Dear Anonymous Reviewer #1,
Our responses to your comments are in red.

Dear Authors,

Thank you for this well-written and detailed manuscript describing this large-scale experiment to attempt to understand the offset in retrieved AOD between the morning and afternoon MODIS instruments. You have presented some analysis to examine the expected magnitude of certain effects and the implications of recent re-examinations of the MODIS L1B calibration, through the lens of the persistent AOD and Angstrom Exponent biases in the MODIS-Terra MOD04 Dark Target (DT) aerosol product compared with the identical product produced from MODIS-Aqua.

I applaud your support for the community in preparing this publication to summarize your results even though the outcome is unsatisfying because the discrepancy is not resolved.

Thank you for your support!

There are always more tests to be done, but my purpose is not to demand that you add multiple papers' worth of analysis (though I hope you will continue to pursue this question and publish more papers about it!). In preparing the final version of this current paper, I think care should be taken to present the outcome of the experiments in terms of what is known, and to separate findings into the categories of "constructive recommendations for analyses combining Terra and Aqua MODIS" and "directions for resolving the discrepancy in MODIS retrieved aerosol properties."

We also hope to publish more good and useful papers. Thank you.

You present AERONET comparisons indicating that the global offset between Terra and Aqua is reproduced when sampled to times and locations of AERONET data, and that the Terra-Aqua differences do not correspond to differences between morning and afternoon AERONET AOT. This is consistent with analysis of AERONET morning and afternoon retrievals (without pairing to MODIS, Figure C1); AERONET also shows no systematic trend of Angstrom exponent between morning and afternoon (Figure C2). These results are consistent with literature e.g. [*Smirnov et al.*, 2002], but an update to Figure 17 from Levy et al. (2013) would be relatively easy and a welcome addition to your Figure 2.

We like your suggestion and have added a plot (panel 2C) showing the Angstrom Exponent.

In light of the AERONET results, the MERRA model experiment does not add much additional weight. MERRA is constrained by MODIS radiances tuned to AERONET, and from that perspective would be expected to show the same trends as AERONET. MERRA assimilation does not provide information on aerosol diurnal variation outside the Terra and Aqua overpass times. Aerosol sources in MERRA introduce sub-daily variation according to the source functions used, some of which are dependent on meteorology (e.g. dust) and some of which are climatological (e.g. diurnal cycle of smoke) or inventory-based (e.g. anthropogenic pollution). The sub-daily variation in aerosol sources in MERRA cannot be considered definitive. Thus neither the aerosol sources nor the data assimilation contribute a large amount of validated information on subdaily variation.

Sorry, we realize we were not clear enough. The model results used in this study do not invoke aerosol data assimilation in any way, and so have no constraint imposed from either AERONET or MODIS.

Although we drive the simulations with meteorology from MERRA-2, which did see aerosol data assimilation, it is only the MERRA-2 wind, pressure, and temperature fields that are used to drive our replay, with the GOCART aerosol scheme following its own lifecycle: Emission sources are prescribed or dynamic, as the reviewer suggests, but the AOD that evolves is then a function of model aerosol processes (e.g., losses), transport, and interplay of the aerosols with other atmospheric processes. This is a configuration similar to other chemical transport models you may be familiar with. So, while there is uncertainty in diurnal variability in aerosol sources and how well those are represented in the model, there is an effort to realistically capture the AOD variability throughout the diurnal cycle due to, for example, transport within and outside the evolving boundary layer and in response to diurnal cycles in relative humidity. The point of this exercise, was to ask, to the best of our knowledge, whether the observed offsets between the two MODIS sensors could be due to diurnal differences in aerosol. That answer is no.

We revise the text within the manuscript.

From the perspective of this reviewer, it seems the strongest case made by your results is that calibration is unlikely to be the cause of the observed differences. The four different L1B calibrations tested (C6, C6+, C6- Doelling, C6.1) are relatively independent, and might as an ensemble be taken to represent the current state of knowledge about how to calibrate a multispectral sensor such as MODIS. None of these calibrations eliminate the Terra-Aqua offsets in retrieved aerosol properties. The observed AOD offsets are at least double the magnitude over land than can be explained by the C6/C6+ difference, and many times larger than the C6/C6+ difference over ocean (Figure 11). In order for L1B radiance calibration to be responsible for the discrepancy, the correct radiance calibration would have to be different from all of variants considered in your paper by considerably more than the difference between them. In the absence of other evidence that these calibrations are systematically flawed, we must consider this unlikely.

Yes. This is one of the main messages of the paper. While calibration is a priori an obvious source of the Terra-Aqua offset, so far, efforts to calibrate the sensor have not been adequate to bring the sensors to agreement in terms of the aerosol retrieval. However, we do not believe that the four calibrations offered to date and described here have exhausted the full range of possible calibration for a multi-wavelength, multi-detector, scan mirror sensor. We have changed some wording to make sure that it is clear that calibration continues to be a possible problem.

If the small residual Terra-Aqua offset cannot be explained by current state-of-the-art work on L1 radiance calibration and is not consistent with our current understanding of aerosol sources, we must consider other possibilities. These are outside the scope of your current paper, but I think you should include some discussion of the potential explanatory power of some other possibilities. For instance:

1) The MODIS retrieval makes simplifying assumptions about particle optical properties because the retrieval is underdetermined. The discrepancy between the single scattering albedo (SSA) assumed in the MODIS-DT retrieval and the actual particle properties has been shown to be a significant source of bias in MODIS retrieved AOT [*Eck et al.*, 2013]. Is there a set of optical properties consistent with the MODIS-Terra bias vs AERONET? Can this be falsified using other data from AERONET?

This is an interesting hypothesis. Over land, the MODIS-DT algorithm assumes a given particle type based on AERONET-retrieved climatology (season and location). As you suggest, we might see systematic biases between MODIS and AERONET (e.g. Eck et al., 2013; Ichoku et al., 2003]. It makes no attempt to use different optical properties for AM versus PM. If there is a systematic difference between particle properties (say refractive index or size distribution), we agree that that could lead to a systematic bias for Terra versus Aqua, and Terra/AERONET versus Aqua/AERONET. However, based on your attached plots showing near 1-1 for AM vs PM AERONET AOD and AE, we can't see this being the reason.

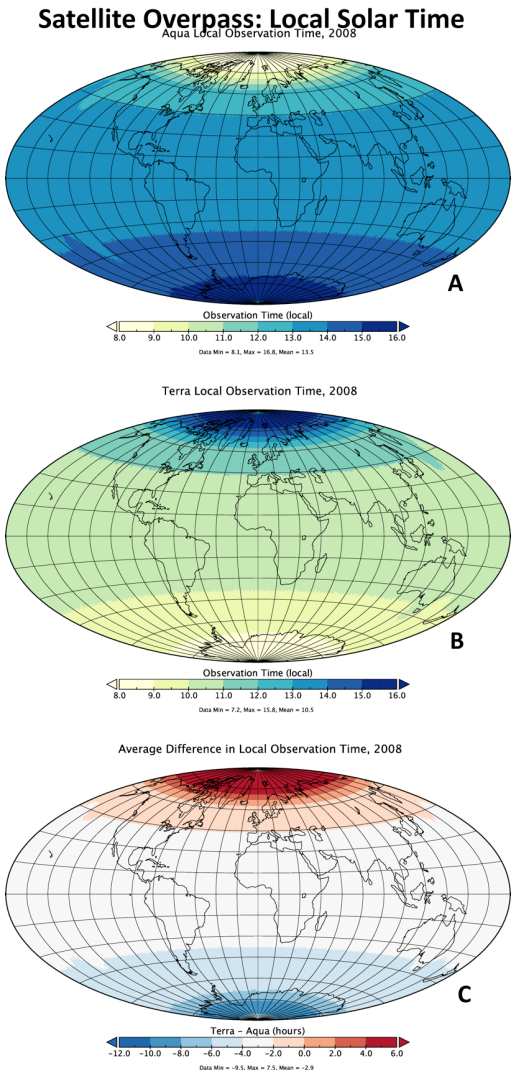
2) It is documented that cloud types and cloud properties show significant diurnal variation in the climatological mean [*Eastman and Warren*, 2014]. Eck et al. [2012] diagnosed how cloud processing affects aerosol particle size distributions. Can an envelope calculation be made of the expected magnitude of diurnal differences in particle optical properties related to cloud processing? Hypothetically, if the MODIS-DT assumed particle properties were assumed to match the real atmosphere for MODIS-Aqua observations, what magnitude of difference in optical properties could account for the MODIS-Terra AOD offset?

If the size or refractive indices of the aerosol particles were to systematically change from morning to afternoon (e.g. due to changes in RH or cloud processing), then there might be a systematic bias between morning to afternoon. However, your attached figures (showing really no AM/PM difference in AOD or AE from AERONET) seems to suggest that we should not expect systematic changes in particle properties from AM to PM. It would be very interesting to get a better handle on delta-optical properties/size versus change in AOD bias, but well beyond the scope of this paper.

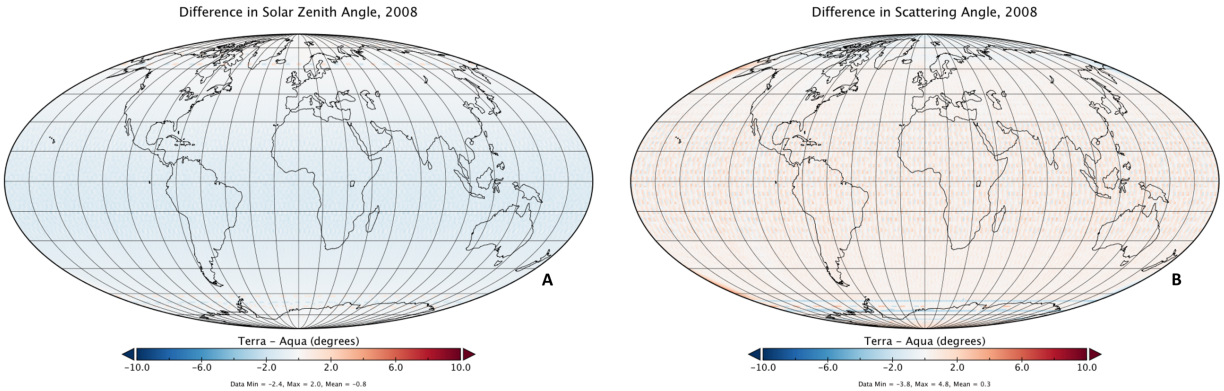
3) The distribution of observation angles is different for MODIS-Terra and MODIS-Aqua. Numerous analyses have shown that MODIS-DT retrievals have some sensitivity to observation geometry (e.g. [*Hyer et al.*, 2011]). I believe that the pole-to-pole uniformity of the Terra-Aqua bias argues against this as a contributing factor, but with some simple additional tests it might be possible to rule it out entirely. I think the analysis you have done is solid. I would like to see the discussion at the end of this paper expanded to:

This is a really interesting comment, and one we originally debated on whether to include more discussion in the paper. To answer you and the other reviewers, we have created some plots regarding the differences in Local Time observed by the two MODIS sensors (As new Figures within the paper), as well as plots showing relative differences of geometry (shown here only). For the geometry, when looking at 2008 only, there is on average a 0.8° difference in solar zenith angle (Terra < Aqua), and associated difference of 0.3° in scattering angle (Terra > Aqua).

We have added some new text within the paper.



NEW Figure 1: Gridded average MODIS local observation time (local solar time) for Aqua (A), Terra (B) and the difference between the two (C).



Extra Figure (not in paper): Gridded average solar zenith (A) and scattering angles (B) for 2008. Each panel represents the difference between averaged MOD04 and averaged MYD04 (Terra-Aqua).

A) Include a quantitative description of how Terra-Aqua offsets compare to the magnitude of sub- daily variation as observed by AERONET. This is the clearest expression of the where we would need to get to in order to use Terra-Aqua to diagnose diurnal processes.

This is beyond the scope of this paper, but there are papers that have looked at this idea for earlier MODIS collections. Kaufman et al., (2000) shows that using AERONET data sampled at the MODIS passing time, the global AOD diurnal cycle is within 2% of the AOD, which is at the same magnitude or much smaller than the discrepancies between Terra and Aqua retrievals that we discovered in this study depending on the time span of AERONET data used. There are other studies focus on aerosol regional diurnal cycle shows wide range of daily variations of AOD depending on locations and/or seasons (Smirnov et al., 2002; Yan et al., 2012,). However, detailed analyses of Terra and Aqua differences regionally is highly depending on the observing conditions and aerosol model assumptions/selections.

B) Include a hypothetical discussion of other differences in the atmosphere that could cause Terra and Aqua to report different answers, drawing on existing literature to estimate the magnitude of different effects.

Again, this is beyond the scope of this paper, but is worthwhile doing. There are a number of reasons why Terra and Aqua might show different AODs

- Actual differences between AOD in morning and afternoon. This could be caused by differences in aerosol size or optical properties (possibly related to changes in RH or to cloud processing), and/or of actual aerosol loading (e.g. smoke maxima in late afternoon). This is not well supported by either the MERRA-2 analysis in the paper or the AERONET analysis you provided in this review.
- Differences between morning and afternoon sampling, either because of differences in cloud fraction (affecting retrievability) or differences in geometry. There appears to be some correlation between cloud fraction (e.g. King et al., 2013) and the AOD differences.
- Although the retrieval algorithm "corrects" for gas absorptions (column water vapor, ozone, etc), unknown differences between morning and afternoon (for example if 12 UTC water vapor was used for both 10:30 and 13:30 overpasses) could lead to systematic biases in retrieved AOD.
- Since the Terra-Aqua bias is similar to the difference between Terra-AERONET and Aqua-AERONET (Terra-Aqua = Terra-AERONET – Aqua-AERONET), and this is coupled with the overall very little change in AERONET-observed AOD or AE, we continue to suspect that MODIS calibration is the main culprit. It's not just calibration in the bands we retrieve upon (e.g., C6+ versus C6), but also maybe requires more detailed analysis of thermal infrared channels and 1.38 μm bands used for cloud detection and masking. If both sensors were observing the exact same scene with the exact same geometry (impossible), would they observe the exact same reflectance and thermal radiance in all channels?

Eastman, R., and S. G. Warren (2014), Diurnal Cycles of Cumulus, Cumulonimbus, Stratus, Stratocumulus, and Fog from Surface Observations over Land and Ocean, *Journal of Climate*, *27*(6), 2386-2404, doi:10.1175/jcli-d-13-00352.1.
Eck, T. F., et al. (2012), Fog- and cloud-induced aerosol modification observed by the Aerosol Robotic Network (AERONET), *J. Geophys. Res.-Atmos.*, *117*, 18, doi:10.1029/2011jd016839.

Eck, T. F., et al. (2013), A seasonal trend of single scattering albedo in southern African biomass-burning particles: Implications for satellite products and estimates of emissions for the world's largest biomass-burning source, *J. Geophys. Res.-Atmos.*, *118*(12), 6414-6432, doi:10.1002/jgrd.50500.
Hyer, E. J., J. S. Reid, and J. Zhang (2011), An over-land aerosol optical depth data set for data assimilation by filtering, correction, and aggregation of MODIS Collection 5 optical depth retrievals, *Atmospheric Measurement Techniques*, *4*(3), 379-408, doi:10.5194/amt-4-379-2011.
Smirnov, A., B. N. Holben, T. F. Eck, I. Slutsker, B. Chatenet, and R. T. Pinker (2002), Diurnal variability of aerosol optical depth observed at AERONET (Aerosol Robotic Network) sites, *Geophys. Res. Lett.*, *29*(23), 4, doi:10.1029/2002gl016305.

Additional new references:

Ichoku, C., Remer, L., Kaufman, Y., Levy, R., Chu, D., Tanre, D. and Holben, B. (2003), MODIS observation of aerosols and estimation of aerosol radiative forcing over southern Africa during SAFARI 2000, *J Geophys Res-Atmos, 108*(D13), 8499–8499, doi:10.1029/2002JD002366.

Kaufman, Y. J., Holben, B. N., Tanré, D., Slutsker, I., Smirnov, A. and Eck, T. F. (2000), Will aerosol measurements from Terra and Aqua Polar Orbiting satellites represent the daily aerosol abundance and properties? *Geophys Res Lett, 27*(23), 3861–3864, doi:10.1029/2000GL011968.

Zhang, Y., Yu, H., Eck, T. F., Smirnov, A., Chin, M., Remer, L. A., Bian, H., Tan, Q., Levy, R., Holben, B. N. and Piazzolla, S. (2012), Aerosol daytime variations over North and South America derived from multiyear AERONET measurements, *J. Geophys. Res., 117*(D5), D05211–, doi:10.1029/2011JD017242.