

Interactive comment on “Nitrogen dioxide observations from the Geostationary Trace gas and Aerosol Sensor Optimization (GeoTASO) airborne instrument: retrieval algorithm and measurements during DISCOVER-AQ Texas 2013” by C. R. Nowlan et al.

C. R. Nowlan et al.

cnowlan@cfa.harvard.edu

Received and published: 18 March 2016

We thank Reviewer 1 for helpful comments. We have made a number of changes to our manuscript based on this review. The reviewer's comments are listed below in italics and our responses and edits follow.

General Comments

C5697

This paper presents an overview of the GeoTASO instrument, retrieval algorithm for NO₂ vertical column data, and comparisons of GeoTASO NO₂ column amounts to ground-based and aircraft-based NO₂ data. GeoTASO was originally conceived as the testbed instrument for the upcoming GEO-CAPE satellite mission, and now also serves as part of mission risk reduction for the GEO-CAPE, TEMPO, and GEMS satellite missions. Thus, it is important to understand the capabilities and limitations of GeoTASO to better understand the capabilities and limitations of the data products from these future satellite missions, especially for short-lived species with heterogeneous sources such as NO₂. The manuscript presents a thorough description of the GeoTASO instrument and NO₂ retrieval algorithm, and is well organized. However, further analysis of how GeoTASO NO₂ column data compare to the other data sets presented in this manuscript is necessary; therefore, I recommend publication after several major revisions.

We have modified several parts of the text and added two figures and one table in response to Reviewer 1. Details follow specific comments below.

Specific Comments

Section 3.2.8: I'm very interested in your use of CMAQ to provide the tropospheric trace gas profiles used in the AMF calculation; I commend your choice! However, please provide a brief explanation of why you chose CMAQ over other regional models or a more traditional global model choice.

A regional model like CMAQ was chosen over global models in order to use the high-resolution simulations to accurately model bay and sea breezes that are important in the Houston area for air quality. CMAQ and WRF were chosen specifically as we have found success in previous applications for modeling bay and seas breezes using these models (see Loughner et al., Atmos. Environ., 2011). In addition, CMAQ has process analysis and source apportionment modes, which were used for work with Texas AQRP, while WRF-Chem at least does not. We have added the following text to

C5698

this section: “The CMAQ simulations used in this analysis have a spatial resolution of 4x4 km² and a temporal resolution of 20 minutes. The local bay and sea breezes that affect air quality in this region are best simulated at the high resolutions of a regional model over those available from most global models. We apply CMAQ and WRF due to previous success capturing local-scale bay and sea breezes with these models (Loughner et al., 2011).”

Section 4.6.2: This comment concerns CMAQ specifically. I'd like to see how the CMAQ profiles/shape factors and surface mixing ratios compare to the observations. I agree with your statement that “uncertainties in model surface estimates vary by time of day” and having a figure that demonstrates this (on average or for just one day) might help explain the nuances in your discussion later in the manuscript of how inferred GeoTASO surface NO₂ compared to in situ surface NO₂. I'd also like to see some comparison of typical profile shapes between CMAQ simulated profiles and the in situ P-3B profiles, to get a feel for where in the vertical CMAQ is simulating the profile and where in the vertical the model struggles. The vertical distribution of a trace gas will go on to affect the shape factor, so this may give a visual indication of how well CMAQ captures the shape factors used in the AMF calculation.

We have added two figures to this section showing 1) profile shapes derived from CMAQ simulations and from P-3B aircraft profiles at locations of aircraft spirals; and 2) average NO₂ surface bias as a function of hour of day, derived from all sites and campaign days. The following text has been added to this section: “Figure 9 shows median NO₂ profile shapes derived from CMAQ simulations and in situ P-3B aircraft profiles collected during the campaign at eight locations of aircraft vertical spirals. These profiles reveal typical uncertainties in profile shape factors are generally less than 20%; uncertainties of this magnitude typically result in a 5% uncertainty in the AMF below the aircraft.”; and “Figure 10 shows the mean bias of CMAQ NO₂ relative to surface observations as a function of local time, derived from all sites and over all days during the campaign.”

C5699

Section 5.1: The two paragraphs dedicated to the discussion of AOD values seem unnecessary here without some analysis. I'd recommend removing these paragraphs, and keeping only the brief discussion of the effect of aerosols on the AMF computation included in Section 4.6.2.

We have removed the majority of this discussion, but kept the following in this section to ensure the three major sources of AMF error are mentioned (surface reflectivity, profiles, and aerosols) for completeness: “Uncertainties in aerosol distribution can contribute large uncertainties to the AMF, and the presence of aerosols can increase or decrease the AMF, depending on aerosol type and altitude (Leitao et al., 2010). Aerosol optical depth and uncertainties specific to the DISCOVER-AQ GeoTASO measurements are discussed in greater detail in Section 5.1.” The following sentence has been moved to Section 5.1, where aerosols on GeoTASO days are discussed specifically: “At these AODs, the biases calculated by Lin et al. (2014) are typically within 25%.” We have also changed the reference from [Wang, 2015] (a proceedings paper) to [Hou et al., 2016] (recently accepted to JQSRT).

Section 5.2.1: The phrasing that correlations “increase with increasing pollution” is unclear to me. It would be helpful to list the four days presented in Fig. 10 in order of pollution, along with r values, to demonstrate how correlation between GeoTASO and Pandora column NO₂ increases with increasing pollution level (as indicated by column amount). It also seems that Sept. 14 and Sept. 18 give essentially the same correlation for different pollution levels (as indicated by GeoTASO column values): are these correlations statistically different or the same, and, if the same, how does this affect your statement that the correlation increases with increasing pollution? I'd also like to see some discussion of the correlation on the other 3 less polluted days, and what the comparison between GeoTASO and Pandora might mean for our ability to remotely sense NO₂ from space under various pollution levels. What is the

C5700

overall correlation over all 4 days of data between the GeoTASO and Pandora data, in addition to the correlation on individual flight days? Why not also present that analysis, which would also lend itself to some discussion of the variability (at least variability as it relates to pollution level) in the comparison between an airborne, high altitude remote sensor and ground-based instruments, and thus some statement on what this might mean for geostationary satellites. How the Moody Tower data were corrected also needs clarification in the text in this section.

This phrase was a poor choice of words on our part. We meant to imply that a larger measured range of NO₂ amounts (higher variability) can produce higher correlations as the signal is more discernible from the measurement noise, and the correlations tend to be more significant. We have added a table which lists the sample size, correlation coefficients, p-values, slopes and intercepts of the linear fit for each day, and for the overall analysis, for all validation observations. All correlations are significant at the p<0.05 level with the exceptions of DISCOVER-AQ surface validation site coincidences on 18 September and Pandora coincidences on 24 September. In all cases September 13 is the only day with highly significant correlations (p<0.001). We have rewritten this section to better describe the correlations and their significance, and have modified the text in the conclusion and abstract to include overall correlations and relationships. We have also modified the text to describe how we apply a correction for Moody Tower Pandora observations that accounts for the partial column below the tower: "At this downtown location with high levels of NO₂, we find that Pandora observations must be corrected for the large partial NO₂ column below the tower. The Pandora total column is corrected by the addition of the column below 70 m as determined from in situ observations collected every 5 min by the University of Houston from both the top and base of the towers. The in situ observations indicate that NO₂ was generally well-mixed below 70 m during these observations, and the partial column is determined from the product of the mean in situ mixing ratio and the CMAQ air column below 70 m."

To address the geostationary question in this and other comments, we have added

C5701

the following paragraph: "It is also useful to consider these results in light of geostationary validation comparisons, such as those planned for TEMPO. Generally, higher pollution levels result in larger variability and a larger NO₂ range, and therefore a higher potential correlation between GeoTASO and coincident ground-based observations. TEMPO will have a much coarser spatial resolution (2.1×4.5 km² at the center of the field of regard) and a higher precision (< 1 × 10¹⁵ molecules cm⁻²). TEMPO will likely see similar features when compared with surface observations (a correlation that typically increases with an increasing range of NO₂), but may see a reduced slope, due to coarser spatial resolution, and a reduced correlation coefficient, due to both coarser spatial and temporal resolutions than those of GeoTASO."

Section 5.2.2: Similar comments apply to this section as to the previous section. I'd like to see some discussion of the correlation between inferred and in situ NO₂ on the other three days presented, as well as of the overall correlation between these datasets. Again, what might this mean for the capabilities of the geostationary satellites?

The previously mentioned table now includes information on other days and overall statistics. We have also modified the text to discuss overall results.

Section 5.2.3: As with the discussion of the comparisons between GeoTASO and Pandora or in situ surface data, why the emphasis on only the Sept. 13 flight day? How do GeoTASO and GCAS compare on one of the less polluted days?

We have added statistics for comparisons with GCAS on other days and for the overall comparison in the new table. The number of samples listed in the table for each day show that September 13 had about 2.5 times more coincidences than any other day. In addition to the fact that September 13 was the day with the fewest clouds, the Falcon and B200 aircraft had the tightest coincident flights in space and time on this day. For instance, on September 14, the Falcon took off nearly 2 hours after the B200, only catching up just before the B200 started its descent. There are also far fewer coincident observations on September 18 and 24, partly due to cloud, but also due to

C5702

less observations coincident in space and time within our coincidence criteria. As with the previous sections using ground-based data, we have modified the text to mention the overall comparisons.

Section 5.2.4: This section seems unnecessary, as very few satellite data were available for this comparison, and the comparison is complicated by several issues. I recommend removal of the comparison to GOME-2 NO₂ data.

Although there is little satellite data available for comparisons with the aircraft data, we believe this figure highlights the impressive spatial resolution of the aircraft data and the inhomogeneity of NO₂ within a satellite ground footprint of spatial resolution from currently available sensors. We have kept the figure and related text for now, but are willing to remove if the editor and reviewer are convinced it is superfluous to the study. We have added the following phrase to the sentence introducing the figure within the text to further emphasize the inhomogeneity point: ", and illustrates the potential inhomogeneity within a GOME-2 ground pixel."

Technical Corrections

Page 4, Line 29: add "the" before "x dimension of the array" Page 26, Line 21: Please omit the second "can" from this sentence.

These changes have been made in the revised manuscript.

References added:

Hou, W., Wang, J., Xu, X., Reid, J. S., and Han, D.: VLIDORT: An algorithm for hyperspectral remote sensing of aerosols: 1. Development of theoretical framework, *J. Quant. Spectrosc. Ra.*, doi:10.1016/j.jqsrt.2016.01.019, 2016.

C5703

Loughner, C. P., Allen, D. J., Pickering, K. E., Zhang, D.-L., Shou, Y.-X., and Dickerson, R. R.: Impact of fair-weather cumulus clouds and the Chesapeake Bay breeze on pollutant transport and transformation, *Atmos. Environ.*, 45, 4060–4072, doi:10.1016/j.atmosenv.2011.04.003, 2011.

Figure captions:

Figure 1: Normalized median profile shapes from CMAQ model simulations and P-3B aircraft NO₂ observations at P-3B spiral locations during the DISCOVER-AQ Texas campaign. The profiles use model output and observations binned to a 250 m altitude grid.

Figure 2: Mean bias in NO₂ surface mixing ratio of CMAQ simulations relative to surface observations averaged at all campaign validation sites. The pale pink (AM) and blue (PM) shadings indicate the time range of GeoTASO flights, while the darker shadings indicate the time range of coincident GeoTASO and surface observations.

Interactive comment on *Atmos. Meas. Tech. Discuss.*, 8, 13099, 2015.

C5704

Table 1. Summary of NO₂ comparisons between Pandora total column observations, in situ surface validation mixing ratios and GCAS aircraft slant columns with GeoTASO observations.

	13 September	14 September	18 September	24 September	All dates
<u>Pandora total column</u>					
N	39	35	31	21	136
r	0.90	0.41	0.48	0.16	0.73
p-value	<0.001	0.01	0.01	0.57	<0.001
slope	1.59	0.79	1.07	0.88	1.31
intercept (10 ¹⁶ molecules cm ⁻²)	-0.47	0.18	0.14	0.12	-0.13
<u>TCEQ surface sites</u>					
N	26	23	21	12	82
r	0.89	0.89	0.60	0.64	0.85
p-value	<0.001	<0.001	0.004	0.03	<0.001
slope	1.36	1.11	0.64	1.40	1.14
intercept (ppbv)	-5.3	-0.6	0.8	-2.5	-1.9
<u>DISCOVER-AQ surface sites</u>					
N	17	16	11	10	54
r	0.91	0.49	0.51	0.74	0.85
p-value	<0.001	0.05	0.11	0.02	<0.001
slope	1.29	1.46	0.54	0.70	1.14
intercept (ppbv)	-3.5	-1.0	1.3	-1.1	-1.8
<u>GCAS slant column</u>					
N	48555	929	18111	9725	77320
r	0.84	0.25	0.72	0.53	0.81
p-value	<0.001	<0.001	<0.001	<0.001	<0.001
slope	0.94	1.41	0.82	1.24	0.91
intercept (10 ¹⁶ molecules cm ⁻²)	-0.01	0.03	-0.08	-0.01	-0.02

N:

number of coincident measurements

r: correlation coefficient

slope and intercept: results of reduced major axis linear regression of observations versus GeoTASO

C5705

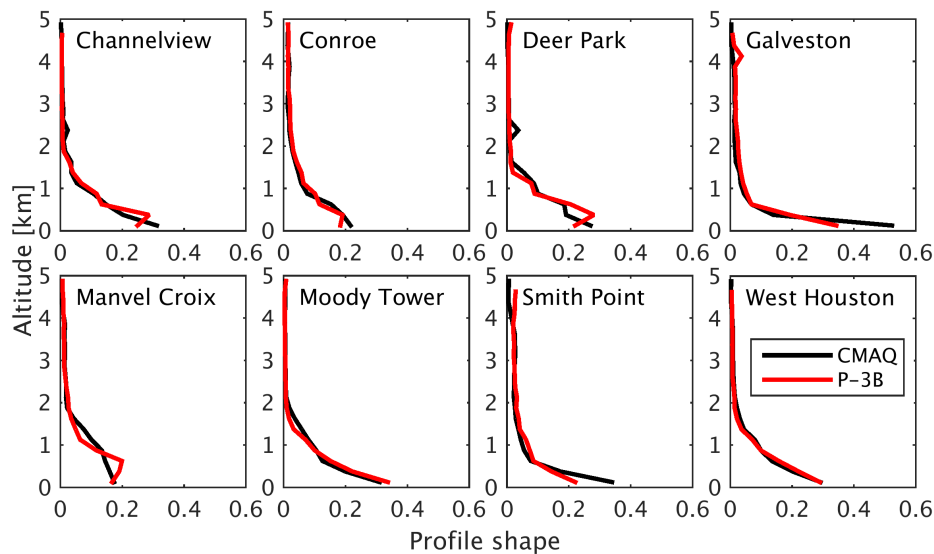


Fig. 1.

C5706

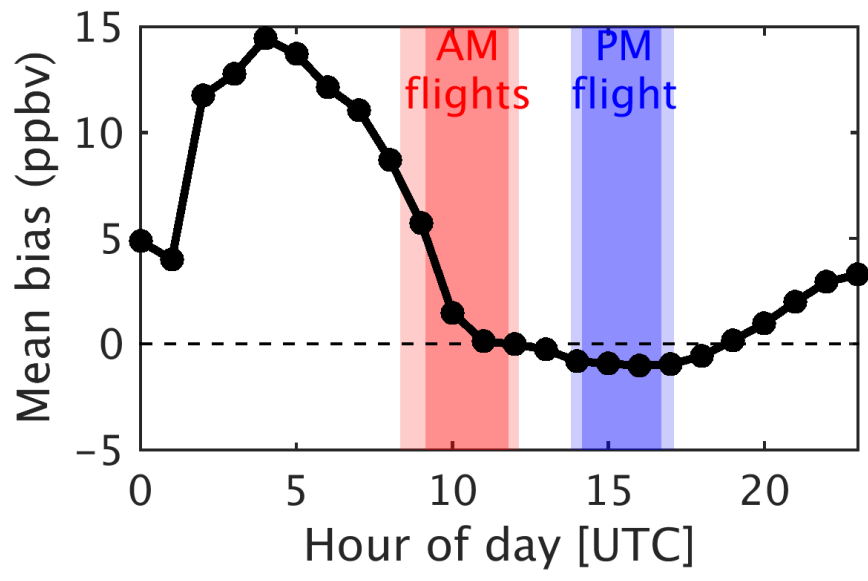


Fig. 2.

C5707