Abbreviations that will be used in response:
UHI: urban heat island
UHII: urban heat island intensity
UHIC: urban heat island circulation
BL: boundary layer
LFT: lower free troposphere

Reviewer 2

Aerosol pollution is of great concern in many megacities all over the world. It is meaningful to study how urbanization affects local meteorology and air pollution. By using WRF-CMAQ model, this manuscript assessed the effects of UHI on the distribution and formation of aerosols in urban atmosphere. The authors separated the impacts of UHI circulation, temperature and humidity, and quantified the contributions of them on inorganic aerosols in the lower free troposphere. Generally, this manuscript is well organized. The findings are very interesting, the discussion is of scientific meaning, and the results can improve the understanding of the formation of inorganic aerosols in urban areas. This manuscript could be considered for potential publication after the following minor revisions.

1) In 2.2, it is better to show the results of the comprehensive model validation for meteorological parameters, such as temperature, relative humidity and wind speed.

Response: Thank you for this suggestion. The model evaluation in terms of temperature, relative humidity, and wind speed has now been added in section 2.2. The time series of hourly ground-level air temperature, relative humidity, and wind speed from both the CTL simulation and observations at the 10 Hangzhou Meteorological Bureau sites are compared in Fig. 2. The correlation coefficient, NMB, and NME between observations and CTL simulations at the 10 meteorological observation sites are shown in Fig. 3a. The IOA, MFB, and MFE for the three meteorological parameters are shown in Fig. S1a). Details of the description of model evaluation are given in section 2.2. In general, the model performed well in the simulation of both meteorological parameters and $PM_{2.5}$ concentrations, as shown in the following Figures (added to the text).

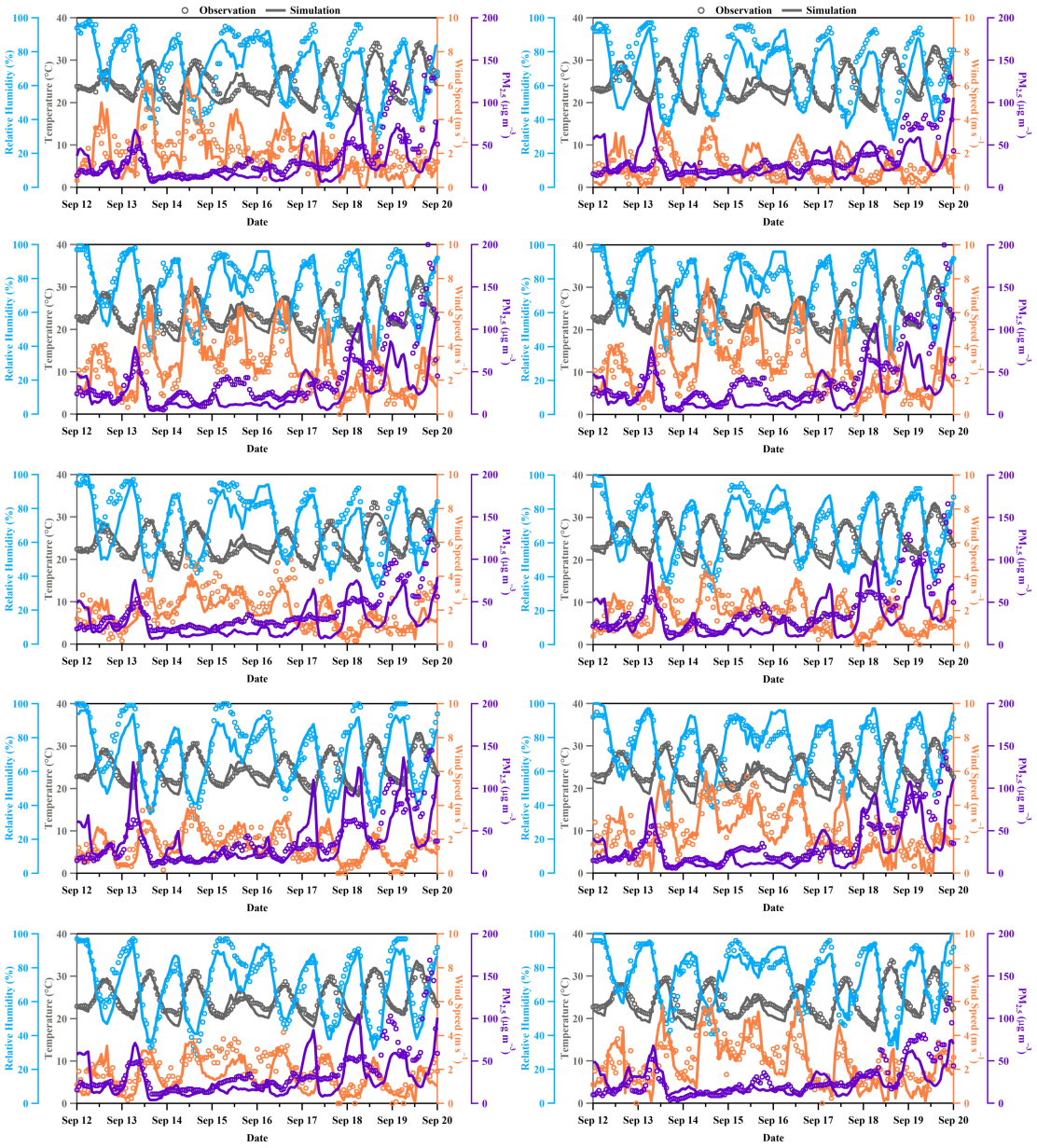

Figure 2. Time series of simulated and observed air temperature, relative humidity, wind speed, and $PM_{2.5}$ concentration from 00:00 LT September 12 to 00:00 LT September 20, 2017 at 10 meteorological and 10 $PM_{2.5}$ monitoring sites.

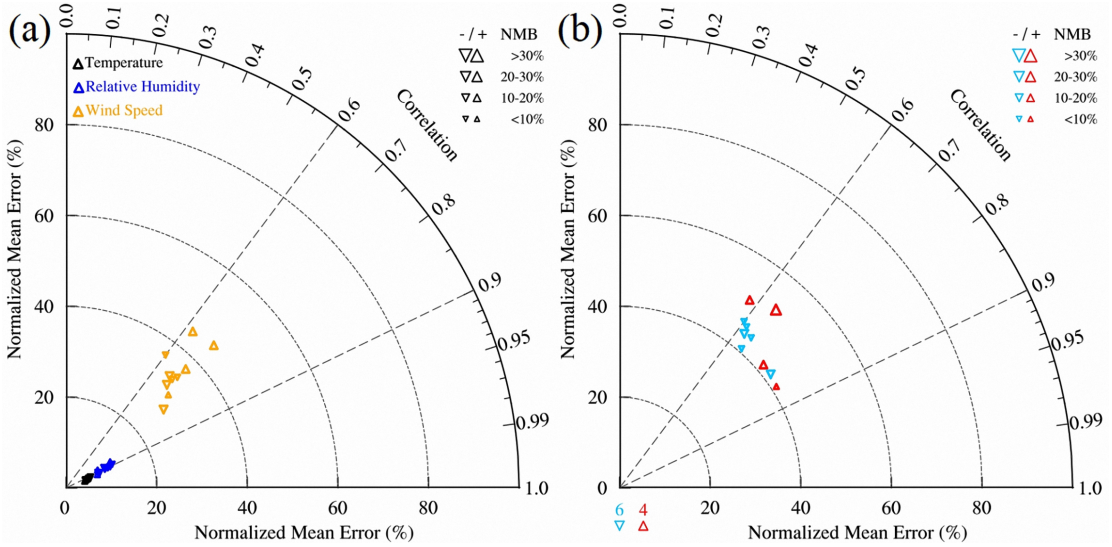

Figure 3. Taylor diagram of correlation coefficient, normalized mean bias (NMB) and normalized mean error (NME) between simulated (CTL experiment) and observed (a) air temperature, relative humidity, and wind speed at 10 sites in Hangzhou (white stars in Fig. 1b); and (b) $PM_{2.5}$ concentrations at 10 sites in Hangzhou (yellow filled circles in Fig. 1b).

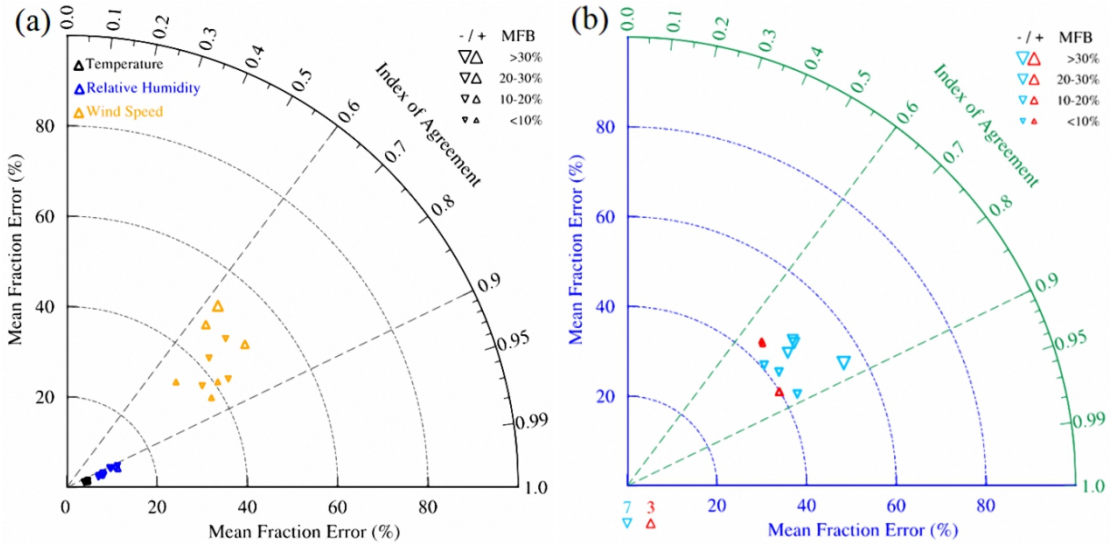

Figure S1. Taylor diagram of index of agreement, mean fraction bias (MFB) and mean fraction error (MFE) between simulated (CTL experiment) and observed (a) air temperature, relative humidity, and wind speed at 10 sites in Hangzhou (white stars in Fig. 1b); and (b) $PM_{2.5}$ concentrations at 10 sites in Hangzhou (yellow filled circles in Fig. 1b).

2) Line 145: "The NMB, NME, MFB, and MFE…" should be "The NMB, NME, IOA, MFB, and MFE…".

Response: Yes, you are right, thank you for catching this omission. We added the "IOA" here now.

3) In 3.2, the process analysis technique was used, but it was not introduced in methodology. It is better to briefly describe it in section 2.

Response: Thank you for your suggestion. We added the description of the process analysis technique to line 129 in section 2 as follows.

The process analysis technique (Gipson, 1999), which can determine the contributions of the physical and chemical processes to atmospheric species, was implemented in the CMAQ simulations. The processes discussed in this study include horizontal advection (HADV), vertical advection (ZADV), vertical diffusion (VDIF), and aerosol (AERO) processes.

4) Lines 337-343: The authors should demonstrate that the conclusions (about the impact of UHI on inorganic aerosol in the lower free troposphere) are universal rather than just in a specific case. Although other 7 UHI cases in the 10-day simulation period were mentioned in the last paragraph, the discussion was not clear. More details about the 7 UHI cases should be provided in section 3.

Response: Thank you for your valuable suggestion. Details about the impact of UHI on aerosol ant its inorganic compositions during the 8-day simulation period (exclude 2 spin-up days) are listed in Table S2. From Table S2, we can find that the impact of the UHI on inorganic aerosols, particularly on nitrate and ammonium aerosols, is evident in all 8 UHI cases. The following detailed discussion has been added to lines 380-392.

Lines 380-392: In the 8-day simulation period, the UHI effects decreased the $PM_{2.5}$ concentrations in the BL by 6% to 33%, and increased the $PM_{2.5}$ concentrations in the LFT by 1% to 19% (Table 2). The simulations show that the impact of the UHI on aerosol is highly dependent on the UHI effect's intensity and background wind speed. The impact of the UHI on $PM_{2.5}$ concentrations in both the BL and the LFT is larger on 18 and 19 September (Table 2), when the UHII and UHIC are stronger and the BL wind speed is lower, than on other days (Table S2). On 14 September the BL wind speed was the highest and on 15 September the UHII was the weakest (Table S2), resulting in a substantially smaller UHI impact on the $PM_{2.5}$ concentrations on these days (Table 2). The UHIC effect consistently plays a dominant role (72% to 93%) in affecting the vertical distribution of $PM_{2.5}$ in all UHI cases (Table 2). During the 8-day experiment, the UHI effect increased the mean concentrations of $PM_{2.5}$, nitrate, ammonium, and sulfate in the LFT by 7%, 20%, 8%, and 3%, respectively (Table 2). This suggests that ammonium nitrate aerosols increased considerably in the LFT due to the UHI effects.

5) Please check the English and avoid the typo errors.

Response: Thank you for your suggestions. We double-checked the English spelling and made sure there were no typo errors.