Response to Reviewer #1

The authors would like to thank Reviewer 1 for the thoughtful comments and constructive suggestions about investigating further into the main uncertainties in the inversion process and adjusting the constraints on the inversion.

The discrepancies between the base model and satellite retrieved NO_2 columns arise primarily because of the lower modeled than observed NO_2 in rural areas. As noted in the review by Streets et al. (2013), wider spreads between urban and rural NO_2 in models than in satellite observations have been reported in other studies. We note in Section 3.3 our methods of alleviating such discrepancies by adjusting the $OH+NO_2$ reaction rate and correcting CAMx's low bias for upper tropospheric NO_2 based on the findings of earlier studies. However, additional model shortcomings likely remain (ENVIRON, 2013). In addition, while the NASA OMI NO_2 product, version 2.1, used in this study is the latest available retrieval, it does have some errors that vary spatially and temporally (Lamsal et al., 2014) and retrieval algorithms continue to be refined. It is beyond the scope of this study to fully diagnose or correct all the causes of uncertainties and discrepancies, and to quantify the possible errors in the retrieval processes over our modeling domain.

We have analyzed the influence on the region-based inversion caused by each of the adjustments we made either to the OMI retrieval product or the CAMx a priori simulations (Table R1). It shows that, in this case, missing emission sources (lightning, aviation and soil NO_x emissions) had the largest effect on the inversion results, especially in rural areas. Using the updated OMI product (with higher resolution) had the second largest effect on the inversion results. Since the new OMI NO_2 narrowed the urban-rural spread, the adjustments over most urban areas and rural areas decrease. The adjustments made in the CAMx model such as decreasing the $OH+NO_2$ reaction rate and adding an artificial NO_2 layer in the upper troposphere had smaller effects on the inversion results compared to the other changes (Table R1).

The seven inversion regions, five urban regions encompassed by two large rural regions, were carefully designed using sensitivity simulations to ensure NO_x emissions in each inversion region is mostly responsible for its NO_2 concentrations (Tang et al., 2013). In addition, the five urban regions were chosen to correspond to the urban ozone control regions that are relevant for regulatory attainment and emission control efforts in Texas SIP. The number of source categories is limited by the categorization of emissions in the TCEQ emission inventory. Visual inspection and pseudo-data testing of the categorized emissions were conducted to ensure that the source categories had sufficiently distinct spatial patterns to enable the Kalman filter to distinguish among the sources.

Table R1. Scaling factors for NO_x emissions in each region under alternate inversion cases.

Region-based inversion					
Emission region	Scaling factor (unitless)				
	Missing emission sources[a]	W/ additional emission sources[b]	Using updated OMI product[c]	Decreasing $OH+NO_2$ rate[d]	Adding a 40ppt layer[e]
HGB	1.31	1.03	1.21	1.18	1.11
DFW	1.32	1.14	1.04	0.98	0.97
BPA	1.90	1.75	1.70	1.72	1.49
NE Texas	1.40	0.56	1.12	1.20	1.10
Austin and San Antonio	1.90	1.70	1.21	1.24	1.15
N rural	2.88	1.98	1.45	1.48	1.24
S rural	3.84	1.72	1.25	1.15	0.98

a. Inversion conducted based on OMI v.2.1 and a priori simulation using base case NO_x emissions; adopted from Tang et al. (2013).

b. Inversion conducted based on OMI v.2.1 and a priori simulation using base case with added lightning and aviation and doubled soil NO_x emissions; adopted from Tang et al. (2013).

c. Inversion conducted based on updated OMI v.2.1 (using an a priori NO_2 profile generated from nested GEOS-Chem simulations with a 2005 emission inventory) and a priori simulation with NO_x emissions from b.

d. Inversion conducted based on updated OMI v.2.1 and a priori simulation with NO_x emissions from b and decreased $OH+NO_2$ reaction rate.

e. Inversion conducted based on updated OMI v.2.1 and a priori simulation from d with an added 40ppt layer in the upper troposphere.

Following are our responses to each of the reviewer's general and specific comments (shown in italics):

General comments:

1. *Both region-based and sector-based NOx emission adjustments were made in the paper, but only "sector-based" approach is mentioned in the abstract.*

A sentence "The region-based DKF inversion suggests increasing NO_x emissions in most regions, deteriorating the model performance in predicting ground-level NO_2 and O_3" was added to the abstract.

2. *In the Introduction section, more references should be added when discussing "studies using satellite NO2 measurements to create top-down NOx emissions for atmospheric modeling".*

References to Martin et al., (2003); Müller and Stavrakou, (2005); Jaeglé et al., (2005); Lin et al., (2010); Konovalov et al., (2006, 2008); Napelenok et al., (2008); Kurokawa et al., (2009); Zhao and Wang, (2009); Chai et al., (2009); and Zyrichidou et al., (2015) were added to the Introduction and Reference sections.

Specific comments:

1. *Please check equation 5 (last term).*

The last term is correct, because we need to consider the difference between prediction and observation at each iteration. The term "Sx" reflects adjustments after each iteration.

2. *Page 24491, line 23, "while it adds 50% ...": Should it be 49% ?*

We have changed the number to 49% in the sentence.

3. *Page 24493, line 16, "0.09 reduction in both modeled NMB ...": Is it 0.09 reduction in NMB? Table 5 shows that it is from 0.09 to -0.02.*

We have changed the sentence to "The model performance is also improved compared against P-3 measurements. For NO_2, NMB is reduced from 0.09 to -0.02, and NME is

reduced by 0.09. For NO_y, NMB is reduced by 0.16 and NME is reduced by 0.11 (Table 5).”

4. *Table 3: Are the "overall" evaluation statistics based on the data from all regions listed above them? Then, the "overall" numbers do not seem to be right. The values should fall between the minimums and the maximums of the separate regions. For instance, in the last column, the NMEs are all above or equal to 0.30, but the overall NME is shown as 0.16.*

We double checked the numbers, and they are correct. The “overall” statistics are calculated based on data from all inversion regions, including two large rural regions that encompass the five urban regions presented in the tables. The OMI observations cover each grid cell, and thus the two large rural regions influence the overall statistics in Table 3. For Tables 4 and 6, there are few observation sites outside the five urban regions, making the overall values more similar to the urban values.

References

Lamsal, L. N., Krotkov, N. A., Celarier, E. A., Swartz, W. H., Pickering, K. E., Bucsela, E. J., Martin, R. V., Philip, S., Irie, H., Cede, A., Herman, J., Weinheimer, A., Szykman, J. J., and Knepp, T. N.: Evaluation of OMI operational standard NO_2 column retrievals using in situ and surface-based NO2 observations. Atmos. Chem. Phys. Discuss., 14, 14519–14573, 2014.

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Streets, D. G., Canty, T., Carmichael, G. R., de Foy, B., Dickerson, R. R., Duncan, B. N., Edwards, D. P., Haynes, J. A., Henze, D. K., Houyoux, M. R., Jacob, D. J., Krotkov, N. A., Lamsal, L. N., Liu, Y., Lu, Z-F., Martin, R. V., Pfister, G. G., Pinder, R. W., Salawitch, R. J., and Wecht, K. J.: Emissions estimation from satellite retrievals: A review of current capability. Atmos. Environ., 77, 1011–1042, 2013.

Tang, W., Cohan, D. S., Lamsal, L.N., Xiao, X., and Zhou, W.: Inverse modeling of Texas NO_x emissions using space-based and ground-based NO_2 observations. Atmos. Chem. Phys., 13, 11005-11018, 2013.

Response to Reviewer #2

The authors would like to thank Reviewer 2 for the thoughtful comments and description of this paper as well written and as interesting to the regional air quality community.

Following are our responses to each of the reviewer's general and specific comments (shown in italics):

General comments:

1. *My concern with using GOES cloud fractions to adjust photolysis rates in the model is that it introduces an inconsistency with the modeled dynamics. Changing the cloud fraction directly affects the heat flux and therefore stability and the height of the boundary layer, both important drivers of ground level O3. I understand that it may take considerable effort to fully include satellite-observed cloud fractions in the chemistry and meteorological models. However, I think the authors should at least include a broader discussion of this topic and frame this analysis as a sensitivity study.*

We agree with the reviewer on this point. The model dynamic and aqueous phase chemistry haven't been adjusted by the GOES cloud fractions, and thus are inconsistent with the GOES-based photolysis rates. This work represents a sensitivity study of the impact of satellite-based photolysis rates but not a complete assimilation of satellite-based clouds. We have more fully discussed this limitation in the conclusion (page 24495, lines 13-15) by the sentences:
"The GOES-retrieved clouds applied here adjusted only the modeled photolysis rates, while modeled clouds continued to drive the dynamics and aqueous phase chemistry. This inconsistency in the placement of clouds is similar to the approach of a previous study (Pour-Biazar et al., 2007). Thus, this work demonstrates a sensitivity study of using satellite-derived photolysis rates on model performance rather than a full integration of satellite-observed clouds into all aspects of the model. Future work could extend the use of GOES-retrieved clouds to also correct model dynamics and aqueous phase chemistry and investigate their impacts on NO_x and O_3 modeling."

2. *The last sentence of the introduction states that the manuscript will also present inverse modeling of VOC emissions, but there is no mention of this in the methodology. Some results of VOC inversions are presented in the Conclusions and the reader is directed to supplementary information. If this analysis is to be presented as one of the main aims of the manuscript, I think that the methodology and results should appear earlier in the manuscript.*

The reason we studied VOC is that we want to see if the uncertainties in VOC emissions will significantly affect our NO_x inversion results. Since this is not the main aim of this paper and the findings are not significant, we have moved the description of VOC emissions part in the introduction section (page 24480, lines 7-20) into the supplementary material. We keep the last sentence regarding the VOC work in the introduction section (page 22481, lines 1-2) and point it directly to the supplement.

3. *The last sentence of the 2.5.1 states that the "the OMI averaging kernels are not applied here." I think this is misleading because it implies that the vertical sensitivity of the retrieval and dependence on the a-priori profile are ignored. This is in fact not the case, as is shown in the supplement, and I would urge the authors to reword this.*

We have changed the sentence in page 24486, lines 12-14 to "Since applying OMI averaging kernels (Eskes and Boersma, 2003) may introduce more uncertainties to the CAMx-derived NO_2 VCD in this case (Supplement, Sect. 1), the CAMx modeled NO_2 are compared to the OMI NO_2 directly." to avoid any confusion.

Specific comments:

1. *Page 24478 Line 13: The term 'ozone design values' is not common outside of U.S. air quality policy circles. Thus a typical reader may not understand the implications of ozone design values above the NAAQS standard. It might be good here to give a brief definition of the term, or phrase this in a different way.*

We have removed the term "ozone design value" and rephrased the sentence in page 24478, lines 11-17 to "First and foremost, the Houston-Galveston-Brazoria (HGB) region and the Dallas-Fort Worth (DFW) region exceed the 2008 O_3 National Ambient Air Quality Standard (NAAQS) of 75 ppb and thus are both classified by US

Environmental Protection Agency (US EPA) as O_3 non-attainment areas. Next, Beaumont-Port Arthur (BPA), Northeast Texas (NE Texas), and Austin and San Antonio regions require attention for closely approaching that standard (Gonzales and Williamson, 2011)."

2. *I think it's misleading to say that GOES measures cloud fraction. The 12 km cloud fraction is derived from the fraction of GOES subpixels that are deemed cloudy. This should at least be made more clear.*

We agree with reviewer on this point. The cloud fraction in the 12km model grid was integrated from GOES sub-pixels. The terms we use in our paper are "GOES-retrieved clouds" and "GOES-derived photolysis rates". We have changed the sentence in page 24483, lines 11-12 to "In this study, hourly GOES observations with integrated 12km cloud properties from sub-pixels have been used." to avoid any confusion.

References

Eskes, H. J. and Boersma, K. F.: Averaging kernels for DOAS total column satellite retrievals. Atmos. Chem. Phys., 3, 1285–1291, 2003.

Gonzales, M. and Williamson, W.: Updates on the National Ambient Air Quality Standards and the State Implementation Plans for Texas, presented in TCEQ Trade Fair, Austin, TX, 4 May, 2011.

Pour-Biazar, A., McNider, R. T., Roselle, S. J., Suggs, R., Jedlovec, G., Byun, D. W., Kim, S., Lin, C. J., Ho, T. C., Haines, S., Dornblaser, B., and Cameron, R.: Correcting photolysis rates on the basis of satellite observed clouds. J. Geophys. Res., 112, D10302, doi: 10.1029/2006JD007422, 2007.

List of changes in the revised manuscript:

1. A sentence “The region-based DKF inversion suggests increasing NO_x emissions in most regions, deteriorating the model performance in predicting ground-level NO_2 and O_3” was added to the abstract.
2. References to Martin et al., (2003); Müller and Stavrakou, (2005); Jaeglé et al., (2005); Lin et al., (2010); Konovalov et al., (2006, 2008); Napelenok et al., (2008); Kurokawa et al., (2009); Zhao and Wang, (2009); Chai et al., (2009); and Zyrichidou et al., (2015) were added to the Introduction (page 24479, line 15) and Reference sections.
3. The number of “50%” in Page 24491, line 23 was revised to “49%”.
4. The sentence in page 24493, lines 15-17 was revised to “The model performance is also improved compared against P-3 measurements. For NO_2, NMB is reduced from 0.09 to -0.02, and NME is reduced by 0.09. For NO_y, NMB is reduced by 0.16 and NME is reduced by 0.11 (Table 5).”
5. The sentences “The GOES-retrieved clouds applied here adjusted only the modeled photolysis rates, while modeled clouds continued to drive the dynamics and aqueous phase chemistry. This inconsistency in the placement of clouds is similar to the approach of a previous study (Pour-Biazar et al., 2007). Thus, this work demonstrates a sensitivity study of using satellite-derived photolysis rates on model performance rather than a full integration of satellite-observed clouds into all aspects of the model. Future work could extend the use of GOES-retrieved clouds to also correct model dynamics and aqueous phase chemistry and investigate their impacts on NO_x and O_3 modeling.” were added to Conclusion (page 24495, line 14).
6. The description of VOC emissions part in the introduction section (page 24480, lines 7-20) was moved to the supplement.
7. The sentence in page 24486, lines 12-14 was revised to “Since applying OMI averaging kernels (Eskes and Boersma, 2003) may introduce more uncertainties to the CAMx-derived NO_2 VCD in this case (Supplement, Sect. 1), the CAMx

modeled NO_2 are compared to the OMI NO_2 directly."

8. The sentences in page 24478, lines 11-17 were rephrased to "First and foremost, the Houston-Galveston-Brazoria (HGB) region and the Dallas-Fort Worth (DFW) region exceed the 2008 O_3 National Ambient Air Quality Standard (NAAQS) of 75 ppb and thus are both classified by US Environmental Protection Agency (US EPA) as O_3 non-attainment areas. Next, Beaumont-Port Arthur (BPA), Northeast Texas (NE Texas), and Austin and San Antonio regions require attention for closely approaching that standard (Gonzales and Williamson, 2011)."
9. The sentence in page 24483, lines 11-12 was revised to "In this study, hourly GOES observations with integrated 12km cloud properties from sub-pixels have been used."