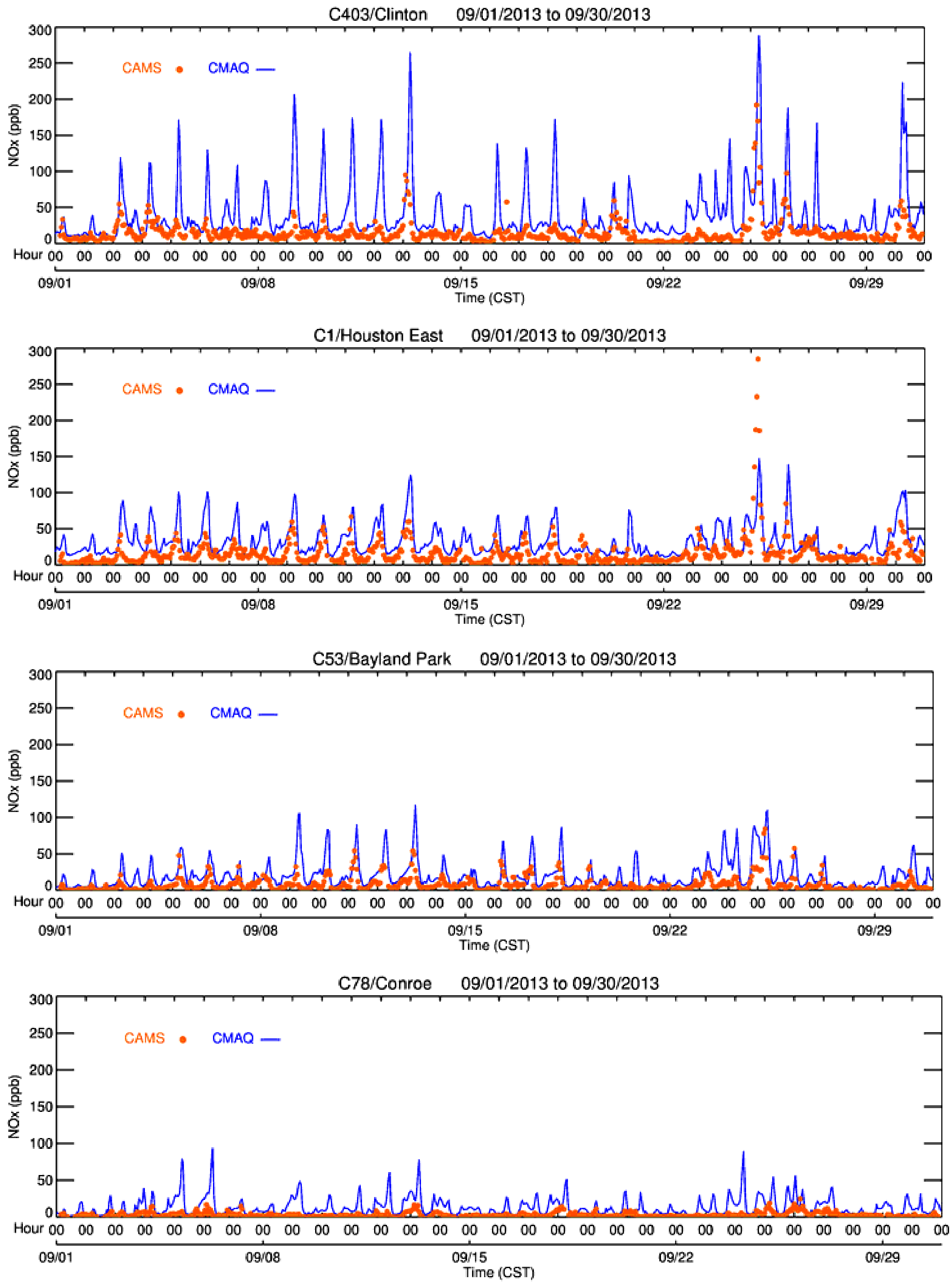
Respond to the first reviewer 1

This paper investigated the applicability of the satellite observed column density of NOx as a constraint for the bottom-up emission inventory of NOx (NEI2005) to improving CMAQ simulations of NOx and O3 concentrations in surface air over the United States. The method is largely based on the previous ACP paper by the author [Choi et al.,2012], and at this time the author additionally focuses on the analysis of the observed and simulated concentrations of O3 in surface air over the two urban areas (e.g., LA and Houston) in order to validate the use of the satellite observation to perturb the bottom-up emissions in the model.

The merit of this study, just as in the previous literature on the top-down estimates of emissions, is to provide adjustment information of the bottom-up emission, which usually lags relative to the present or the period of interest. As mentioned by the author, the old NEI2005 inventory is still widely used although the NEI2008 is available to the public because the latter has not extensively been evaluated yet. This work is also based on the NEI2005 with a few modifications, which primarily update the point sources.

1. Therefore, I wonder how the adjusted NO_x emission based on the satellite observation compares with the latest emission inventory (NEI2008) in the contiguous US. Please discuss the difference if any or the consistent changes compared to the NEI2005.

   Thanks for sharing a good idea. For this study, we have focused on the modified NEI2005 in 12km CMAQ domain over the southern California and the southeastern Texas. We have our forecasting results from the Air Quality Forecasting system at UH (AQF-UH, http://spock.geosc.uh.edu) based on NEI2008 in the 4km CMAQ domain over the Southeast Texas for the DISCOVER-AQ Houston aircraft campaign (September, 2013). From the comparisons between the 4km CMAQ Air Quality Forecasting results and the CAMS urban site observation data, we also found that the 4km CMAQ simulated NO_x concentrations with 2008 NEI are shown to be larger than the observed ones from the CAMS measurement sites. The other referee recommended that the manuscript needs to be shorten. Thus, I briefly added this discussion in the section of Discussion and put the comparisons as Appendix 1 in the revised manuscript. The short discussion is added by saying, "More interestingly, the high simulated NO_x biases are still shown in the comparison of the NO_x concentrations from the CMAQ including NEI2008 from Air Quality Forecasting system at UH (AQF-UH) and the corresponding observations from the CAMS sites over Southeast Texas for the DISCOVER-AQ Houston campaign (September of 2013) (Appendix 1), but they are not shown to be significant as much as in those of CMAQ including the modified NEI2005 in this study. The detailed study needs to be followed to examine how the biases of NO_x emissions found in this study are changed in the modeling study with NEI2008 using same resolution and same time simulations".



Appendix 1. Surface NO_x concentrations at the CAMS stations over Texas (pink color) and corresponding 4km CMAQ simulations with NEI2008 from Air Quality Forecasting system at UH (http://spock.geosc.uh.edu) (blue color). The CAMS sites are from Clinton site, Houston East Site, Bayland Park site, and Conroe site.

2. The model with the top-down emission supports the validity of the adjustment of the bottom-up emission based on the satellite observations in the context of NOx concentration in surface air but appears to be inconsistent with the observed O3 concentrations especially for Houston. Why? This issue needs to be investigated more.

   Thanks for initiating a good point. As we described in the manuscript, the adjusted NO_x mitigated the NO_x and O_3 biases for Los Angeles, but the adjusted NO_x emissions only mitigated the NO_x discrepancies for Houston. The adjusted NO_x emissions are useful for mitigating the high O_3 biases near and around the city of Houston, but as the review indicated, the high O_3 biases became worsen over the central Houston. One message from this study is that the better simulated NO_x does not mitigate O_3 biases over the central Houston. Also, please note that the main scope of this study is not to find the main cause of high biased O_3 over the central Houston, but to investigate the impact of satellite-adjusted NO_x emissions on simulated NO_x and O_3 biases in the urban and outflow areas (as the title indicated). In addition to this, to see the high O_3 bias issue over Houston, I think that we should see NO_x and VOC at the same time, which will make this manuscript longer. The other referee recommends to make this manuscript shorten.

3. Following up the comment above, are there any issues of the top-down estimates due to the uncertainties of the satellite measurements such as AMF, cloud contaminations, sampling mismatch between the model and satellite, etc?

   Thanks for initiating the point. That's why we only used the data including monthly averaged NO_2 column of $>10\times10^{15}$ molecules cm^{-2} which are more accurate (or above the sensitivity of satellite sensor). I added this sentence in Section 4.2 of the revised manuscript, "To filter out the remote region data (with low sensitivity of the satellite sensor), the monthly averaged GOME-2 and CMAQ NO_2 column were estimated and only the regions showing NO_2 column densities $>1\times10^{15}$ molecules cm^{-2} were considered to adjust the emission inventory as in our previous study (e.g., Choi et al., 2012)".

   In this study, some data were filtered out with a cloud fraction of > 40%. We added this sentence in Section 3.1, "Some data were filtered out with a cloud fraction of >40%".

   Through the contents, I addressed the uncertainties regarding on remote sensing measurements, in order not to mislead the readers.

   For example, I addressed this issue in the introduction, by saying "The main purpose of this study is not to obtain an accurate emissions inventory or estimate the absolute uncertainty of the emissions inventory, but instead to perform an evaluation of the relative uncertainties of both the NO_x emissions inventory and adjusted NO_x emissions inventories using remote sensing in the two urban areas that showed large discrepancies between simulated surface O_3 and corresponding observations."

   In the discussion section, I also addressed this issue again, "The direct satellite-adjusting method in this study gave general success in mitigating the discrepancies of model-simulated surface NO_x concentrations compared with in-situ measurements, but further research is needed to address some of remaining issues. First, the assumption that remote-sensing NO_2 columns are closer to actual true values compared with model-simulated NO_2 columns was not perfectly met by the results. Thus, ideally, in order to get accurate emission inventories, we need to estimate uncertainties of remote-sensing NO_2 column and model simulated NO_2 column/NO_x emission inventories and the uncertainties for the application of data assimilation

approach (e.g., Napelenok et al., 2008; Chai et al., 2009; Zhao et al., 2009). Second, emissions were adjusted using morning time satellite NO_2 column data (e.g., GOME-2) and the resulting emission inventory could miss its diurnal cycle. In a following study, we will adjust the diurnal cycles of emissions using two different remote sensing data from GOME-2 (morning time) and OMI (afternoon). Some other uncertainties regarding the use of NO_2 columns as a proxy for NO_x concentrations/emissions over the surface were described in detail in the previous study (Choi et al., 2012)."

4. Are there any other regions, showing similar responses of the model such as improved agreements for NOx but degradations for O3? It might be useful to look at other area with high VOC emissions (e.g., southeast US) and examine the sensitivity of the model with the respect to the NOx perturbations.

   Thanks for sharing a good idea. It is a good suggestion, but I think that the NO_x and VOC should be simultaneously investigated over the region. The specific study over the region could be a single story by itself (actually, one of the graduate students in my group is working on the high O_3 bias issue for the SENEX campaign over Southeast US (June-July of 2013)). Also, the southern California and the southeastern Texas have been investigated as nonattainment areas. The simulated high O_3 biases (model – observation) are significant over Southeast US, but the observed daytime O_3 concentrations over Southeast US are relatively lower than those of two other regions in this study (e.g., South California and Southeast Texas). I addressed this point and why I chose two regions in the introduction, "As we mentioned above, among these cities, Los Angeles and Houston have been investigated by previous NO_2 remote sensing studies (e.g., Kim et al., 2009; 2011; Eder et al., 2009) because of their characteristic as an O_3 nonattainment area and a large discrepancy area of O_3 compared with in-situ measurements".