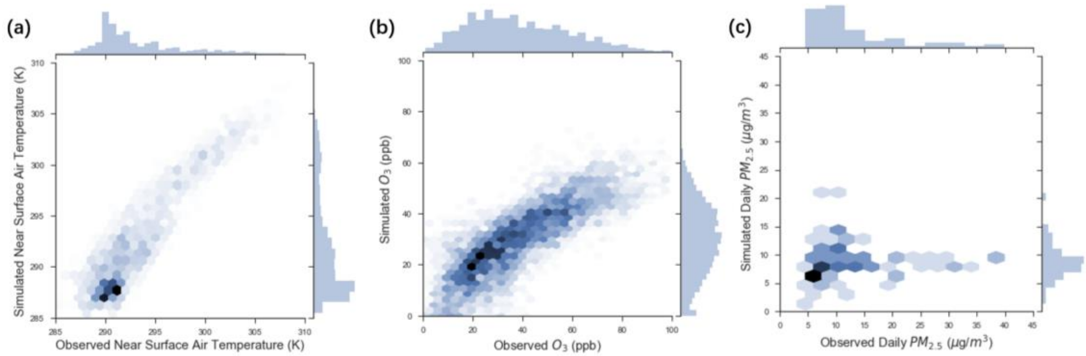


Figure 3. Comparison between modeled and observed (a) hourly near-surface air temperature (K), (b) hourly O_3 concentrations (ppb), and (c) daily $PM_{2.5}$ concentrations ($\mu g/m^3$). Note that daily $PM_{2.5}$ concentrations from simulations include sea salt, but exclude dust. Darker hexagonal bins correspond to higher point densities in the scatter plots. Histograms of both observations and modeled values are also shown at the edges of each panel.

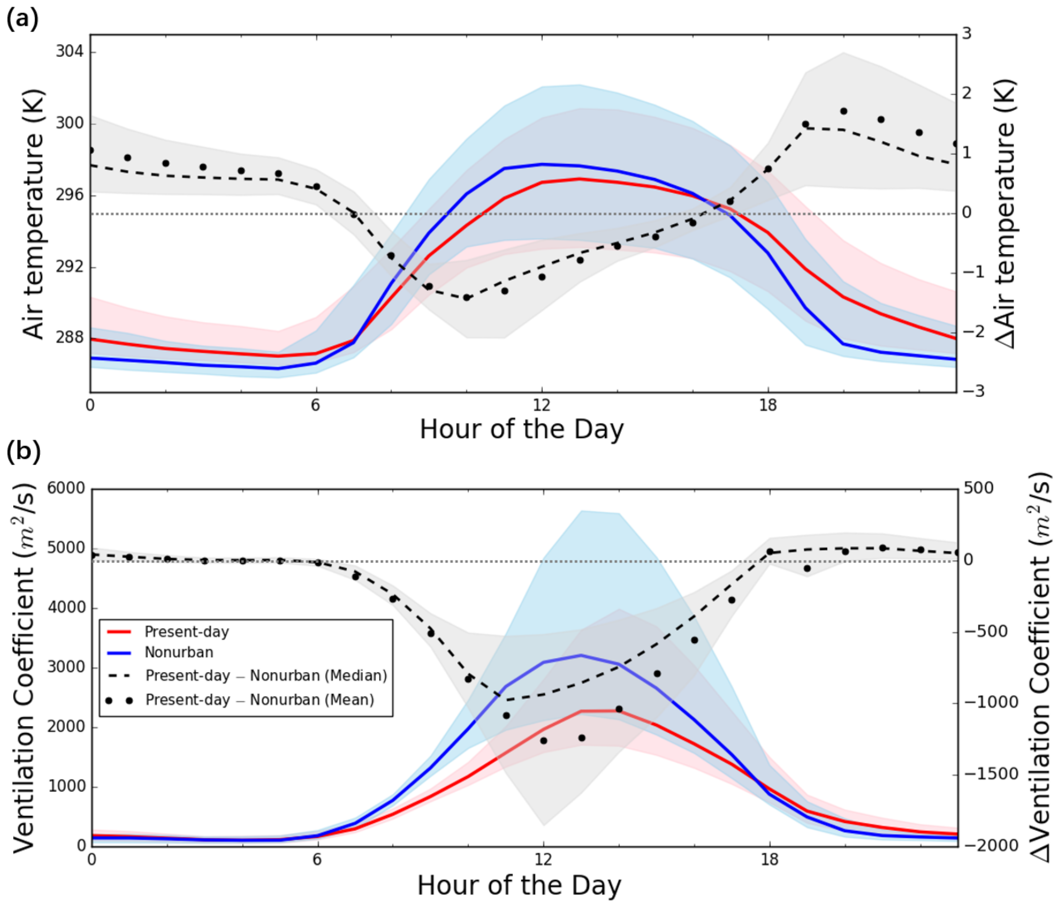
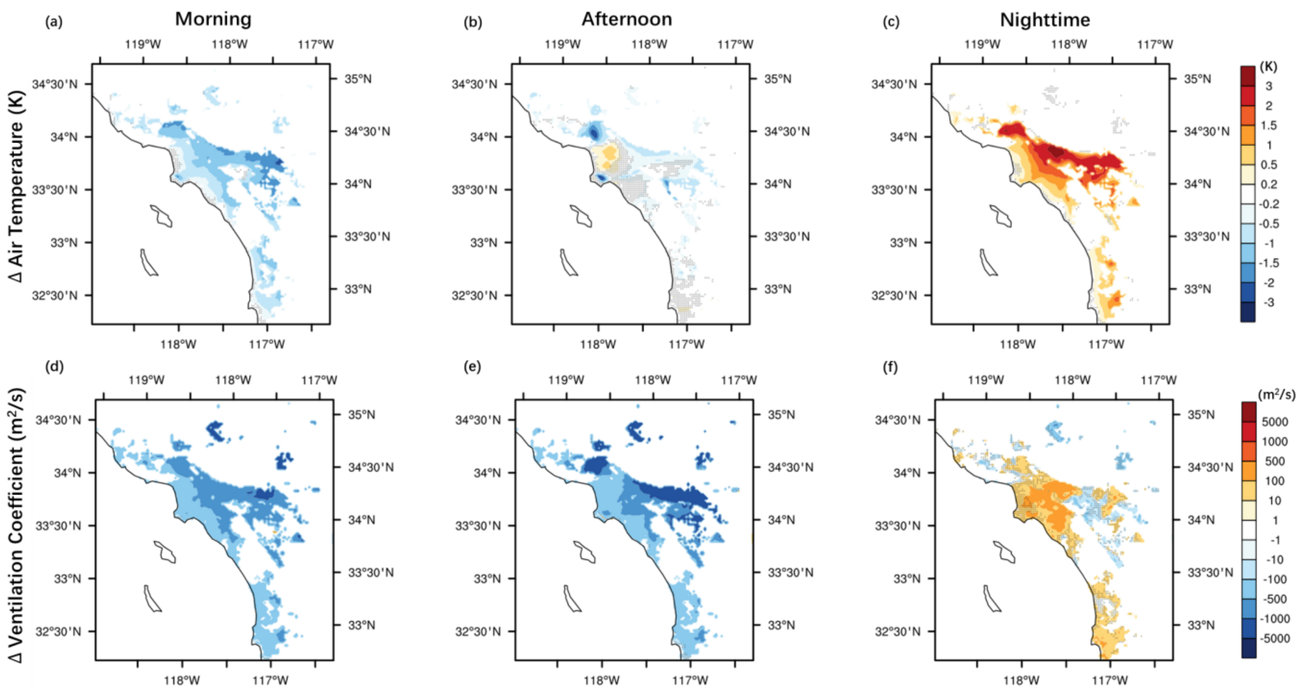


Figure 4. Diurnal cycles for present-day (red), nonurban (blue), and present-day – nonurban (black) for (a) air temperature in the lowest atmospheric layer (K) and (b) ventilation coefficient (m^2/s). Values are obtained by averaging over urban grid cells and the entire simulation period for each hour of day. The solid and dashed curves give the median values, while the shaded bands show 25th and 75th percentiles. Dots indicate mean values for differences between Present-day and nonurban. The horizontal dotted line in light grey shows $\Delta = 0$ as an indicator of positive or negative change by land surface changes via urbanization.



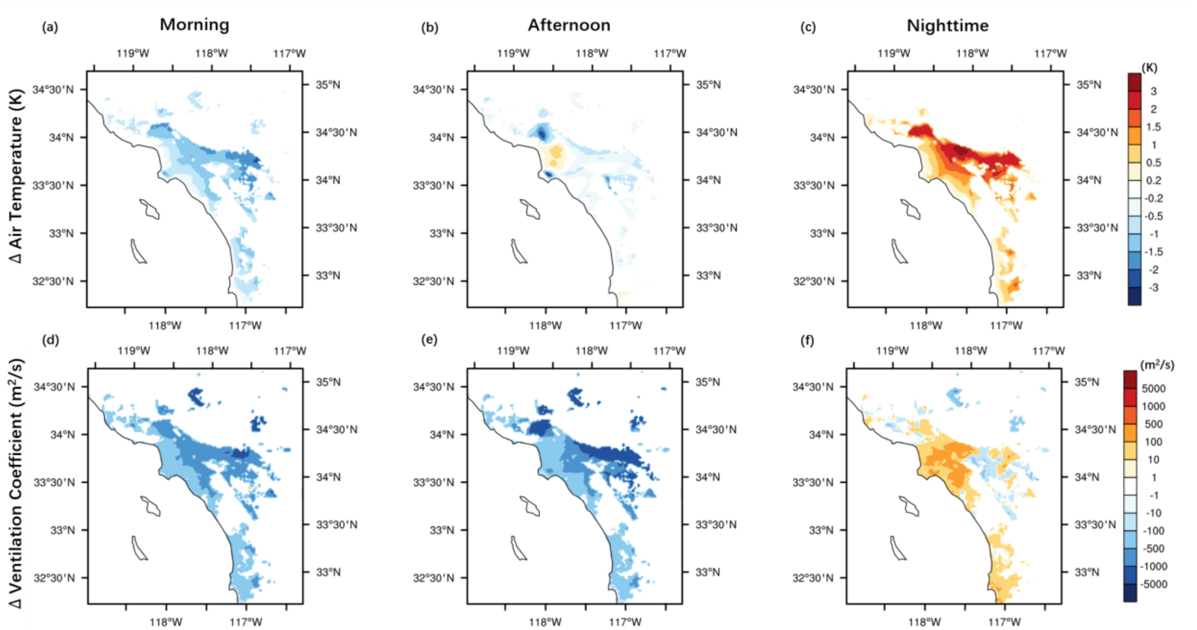


Figure 5. Spatial patterns of differences (Present-day – nonurban) in temporally averaged values during morning, afternoon and nighttime for (a,b,c) air temperature in the lowest atmospheric layer, and (d,e,f) ventilation coefficient. Morning is defined as 7 PST to 12 PST, afternoon as 12 PST to 19 PST, and nighttime as 19 PST to 7 PST. We refer to

morning and afternoon as daytime. Note that values are shown only for urban grid cells. Black dots indicate grid cells where changes are not significantly different from zero at 95% confidence level using the paired Student's t-test with n=7 days.

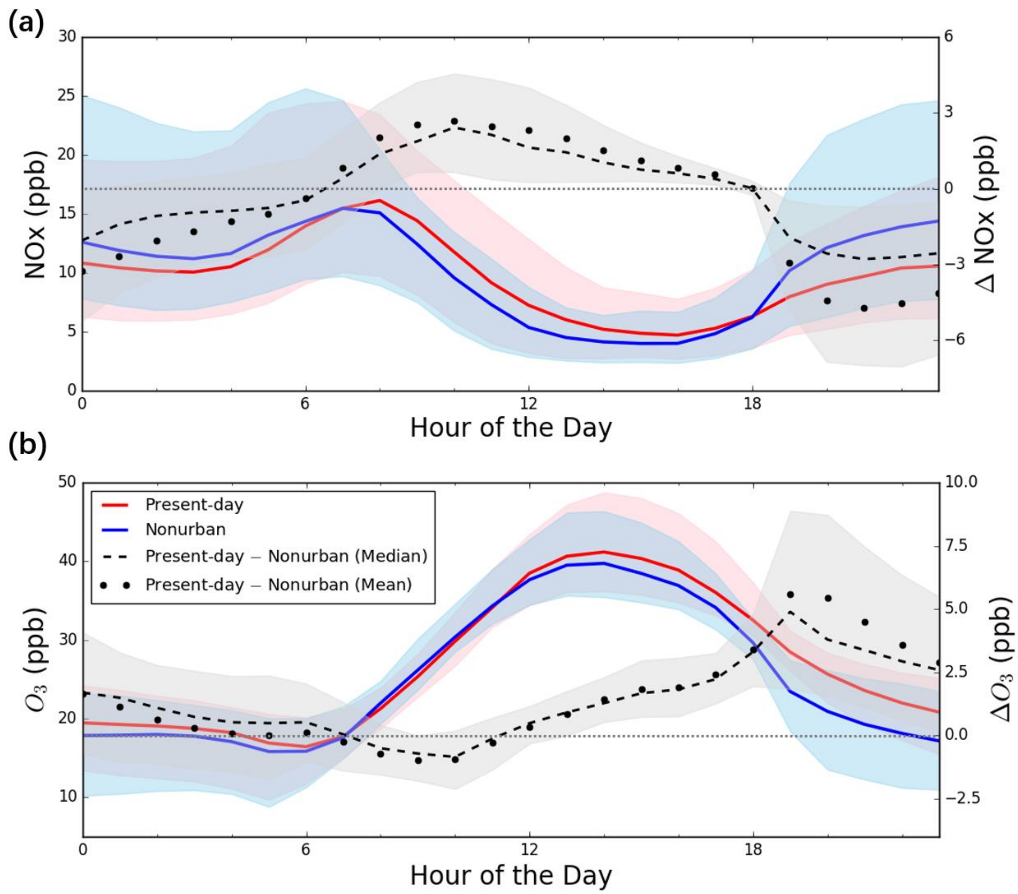
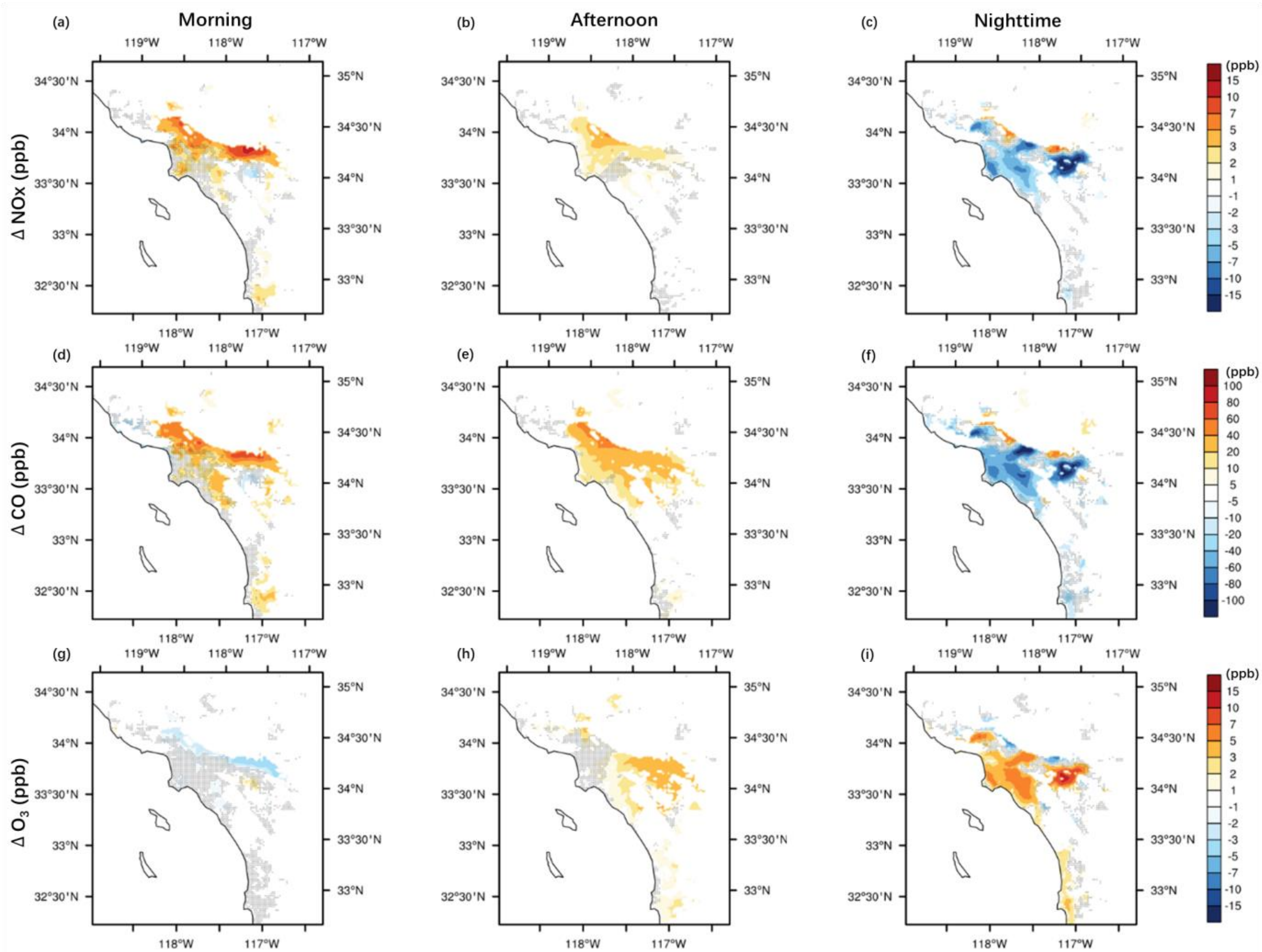


Figure 6. Diurnal cycles for present-day (red), nonurban (blue), and present-day – nonurban (black) for (a) NOx (ppb) and (b) O_3 concentrations (ppb). Values are obtained by averaging over urban grid cells and the entire simulation period for each hour of day. The solid and dashed curves give the median values, while the shaded bands show 25th and 75th percentiles. Dots indicate mean values for differences between Present-day and nonurban. The horizontal dotted line in light gray shows $\Delta = 0$ as an indicator of positive or negative change by land surface changes via urbanization.



(a)
Morning
(b)
Afternoon
(c)
Nighttime
(d)
(e)
(f)
(g)
(h)
(i)
Δ NOx (ppb)
Δ CO (ppb)
Δ O3 (ppb)
(ppb)

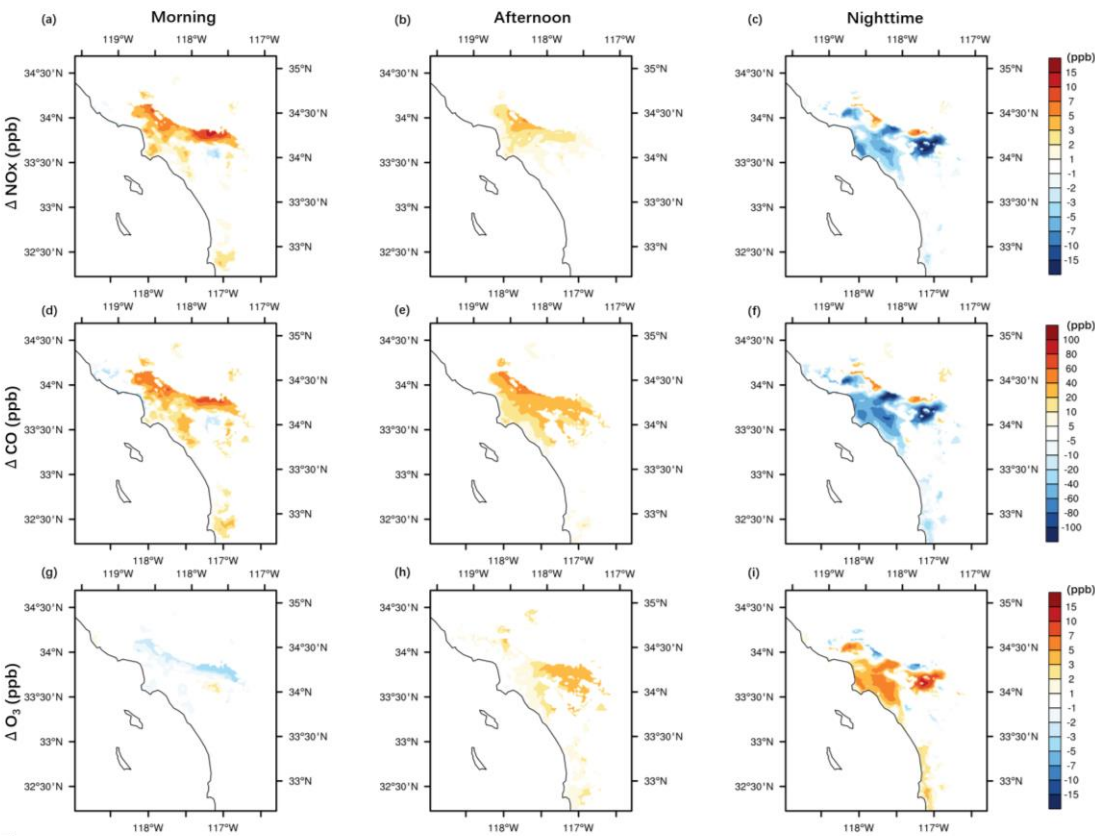


Figure 7. Spatial patterns in differences (Present-day – nonurban) of temporally averaged values during morning, afternoon and nighttime for (a,b,c) NOx, (d,e,f) CO, and (g,h,i) O_3 concentrations. Morning is defined as 7 PST to 12 PST, afternoon as 12 PST to 19 PST, and nighttime as 19 PST to 7 PST. Black dots indicate grid cells where changes are not significantly different from zero at 95% confidence level using the paired Student's t-test with n=7 days.

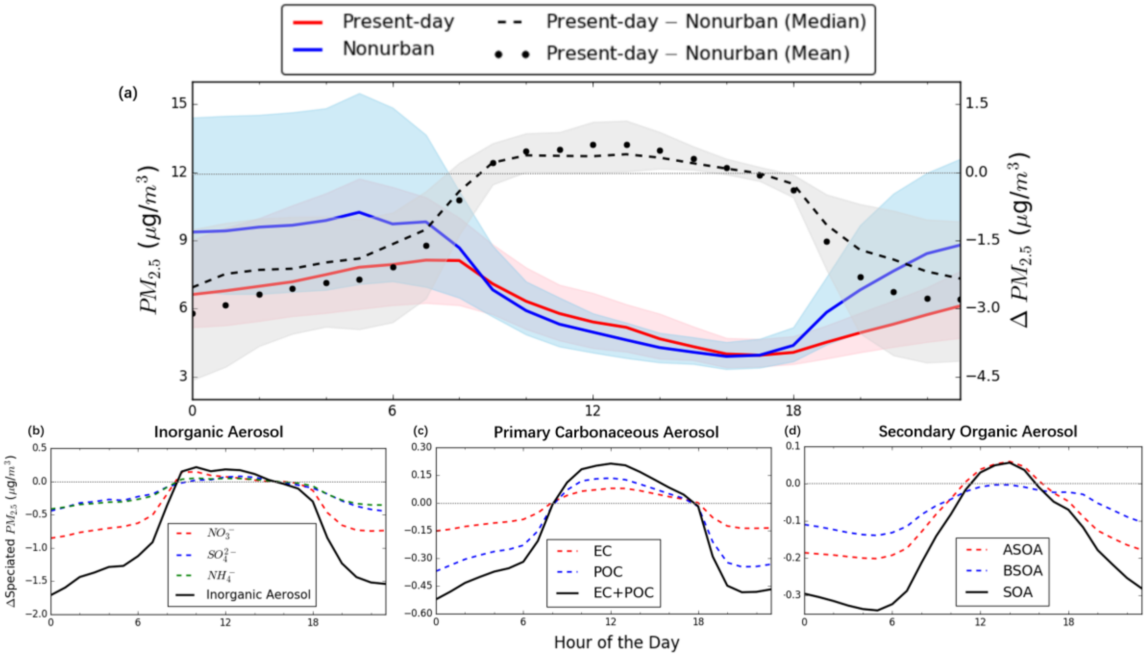
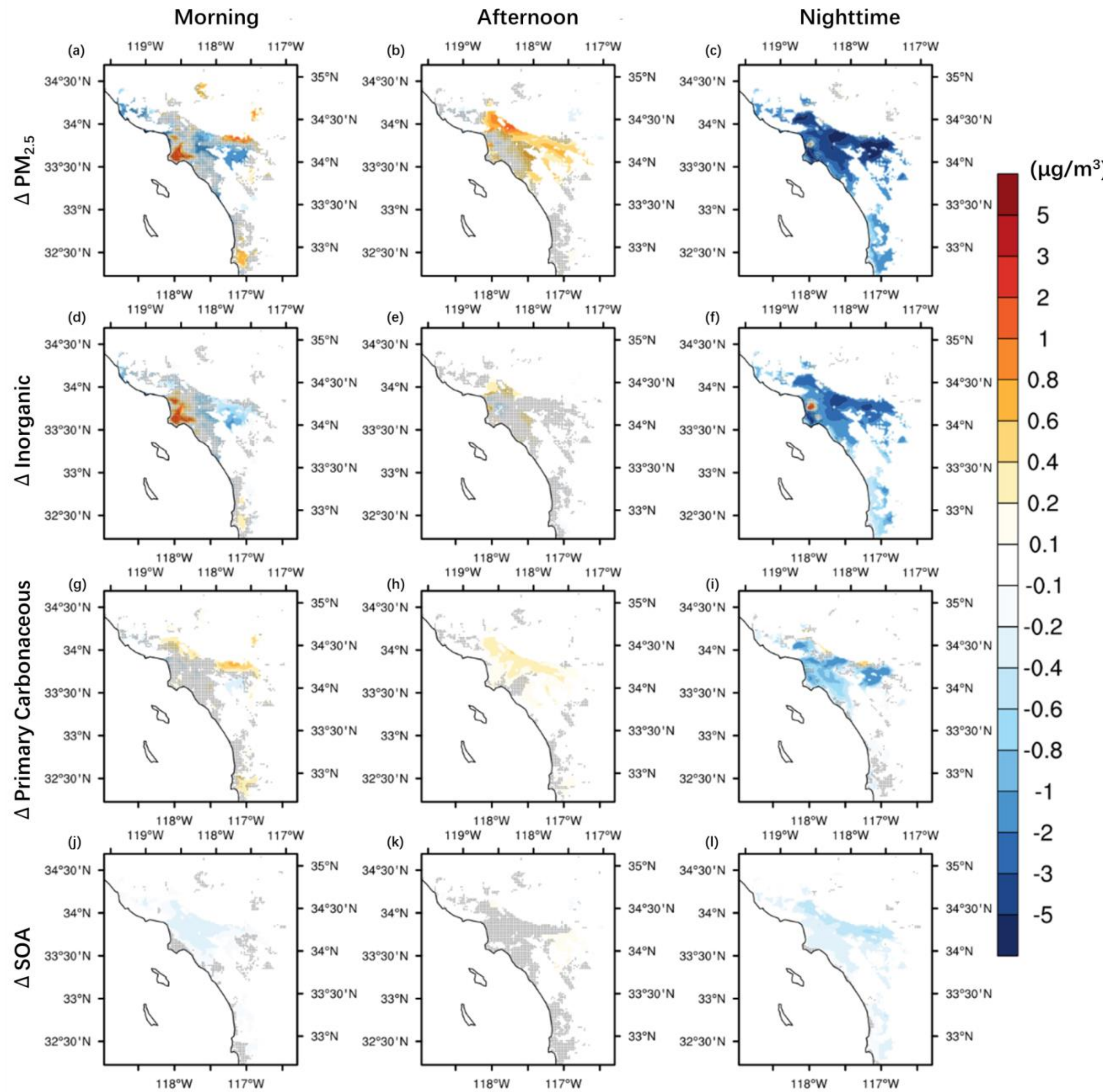


Figure 8. Diurnal cycles for spatially averaged $PM_{2.5}$ concentrations. Panel (a) shows Present-day, nonurban, and present-day – nonurban for total $PM_{2.5}$ (excluding sea salt and dust). The lower row shows differences (Present-day – nonurban) in speciated $PM_{2.5}$ including (b) inorganic aerosols (NO_3^-, SO_4^{2-}, NH_4^+), (c) primary carbonaceous aerosols (EC, POC), and (d) secondary organic aerosols (ASOA, BSOA). The horizontal dotted line in light grey is shown for $\Delta = 0$ as an indicator of positive or negative change by urbanization.


Morning
Afternoon
Nighttime
Δ PM2.5
Δ Inorganic
Δ Primary Carbonaceous
Δ SOA
(a)
(b)
(c)
(d)
(e)
(f)
(g)
(h)
(i)
(j)
(k)
(l)
(μg/m³)
5
3
2
1
0.8
0.6
0.4
0.2
0.1
-0.1
-0.2
-0.4
-0.6
-0.8
-1
-2
-3
-5

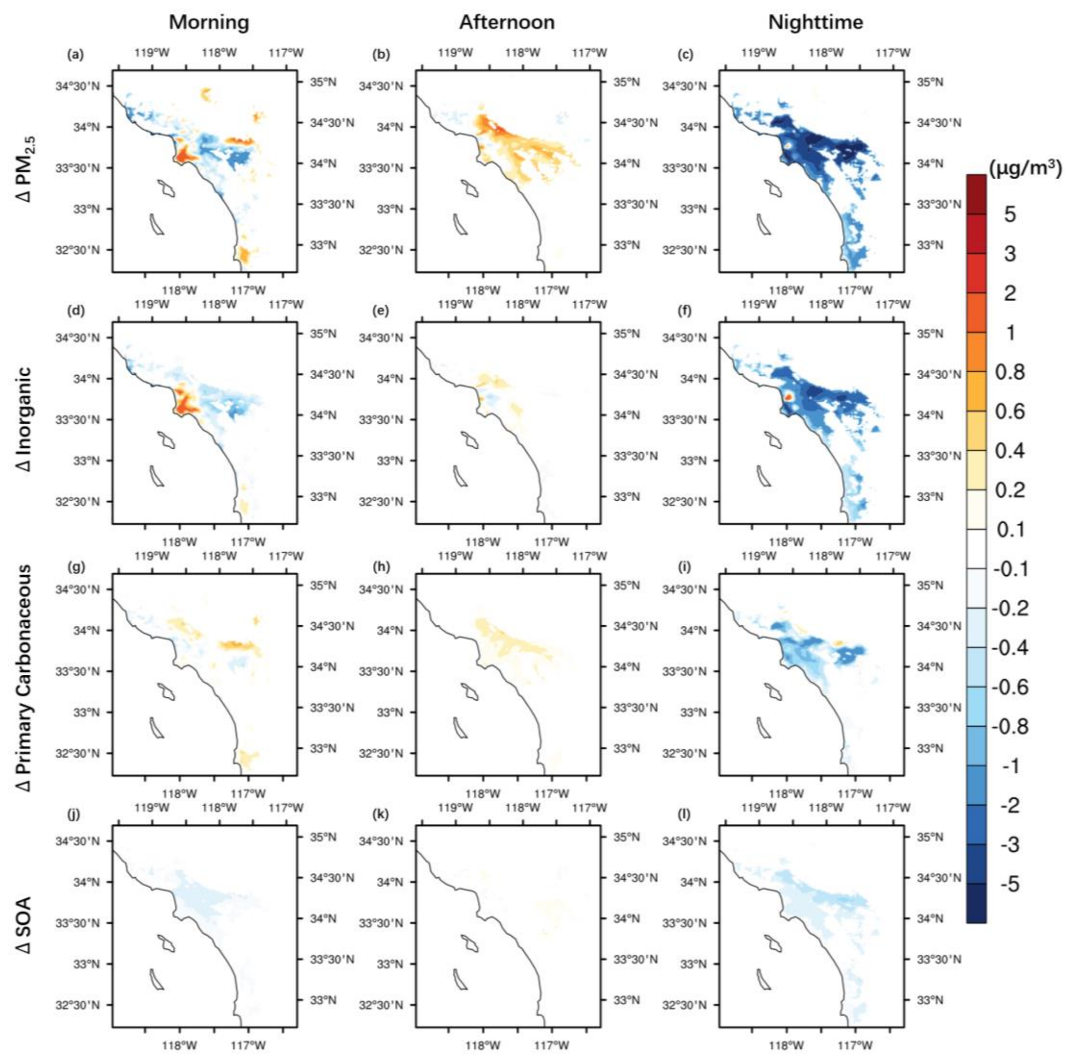


Figure 9. Spatial patterns in differences (Present-day – nonurban) of temporally averaged values during morning, afternoon, and nighttime for $PM_{2.5}$. Panels (a)–(c) show total $PM_{2.5}$; (d)–(f) inorganic aerosol; (g)–(i) primary carbonaceous aerosol; and (j)–(l) secondary organic aerosol. Morning is defined as 7 PST to 12 PST, afternoon as 12 PST to 19 PST, and nighttime as 19 PST to 7 PST. Black dots indicate grid cells where changes are not significantly different from zero at 95% confidence level using the paired Student's t-test with n=7 days.

Table 1. Summary statistics (mean bias (MB), normalized mean bias (NMB), mean error (ME), and root mean square error (RMSE)) for model evaluation, which compares simulated hourly near-surface air temperature (T2), hourly O_3 and daily $PM_{2.5}$ concentrations to observations.

Variable	N [a]	Mean		MB [b]	NMB [c]	ME [d]	RMSE [e]
		Observations	Simulations				
T2	1944	293.0 K	292.0 K	-1.0 K	-0.3%	1.9 K	2.2 K
O_3	5171	38.7 ppb	30.0 ppb	-8.7 ppb	-22%	11.8 ppb	14.6 ppb
$PM_{2.5}$	81	12.9 $\mu g/m^3$	9.2 $\mu g/m^3$	-4.0 $\mu g/m^3$	-31%	6.2 $\mu g/m^3$	9.5 $\mu g/m^3$

[a] Total number of data points comparing modeled versus observed values across all measurement station locations over the simulation period

[b] $\mathrm{MB} = \frac{1}{\mathrm{N}}\sum(mol_i - obs_i)$

[c] $\mathrm{NMB} = \frac{\sum(mol_i - obs_i)}{\sum obs_i}$

[d] $\mathrm{ME} = \frac{1}{\mathrm{N}}\sum|mol_i - obs_i|$

[e] $\mathrm{RMSE} = [\frac{1}{\mathrm{N}}\sum(mol_i - obs_i)^2]^{\frac{1}{2}}$