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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Inverse modeling of Texas NO_x emissions using space-based and ground-based NO_2 observations" *by* W. Tang et al.

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The authors would like to thank the Reviewer 2 for the thoughtful and constructive comments. Following are responses to each of the reviewer's specific comments:

1. "The authors restricted their inversions to 2 time periods of 1 month during which high ozone concentrations were measured at either Dallas or Houston regions. Since the authors used OMI satellite data and continuous AQS ground site measurements, I don't see the reason for restricting the analysis to those 2 months specifically. I would extend the analysis to several months to minimize the uncertainty in the posteriors."

The main objective of this study is to test the applicability of two inverse methods to state-level regulatory attainment modeling. The two modeling episodes (May 31 to

July 2, 2006 and August 13 to September 15, 2006) chosen in this study are especially developed by TCEQ for the Texas SIP development. The inversions are conducted separately to develop independent scaling factors for episodes that began with slightly different meteorology and emissions estimates, the June episode having been prepared for the Dallas-Fort Worth SIP and the August-September episode for Houston-Galveston-Brazoria. Unfortunately, additional modeling episodes are unavailable and are beyond the scope of our current study, whose funding has concluded.

While extending the inversions to several months may reduce the uncertainty in posteriors to a certain extent, the discrepancies between satellite- and ground-based inversion results arise mainly from difference between the model and two types of observations that are persistent across the two episodes (Fig. R1).

The satellite measures total tropospheric NO2 column, while AQS ground monitors measure surface NO2. The comparisons between modeled NO2 using a priori NOx emissions and these two independent observations (Fig. R1) indicate that model overpredictes ground NO2 concentrations, but underpredictes total tropospheric NO2 column. Therefore, before performing inversions, we can anticipate that the adjustments based on these two independent observations will be heading different directions.

2. "In section 2.4.2, page 17487, line 14 and 15: the authors say that they assumed an uncertainty of 0.15 for the ground site measurements, 0.3 for the OMI data, and 2.0 for the prior inventory in the covariance matrices. An uncertainty should have a unit. I assume that those values are standard deviations relative to the mean value, basically uncertainties of 15%, 30% and 200% respectively. In an inversion, the covariance matrices play a key role in the posterior results. The prior uncertainty estimate used in this study, 200%, is in my opinion quite large based on the work done by TCEQ. The authors justified this assumption because of the assumption made by Napelenok et al. (2008) who assumed an uncertainty of 200% for the EPA NEI 1999 inventory. The authors cannot assume the same uncertainty for EPA NEI 1999 and TCEQ 2005 inventory without a minimum of justification. The authors tested the sensitivity of the

posterior on the covariance matrix of the prior using a test based on pseudodata in section 3.1. The underlying assumption in this kind of test is basically a perfect (unbiased) model. I am not convinced that this test can be used as a test for the sensitivity of the posterior on the assumptions made on the error in the observations (15 to 30%) and the prior (200%). The authors should use the OMI and AQS ground site observations for testing the sensitivity of the posterior on the assumptions made in the covariance matrices, and not pseudodata."

The uncertainty factors are unitless, and values of 0.15, 0.3 and 2.0 represent 15%, 30% and 200%. The uncertainty of ground measurement was estimated by U.S. EPA (2006), and the uncertainty of OMI was estimated by the NASA retrieval team (Bucsela et al., 2013). Direct estimation of the uncertainty of specific emission inventories is usually from an expert elicitation study (Hanna et al, 2001) and is rare, and is unavailable for the TCEQ emission inventory used in this study. Hence, the factor of 2.0 adopted here corresponds to the value assumed by the previous satellite NOx inversion by Napelenok et al. (2008). We agree with the reviewer that the factor of 2.0 may be unnecessarily large for the emission inventory considered here. However, testing of alternate values for this factor shows that the DKF inversion results are insensitive to the presumed value (Fig. R2). As suggested by the reviewer, we also performed sensitivity tests by varying uncertainty values in the error covariance matrices for both OMI-based and ground-based DKF inversions in the actual inversion case (Fig. R2). The results are very similar to the pseudodata test: the adjustments for the posteriors are insensitive to the emission error covariance matrix, and slightly responsive to the assumed observation errors. Results from Figure R2 will appear in the supplementary material.

3. "I am not sure to understand why the direct scaling inversion method is used in the paper. Is it to prove that this method shouldn't be used? If so, the authors should state that more clearly in the text and conclusion."

As detailed in our response to Reviewer 1, comment 2, we agree that Discrete Kalman

Filter is the more effective inversion method for this study, and thus move the direct scaling results to the supplementary material.

4. "The fact that the authors used monthly average measurements from OMI to drive the inversion is probably okay. However, since they restricted their analysis to 2 months, it is as if the inversion was driven by only 2 independent observations in each region, which is not a lot of observations to have good confidence in the posterior results. However, using 24h-average NO2 measurements from AQS ground sites to minimize the uncertainty from the influence of PBL height on NO2 concentration is not a good idea. At night, power plant plumes are lifted above ground because of the buoyancy of the stack when they are emitted. Since the PBL at night is stable, they don't mix down to the ground. Hence the AQS ground site measurements are not representative of the concentration higher in altitude. If I understand correctly, the power plants are emitted in the model at the surface, without buoyancy. Therefore, one can expect a bias in the model results at the AQS ground site locations at night. Therefore, using 24h-average AQS NO2 measurements in the inversion will probably make the inversion underestimates the surface NO2 emissions. This is probably the reason why the posterior from the inversion based on AQS ground sites is much lower than the inversion based on OMI data. The authors should use daily average NO2measurements instead. PBL uncertainties are not a problem in an inversion as long as the PBL height is not systematically biased."

One advantage of using satellite observations to perform inverse modeling rather than ground measurements is the rich spatial coverage of the dataset. However, satellite data on individual days can be noisy or unavailable due to cloud cover or other data screening. Therefore, in this study, the monthly averaged satellite measurements ensure complete spatial coverage in every modeling grid cell and more robust posteriors. The CAMx model has its shortcoming in simulating the PBL heights in the early morning and late afternoon (Kolling et al., 2013). Therefore, we chose 24-h averaged daily data to perform the ground-based inversion, trying to alleviate the problem from PBL simulations. The power plant in CAMx modeling are treated as elevated point source emissions with assigned stack parameters (height, diameter, exit velocity and temperature) for each stack used for calculating the plume rise (ENVIRON 2010). Hence, the power plant emissions in this study are not emitted at the surface and will not create a bias in the inversion.

We have also tried to use the daytime ground NO2 to constrain the NOx emissions. The data in a 6-h window, 9am to 2pm, was chosen to perform the inversion, but the scaling factors are still far below the OMI-based inversion (Table R1), and the inversion results don't show any improvements relative to the 24-h data-based inversion in simulating hourly AQS ground NO2 (Table R2) and P-3 aircraft measured NO2 (Table R3). In addition, the scaling factors up to a factor of two inconsistencies between the two episodes (Table R1). The use of 24-h averaged ground data is thus retained for the inversions in this paper.

5. "I don't see any validation of the meteorology in the paper. How good is the wind speed, wind direction, PBL height? The meteorology from MM5 must be evaluated. You can use for instance the aircraft measurements from TEXAQS 2006."

The reviewer raises an important point that we address by performing evaluations of modeled temperature, wind speed, wind direction, and PBL heights from the MM5 meteorology outputs for both episodes. The following results will appear in the supplementary material.

The MM5 modeled hourly temperature, wind speed, and wind direction are evaluated with measured data from 34 ground monitoring sites over the 12km CAMx domain for both modeling episodes (Table R4). The model simulates the temperature and wind speed well, showing that the mean bias error (MBE) is less than 0.5K and the root mean square error (RMSE) is less than 2K for the temperature, and the MBE and RMSE are around 2 m/s for the wind speed, which are similar to evaluation results from the study done by Kim et al. (2011). The simulated wind direction shows slightly

weak performance in terms of RMSE. However, according to TCEQ (2010 and 2011), the large discrepancy between observed and modeled wind directions mostly occurred with very low wind speed, thus, it only has slight influence on the CAMx modeling.

Modeled PBL heights are evaluated with available measurement data from two sites at Huntsville airport, Huntsville, TX (30.75°N, 95.58°W) and Jefferson airport, Port Arthur, TX (29.94°N, 94.00°W) for the June episode (Fig. 3), and from three sites at Huntsville airport, Jefferson airport, and LaPorte airport, La Porte, TX (29.67°N, 95.06°W) for the August-September episode (Fig. R4). The model overpredicts the daytime PBL height for the June episode with exceptions at 8am and 7pm in the Huntsville site and 8am at Jefferson site (Fig. R3), while the August-September episode tends to underpredict PBL heights at Huntsville and Jefferson sites, but it shows a good agreement with measurement at LaPorte site (Fig. R4). The overprediction of PBL heights in the June episode, but underprediction of that in the August-September episode is probably caused by using different vertical mixing schemes in the MM5 modeling. The June episode uses ACM2 vertical mixing scheme to simulate PBL heights which tends to have stronger mixing.

6. "In the conclusion, the authors say that DISCOVER-AQ flights in fall 2013 will help in reducing the discrepancy in the posteriors with spirals that will be performed over Houston. Why don't the authors use the NOAA flights during TEXAQS 2006 in September/October 2006 to drive the inversion then? At least the NO2 measurements during TEXAQS 2006 are of better quality than the EPA AQS ground sites. They could even use a longer lived species like NOy which will reduce the uncertainty of the inversion."

The DISCOVER-AQ campaign will have the advantage of conducting aircraft spiral measurements specifically aimed at representing vertical profiles.

The limited spatial and temporal coverage of P-3 aircraft data, which are available on only four days of the episode, make them an insufficient basis for a DKF inversion. The reviewer makes a valid point that longer-lived NOy could provide a valuable check of inversion results. We thus use observations of NOy from the P-3 aircraft to evaluate model performance in the original and inversion cases. The results are shown in the Table R5 and will be added to the main text.

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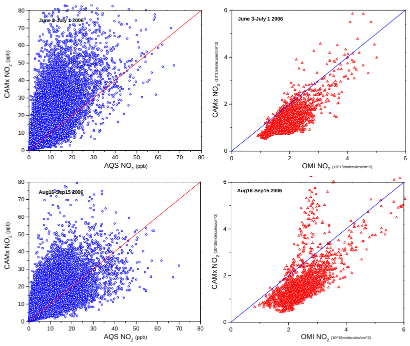


Fig. 1. Fig R1. Comparisons between CAMx NO2 with a priori NOx emissions and AQS ground NO2 observations (left), and OMI NO2 (right) in both June episode (top) and August-September episode (bottom).

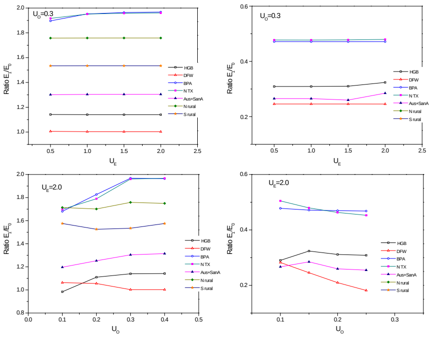


Fig. 2. Fig R2. Sensitivity analysis of Kalman filter inversion by changing emission uncertainties (top), and observation uncertainties (bottom) using OMI NO2 (left) and AQS ground NO2 (right).

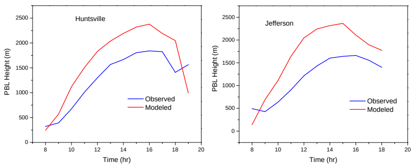


Fig. 3. Fig R3. Temporal variations of monthly averaged modeled and measured PBL heights at Huntsville airport (30.75°N, 95.58°W) and Jefferson airport (29.94°N, 94.00°W).

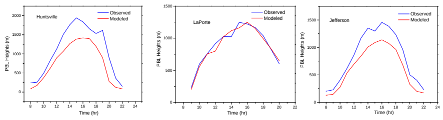
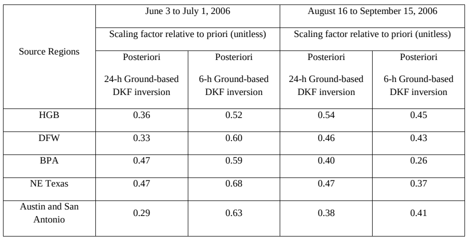
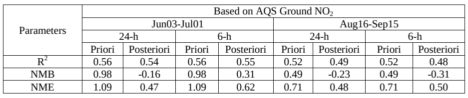


Fig. 4. Fig R4. Temporal variations of monthly averaged modeled and measured PBL heights at Huntsville airport (30.75°N, 95.58°W), LaPorte airport (29.67°N, 95.06°W), and Jefferson airport (29.94°N, 94°W)



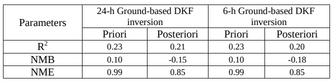
Source Regions	June 3 to July 1, 2006		August 16 to September 15, 2006	
	Scaling factor relative to priori (unitless)		Scaling factor relative to priori (unitless)	
	Posteriori 24-h Ground-based DKF inversion	Posteriori 6-h Ground-based DKF inversion	Posteriori 24-h Ground-based DKF inversion	Posteriori 6-h Ground-based DKF inversion
HGB	0.36	0.52	0.54	0.45
DFW	0.33	0.60	0.46	0.43
BPA	0.47	0.59	0.40	0.26
NE Texas	0.47	0.68	0.47	0.37
Austin and San Antonio	0.29	0.63	0.38	0.41

Fig. 5. Table R1. Scaling factors for each region from different inversions.



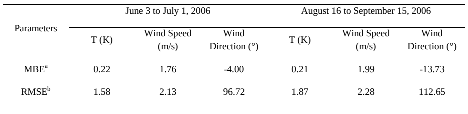
Parameters	Based on AQS Ground NO_2							
	Jun03-Jul01				Aug16-Sep15			
	24-h		6-h		24-h		6-h	
	Priori	Posteriori	Priori	Posteriori	Priori	Posteriori	Priori	Posteriori
R^2	0.56	0.54	0.56	0.55	0.52	0.49	0.52	0.48
NMB	0.98	-0.16	0.98	0.31	0.49	-0.23	0.49	-0.31
NME	1.09	0.47	1.09	0.62	0.71	0.48	0.71	0.50

Fig. 6. Table R2. Performance of CAMx in simulating AQS Ground-level NO2.



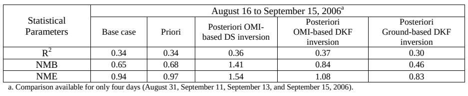
Parameters	24-h Ground-based DKF inversion		6-h Ground-based DKF inversion	
	Priori	Posteriori	Priori	Posteriori
R^2	0.23	0.21	0.23	0.20
NMB	0.10	-0.15	0.10	-0.18
NME	0.99	0.85	0.99	0.85

Fig. 7. Table R3. Performance of CAMx in simulating P-3 aircraft-observed NO2.



Parameters	June 3 to July 1, 2006			August 16 to September 15, 2006		
	T (K)	Wind Speed (m/s)	Wind Direction (°)	T (K)	Wind Speed (m/s)	Wind Direction (°)
MBE[a]	0.22	1.76	-4.00	0.21	1.99	-13.73
RMSE[b]	1.58	2.13	96.72	1.87	2.28	112.65

Fig. 8. Table R4. Evaluation of MM5 in simulating hourly temperature, wind speed and wind direction from 34 ground monitoring sites for both June and Aug-Sep episodes.



Statistical Parameters	August 16 to September 15, 2006[a]				
	Base case	Priori	Posteriori OMI-based DS inversion	Posteriori OMI-based DKF inversion	Posteriori Ground-based DKF inversion
R^2	0.34	0.34	0.36	0.37	0.30
NMB	0.65	0.68	1.41	0.84	0.46
NME	0.94	0.97	1.54	1.08	0.83

a. Comparison available for only four days (August 31, September 11, September 13, and September 15, 2006).

Fig. 9. Table R5. Performance of CAMx in simulating P-3 aircraft-observed NOy.