Response to Reviewers

We would like to thank all reviewers for their helpful comments and criticism on this work. We believe we have addressed the comments and made changes to the methodology and manuscript where possible. We now include supplementary fig ures and several of the figures in the manuscript have been updated.

Key changes include:

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- Analysis and statistics generated for log(AOD) rather than AOD
- Instrument uncertainty included in the estimate
- Regional bias correction of satellite data by AERONET
- Uncertainty in bias correction propagated through analysis
- Marine Aerosol Network (MAN) data included
- Supplementary figures of AERONET and satellite AOD histograms
- Comparison of model AOD with daily AOD from MAN
- Supplementary comparison with deposition flux

The key changes are that the global dust AOD is decreased from 0.033 to 0.030 and the uncertainty increased from 0.006 to 0.011 (2a) as a result of considering instrument uncertainty and the uncertainty on the updated AERONET bias correction of the satellite retrievals. The observational estimate is hence closer to the AEROCOM model estimate. We believe that this better corrects for regional biases in the satellite retrievals while representing the inherent uncertainty in using limited in-situ measurements to apply correction factors over large regions. The regional estimates of seasonal dust AOD from the different satellite instruments are generally in closer agreement. The observational estimate is also brought closer to the MERRAero dust AOD; the previous discrepancy was of some concern because MERRAero assimilates MODIS AOD and may be expected to represent the dust AOD better than models without assimilation. The agreement between model and observational estimate improves over the mid-Atlantic, reducing (but not eliminating) the potential for systematically high dust removal in the models. While many of the quoted numbers change as a result of our reanalysis, all other conclusions remain essentially the same.

Please find the reviewer-specific comments and responses (blue italics) listed below.

30 Kind regards, David Ridley

Amato Evan

This manuscript describes a method of combining satellite and model data in order to estimate the global dust AOD (DAOD).

The principal idea here is that models do a good job of simulating non-dust AOD, and satellites do a good job of retrieving the total AOD, so the difference between the two should be a good estimate of DAOD. While I applaud the authors on their creative effort, and the obviously massive amount of time undertaken to complete this work, I find there to be a couple of major issues with the methods that I suspect are contributing to a bias in their global DAOD estimate, and increase the uncertainty. Thus, I am suggesting a major revision.

Major Comments

1. Å major assumption of this method is that model DAOD is biased, but that model AOD is not. However, this assumption, at least the part about model AOD not having any systematic bias, isn't justified. The authors suggest that they are accounting for errors related to underestimation of the non-dust AOD by reporting their global DAOD with a 2-sigma uncertainty range (P13, L15). However, if the models systematically underestimate the non-dust AOD, this will induce a high bias in their reported global DAOD, and thus simply increasing the uncertainty range isn't really appropriate. We need to know if there is a bias, particularly because a low bias in modeled non-dust AOD would serve to push the hybrid global DAOD estimate closer to the aerocom mean, and possible closer to the MERRAaero estimate. One could determine if such a bias exists by comparing histograms of AOD for the models and AERONET, over land regions and over-water regions where there is no dust (but there

is smoke, anthro. aerosols, and marine aerosols). The difference in those histograms can be used to calculate a bias (which could be corrected) and uncertainty in the models' non-dust AOD. These errors can then be carried through to the final global DAOD calculation.

5 > A major assumption of this method is that model DAOD is biased, but that model AOD is not. However, >this assumption, at least the part about model AOD not having any systematic bias, isn't justified.

This is certainly a valid concern. We use multiple models to estimate the uncertainty and consider regions where dust aerosol dominates the AOD to minimize the impact of errors in modeled non-dust AOD. For example, the Gulf of Guinea region is not considered explicitly because of the influence of biomass burning. However, as you point out, this may not be sufficient if all models are biased in the same direction. The difficulty is in isolating cases in the dust-influenced regions we consider, but when the dust AOD is not significant. For example, even if we look during wintertime in the Middle East, when dust emissions are low, there is no guarantee that the non-dust AOD will adequately represent the non-dust present during the summertime (looking in other regions, as suggested, is problematic as we may just be observing a local bias that is irrelevant to the dusty locations).

To explore potential biases in modeled non-dust AOD we separate the daily coincident AERONET and model AOD based on whether the model dust AOD contributes >60% or <60% of the total AOD. There is indeed evidence of a bias in CESM and GEOS-Chem. From the added supplementary materials:

"We find that there is a bias between these two cases where CESM and GEOS-Chem both underestimate the AOD relative to AERONET in low dust cases and overestimate the AOD in high dust cases. WRF-Chem and MERRAero show a smaller bias in the opposite direction. Relative to AERONET, the models are biased by -23%, -20%, +3%, +10% (GEOS-Chem, CESM, WRF-Chem and MERRAero, respectively) for the low dust cases, and biased +33%, +12%, +14% and +6% for the high dust cases. The days with low dust AOD in the models are biased low most at AERONET sites in the Thar Desert and Kyzyl Kum, that have limited AERONET data, in the Middle East, and across Africa. This suggests that the non-dust AOD in the models may be biased low on average, which would lead to a high bias in the observational estimate of the dust AOD.

If we re-run the analysis including a regional bias correction factor for the models, we find that the mean estimate of the global dust AOD is reduced to 0.028, a 7% decrease but still well within our uncertainty estimate. However, with the bias correction applied, the observational estimate of dust AOD in the Mid-Atlantic is unrealistically close to zero in winter and can end up being consistently negative in the Thar desert, suggesting the bias correction is overcompensating. The agreement in seasonal dust AOD between different satellite-model realizations is also worsened, rather than improved. Finally, there is no guarantee that the model dust AOD is an adequate filter to partition the data into low/high dust days, the filter may simply select for seasons when less dust present, which might not tell us much about the non-dust AOD in seasons when dust is present. For this reason, we do not bias-correct the model non-dust AOD for the observational estimate of global dust AOD presented in this work. However, we highlight this potential source of uncertainty in the main text and included a reference to this supplementary text in the summary of explored biases and uncertainty (Table 2)."

40 In the main text (Section 4.4, pg 15) we add:

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"...the non-dust AOD in all models may be systematically biased high or low, which would bias the observational estimate of the dust AOD low or high, respectively. Comparison between modeled and observed AOD at the AERONET sites and MAN ship locations does suggest a low bias in the modeled total AOD in some of the regions considered, although there is no clear systematic bias in the models (see Figures S5 – S9). Comparison of model and AERONET AOD in low and high dust cases (using the model dust AOD to discriminate) suggests that two of the models are biased high and two biased low (Figure S4). Overall, the ensemble of models appears to underestimate the non-dust AOD; correcting this results in a 7% decrease in the global dust AOD estimate (0.028). However, the uncertainties involved in this method are such that we do not include the bias correction in our final estimate (see Supplementary Materials)."

2. I am also very concerned about use of the models' spatial structure of DAOD (the horizontal pattern of long-term mean DAOD). In Eqn 2 the authors rely on the spatial structure of modeled DAOD in order to estimate their hybrid global DAOD. The implicit assumption is that while the models' may exhibit biases in the absolute value of DAOD, they do a good job of reproducing the long-term mean spatial structure. However, later on in the paper (P11, Section 4.3) the authors examine the signs of the difference between modeled DAOD and that from their hybrid method in Fig 9 (Africa, N Atl, Gulf of Guinea), suggesting that the models emit too much dust at the source to compensate for the fact that wet and dry deposition is far too strong. So on the one hand you are saving that the spatial structure of model DAOD is good (Ean 2) and on the other hand it's not (Fig 9). If your hypothesis is correct, that the models emit too much dust because deposition is too strong, then Eqn 2 will introduce a bias into your global DAOD estimate depending on the relative fraction of regions (Fig 1) that are over dust emitting areas and those that are downwind. I think this means that because your regions in Figure 1 are overwhelmingly near or over dust sources, your final global DAOD estimate could be biased low? I'm not entirely sure. . . But the bottom line is that, given this bias in the spatial structure of dust from the models, there is an additional source of uncertainty in the global DAOD estimate, and potentially a bias, related to the distribution of the regions you choose (Fig 1). I'm not exactly sure how you can address this. Maybe add more over-water regions and redo the estimate only using over-water regions, the only using over-land regions, then using both (via Eqn 2)? Or maybe the way to address this potential bias/uncertainty is to recalculate global DAOD using an equal distribution of regions over dust sources and regions downwind of dust sources (also in Eqn 2).

Yes, this is a fair point. The short answer is that there is could be a bias, assuming the excessive removal we infer in the Atlantic is a global issue, but comparison of satellite and model DAOD in remote locations is not possible. We were unable to use the bootstrapping method to determine satellite DAOD over remote regions simply because the DAOD is too low relative to other aerosol AOD and the retrieval sensitivity is too weak (we compared the satellite-retrieved AOD to the MAN network and found that the agreement is much poorer and of limited use over remote locations in the Southern Ocean and Arctic Ocean).

We compare the models with the MAN observations in remote locations to see if there is an obvious low bias. While correlation is poor in remote regions, there is no clear systematic bias present in the models. However, it is not clear how much of a role dust versus non-dust aerosol plays in this. We refer to previous comparisons of modeled and observed dust surface concentration that show considerable spread in the agreement in remote regions (see Figure 4 of Huneeus et al.; 2011) that limit our ability to discern whether the models used here are unbiased in their representation of the local-to-remote dust distribution. We have added the supplementary figures of comparison of AOD between models and AERONET and MAN, and in the manuscript we have more clearly highlighted this uncertainty in Section 4.4 ("Discussion of the remaining uncertainties"):

"Modeled dust AOD is used as a scaling factor to determine the global dust AOD from the regional observational estimates. We use multiple models to represent the uncertainty, but there may be a systematic bias present, rather than the ±6% uncertainty presented (Table 2). If the over-zealous removal of dust in models, highlighted in the mid-Atlantic, is a global phenomenon then the models would predict too much dust in the source regions relative to downwind and yield a low regional-to-global scaling factor. Similarly, dust emissions schemes currently used in the models are unlikely to reproduce emissions where vegetation cover is variable and will not represent dust from agricultural regions (Ginoux et al., 2012). If those emissions are substantial, then it is possible that tuned emissions in models overestimate emissions from large, permanent dust sources to compensate for the lack of agricultural emissions, which could partially explain model bias towards African emissions.

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3. Lastly, I think models report AOD even in the presence of 100% cloud cover. So, in the model, there could be an aerosol layer overlaying stratus clouds, and the model would save an AOD value. However, in the satellite world, there would be no

AOD retrieval. Does this discrepancy induce a bias? Can you examine the model data (I guess you'd need daily or hourly output) to see?

Yes, we have performed this analysis to compare the DAOD with all GEOS-Chem data and with the model data masked where any grid box in a column has >50% cloud cover (based on MERRA reanalysis). We find that the change from masking is small, resulting in a 2% increase in the DAOD. We now mention this explicitly in the main text (pg 11):

"We also calculated GEOS-Chem global dust AOD after masking columns that have >50% cloud cover in any grid box, based on MERRA reanalysis. This causes the global dust AOD to increase by 2%, relative to when no masking is used, indicating that the difference between clear-sky and all-sky dust AOD is small. However, we acknowledge that poor representation of clouds in the reanalysis meteorology or potential satellite misclassification of heavy dust loading as cloud (Darmenov and Sokolik, 2009) could lead to a stronger perceived relationship between dust loading in cloudy and clear sky conditions."

also by masking the model AOD with satellite retrievals. The latter effectively masks for clouds as well as for overpass frequency for better comparison between model and observations. Masking the model DAOD with Aqua and Terra has a negligible effect on the global DAOD (<1%) and masking with MISR increases the global DAOD by 1-2%. This rather surprising finding indicates that the model global DAOD is not significantly different whether or not cloudy locations are included. We have made this more explicit in the text as follows:

"We calculate the modeled global dust AOD with and without masking to match the MODIS and MISR sampling, testing whether sampling affects the derived global dust AOD. We find negligible (<1%) changes in the modelled global dust AOD when sampling to the MODIS instruments and an increase of 1 - 2% when sampling to MISR. Therefore, we determine that sampling frequency is sufficient to represent the AOD in the regions considered. Furthermore, because the masking effectively removes cloudy regions, the very small change in the modelled global dust AOD indicates that there is no obvious bias in the global dust AOD when including regions within cloudy air masses, relative to clear-sky only"

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Minor Comments
1. P7, L4: Spelling, "main" not "man"

35 Changed

2. Should alpha have a region superscript in Eqn 1?

Previously no, but now that the bias correction is regional it has been added.

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3. P11, L25: Why would a lack of convectively driven dust emissions cause an overestimation of DAOD? Seems like it would be the opposite.

The convectively driven dust is strongest in summer, so a lack of that source causes an over-emphasis of winter and spring relative to summer. I think the sentence reflects your intuition.

4. P7, L26: You write, "In the regions analyzed here the AOD is predominantly driven by dust aerosol, limiting the influence of the model non-dust AOD" but this simply isn't true. Region 1 (N. Atl) also has a big biomass burning contribution in the

boreal winter. Regions 8 also has a contribution from anthro. aerosols from N. India during the dry monsoon season. Same for region 10 (from Pakistan and Iran).

Yes, this statement does overemphasize the importance of dust aerosol. We have softened the language as follows:

"In the regions analyzed here dust aerosol plays a key role and often dominates in the spring and summer, limiting the influence of the model non-dust AOD. Exceptions to this are in South America, South Africa, and Australia, that have a minimal impact on the global dust AOD, and the Gulf of Guinea, where significant biomass burning aerosol is present (we consider results with and without these regions, see Table 1)."

- 5. P10: Cloud filtering: Interesting that you are getting such a strong correlation between the two. Misclassification of optically thick dust as cloud may be pretty common, FYI.
- Yes, we thought this was interesting. We have added a reference to Darmenov and Sokolik (2009) in there to point out that misclassification of optically thick dust may be occurring.
 - "We acknowledge that satellite misclassification of heavy dust loading as cloud may occur (Darmenov and Sokolik, 2009) potentially leading to a stronger relationship between dust loading in cloudy and clear sky conditions."

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Anonymous Referee #3

In this manuscript the authors present a global reconstruction of dust AOD based on satellite data and sun photometer retrievals, using sun photometer data to correct satellite bias and various model simulations to separate the regional contribution of dust from other aerosols. This is a really nice manuscript with some good ideas and a dataset the has the potential to be a widely cited reference. Because of this potential it

is necessary to be extra careful, though. The authors have followed previous methodology, including the weaknesses. I'd like to see these addressed before I support the publication of this manuscript.

30 We thank the reviewer for their comments and generally positive view of the research. We appreciate the points raised and believe that the major concerns have been addressed, detailed below.

- 35 Major Comments:
 - 1. One of my major concerns is the use of the different emission schemes in different models. This will have an impact on the calculation of the dust AOD (eq. 1). How much of the model-ensemble uncertainty is due to different emission schemes and how much due to inter-model variability?
- Without the ability to plug the same emission scheme into each of the models this is difficult to assess. Through the ensemble of satellite instrument and model combinations we can separate the impact of model diversity in non-dust AOD from the model regional-to-global scaling. This is presented in Table 2, where we assume that the bias is symmetrical around the estimate.

- 45 2. I haven't found an explanation why the AOD reconstruction is limited to the 15 regions. Why do you not reconstruct AOD over the whole globe and show it on a map (e.g. using a yearly median)? You can still only calculate the correction factor using the dust-dominated regions.
- Our rationale for using the 15 regions is to isolate the regions that are more influenced by dust than other aerosol species to 50 minimize the effect of mis-categorizing errors in non-dust AOD from the model as dust AOD. For example, the large AOD

from pollution over East China is not always fully represented by the models and would result in a significant bias in the dust AOD inferred over this region. We therefore exclude regions such as this, Europe, and remote areas far from dust sources. Furthermore, through comparison between satellite retrievals and AERONET and the Marine Aerosol Network (MAN) we find that the retrievals are much less reliable in some locations, such as the Southern Ocean, and therefore unlikely to produce useful results. Lastly, the bootstrapping process requires many iterations of the dust AOD calculation at each location and is time-consuming. We have added this rationale to the manuscript as follows (pg 8):

"We focus on regions in which the dust AOD often dominates to reduce potential errors from biases in modeled non-dust AOD."

This is followed by examples of key uncertainties in the non-dust AOD (e.g. regions influenced by biomass burning).

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- 3. The assumption on which the correction factor are based seem weak to me. It is true that most of the global dust AOD is dominated by the North African and East Asian region. But this doesn't mean that you only need to concentrate on a few region, but that the spatial distribution is no Gaussian. In fact, if you look at a histogram of a snapshot in time you will probably find that dust is spatially log-normally distributed. My suggestion to the authors is to look at the spatial distribution of the satellite and sun
- 20 photometer data and if it's lognormal, try to take the logarithm of all initial AOD data such that it is spatially normally distributed and rethink their calculations (especially equations 1-3 and Figure 2) and discussion from that perspective.

We agree that the AOD is usually log-normal, thank you for this observation. We have repeated the analysis assuming the AOD is log-normally distributed when aggregating daily AOD retrievals into a seasonal mean with a standard deviation. Interestingly, we find that this has a relatively small impact on the results, reducing the global dust AOD only slightly. However, this is certainly a better way to represent the statistics and have updated the manuscript accordingly.

We did use AERONET data from all stations when calculating the correction factor, not just those within the 15 regions (see pg6 line 29). However, based on comments from reviewers we have revisited the bias correction methodology and altered this to provide bias corrections for each region, based on the AERONET sites in that region (and using the Marine Aerosol Network where relevant). Histograms of the distribution of AOD for the satellite retrievals and for AERONET within each region are shown in Figure S1 to S3.

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- 4. Global means make sense for GHG but not for aerosols. Talking about a global mean AOD is meaningless. It gives you absolutely no information about what the AOD could be on any point on Earth. I know everybody's doing it and there's a weak argument that can be made for inter-paper comparison's sake. But this manuscript has the potential to be a widely cited reference and it has the means to provide data for more regionally-based comparisons in the future. Figure 4 looks very fancy but gives very little useful information. Maybe in addition to Figure 4 that compares with previous papers you could prepare a synthesis figure or table with which people writing papers in the future can easily compare their results (something like figure 9 but less messy no offense to figure 9).
- We understand the concern with using the global average AOD and its limitations. However, it is necessary to compare with the modeled estimates of Huneuus et al. (2011) and we find that the global dust AOD metric is useful for discussion of the factors leading to uncertainty in the observational estimate. We highlight the importance of regional assessment in Section 43.

"...tuning the models globally will not necessarily produce the right spatial and seasonal distribution. Here we use the observational constraints developed in this study to highlight regional and seasonal discrepancies between models and observations in an effort to isolate potential errors that affect multiple models."

5 Regarding comparisons with future work: we intend for the regional data to be available for future studies to compare against; however, we now provide a summary (Table 3) that gives our observational estimate of average seasonal dust AOD in the regions considered to facilitate quick comparison.

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Minor comments:

The references to air quality and health seem out of place in this manuscript. There is no need to mention these aspect if they are not discussed anywhere.

- 15 We respectfully disagree. We believe it is important to highlight the influence of dust on air quality when AOD retrievals are used in assessing surface PM2.5 in poorly monitored regions, such as Africa, as part of studies informing World Health Organization assessments (e.g. van Donkelaar et al., 2006; Evans et al., 2013).
- 20 Page2, lines 7-9: I don't know if that's a mistake in the original Huneeus paper, but if you give the median because the distribution is not Gaussian, then you shouldn't give the standard deviation, which is a parameter in the Gaussian distribution. The "AEROCOM median" was a combination of the models in the AEROCOM analysis and treated as a separate model. This has been clarified by referring to it as the AEROCOM "model median".
- 25 Chapters 2.1, 2.2, 2.3: I would appreciate it if the description of errors was consistent between the three instruments.

We have homogenized the error format

Page 6&7, lines 32-7: Looking at the data in Figure 2 I would guess that the data is not normally distributed. The choice of a linear regression to calculate the bias between AERONET and satellites is therefore doubtful. See my major comment 3. Hopefully the response to comment 3 addresses this point.

Page 8, Eq.1: In my experience, aerosol concentrations, loads, and therefore AOD are not normally distributed in space. The mean AODs calculated here may not be representative of the central tendency in each region. See major comment 3.

35 Hopefully this was addressed in response to the major comment. The manuscript has been updated to reflect the use of log-normal distributions.

Page 10 line 3: AE<0.4 Figure 2: In the MISR panel, there are values only for one of the two regressions. Also I can see only one regression line

40 This figure has been removed, following suggestions to improve the bias correction of satellite data to AERONET observations, and no longer applies a split regression for MISR. Information on the bias correction of the satellite retrievals is now presented in Table 1 and Figures S1 – S3.

45 Anonymous Referee #4

The study combines estimates of AOD from satellite and sun-photometer (AERONET) observations. The authors evaluate the statistical uncertainty of dust AOD calculated from model simulations against in-situ observations. The manuscript is well written and scientifically sound.

Thank you for your comments, we hope to have covered your concerns below.

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General comment:

Why do you scale the model AOD from regional to global (page 8 & 9, Eqn 3)? The general scaling approach does not consider the regional variability in soil properties (determining dust emission fluxes), meteorological drivers, size distributions (affecting AOD and life time), etc. What is the motivation for ignoring these factors despite knowing that they affect on dust concentrations and dust properties? Are the results after scaling still representative? Please consider including some words on how meaningful the scaling approach is.

The scaling to global dust AOD does rely on the global distribution of dust aerosol in the four models used, and will represent the regional variability in soil properties, meteorological drivers and size distributions to the extent that those models reproduce those properties. We are unable to account for potential biases that exist in all the models; however, the purpose of using four models is to both reduce the impact of these biases and to propagate their effect on the uncertainty in the DAOD by providing a range of scaling factors.

We derive the dust AOD over regions in which dust aerosol makes up a significant fraction of the total AOD to minimize errors from both retrieval uncertainty and model representation of non-dust AOD. Ideally we would derive the dust AOD in all regions to eliminate the need to use the models; however, the uncertainties prevent meaningful results in remote regions. We found that comparison of satellite retrievals of AOD with the Marine Aerosol Network (MAN) showed poor correlation and bias in remote locations. We have added the following text to clarify this in the manuscript (pg 8):

"We focus on regions in which the dust AOD often dominates to reduce potential errors from biases in modeled non-dust AOD."

This is followed by examples of key uncertainties in the non-dust AOD (e.g. regions influenced by biomass burning).

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Related to that, can global averages of dust AODs considered as an appropriate measure for model skills with regard to dust distribution? Regional errors may equal out and thus a global average may be misleading. As also pointed out in the result section, dust varies strongly with regions and depends on the model skills for the regions. Furthermore, on the one side you are arguing with global averages of AOD (i.e. abstract and conclusion), on the other side you are suggesting that regional means are the more appropriate measure. It sounds somewhat inconsistent. Please clarify.

We agree that there is limited use for global dust AOD. However, this is a common metric used to assess models and is presented here to allow comparison with model estimates. Because of the limited use we have provided specific information on the seasonality and the magnitude of the dust AOD in different regions. Between the global dust AOD and the more detailed regional interpretation of the results is a reasonable framework. We address this in opening paragraph of the Section 3.4 on regional dust AOD and have added the following statement to the conclusions:

"...it is essential to evaluate models on regional and seasonal scales, at which we find considerable differences."

p5 114 remove parenthesis for reference Fast et al., 2006 Done
p5 114 remove parenthesis for reference Barnard et al., 2010 <i>Done</i>
p7 14 "man" should be "main" p7 119 should be MERRAero to be consistent Done
p9 114 It appears a bit odd to me to have one of the co-authors cited as "personal communications". Changed
Maybe omit the "personal communication" part and only provide the "manuscript in preparation" part? Done
p9 123 "Eqn. 3" to be consistent Done
p9 127 "Eqn. 1" to be consistent Done
p11 130 As the naming of the regions are erroneous on the figures (see below), please check if it's correct in the text. Thank you, these have been corrected throughout the text.
p13 11 Please consider shifting "(the Gulf of Oman)" to line 26 where the Kyzyl Kum region was mentioned first. Here we are discussing the Gulf of Oman as the region between the Southern Middle East and Kyzyl Kum desert regions. Therefore, we believe the reference to the Gulf of Oman should stay in its current location.
Fig. 1 something went wrong with assigning geographical names to the numbering of the areas. Area number 5 is definitely not the Atlas Mountain region. Maybe confound with the Adrar des Iforas Mountain region? Similarly, the Bodele Depression covers the Sudan, too. Please clarify. Thank you, the Atlas mountains region was mis-labelled and has been corrected to Mali/Niger
Fig. 7, 9, 10 Base on the numbering issue appearing in Fig. 5, there may be a consequent mis-naming of the Atlas region. Please check. Fig. 7, 9, 10 Taklamakan These have been corrected to better represent the regions: Mali/Niger and Bodele/Sudan, and the Taklamakan spelling used throughout.
Anonymous Referee #5 The manuscript describes a new potential tool for validation of mineral dust in global and regional models, based on a combination of remote sensing data and global climate models. The work is certainly of interest and could provide an additional useful tool to the modeling community. In general the methodology appears sound and the paper is well organized and written. A few minor revision are nevertheless needed in my opinion before the paper could be published. Thank you for your assessment of the work. We have addressed your concerns below.

50 Major comment

The construction of the global AOD dataset is the central part of this work. It stems mainly from remote sensing observations, form both satellites and ground-based AERONET stations. I think that too little information is provided regarding data processing (e.g. temporal aggregation) and uncertainties in these types of observations and their relation to dust AOD.

- 5 In Section 3.1, we have added more information to the methodology on the process of developing the seasonal AOD from observations and the revised bias correction process using AERONET. We discuss the aerosol properties assumed in MODIS and MISR retrievals in Sections 2.1 and 2.2. In addition, we have incorporated the instrument AOD retrieval uncertainty into the bootstrapping process and acknowledge uncertainties related to these factors in Section 4.4 "Discussion of the remaining uncertainties". From that section:
 - "Some of the discrepancy between the dust AOD from models and observations is likely born out of simplifications in representing particle morphology and minerology and the resulting impact on the AOD. The models in this study assume a globally fixed refractive index for dust and either spherical or spheroid particle shapes. We do not quantify the uncertainty from mineralogy and morphology here; however, several studies have shown the influence of refractive index and shape upon the derived optical and radiative properties (e.g. Balkanski et al., 2007; Kalashnikova and Sokolik, 2004; Scanza et al., 2015). Scanza et al. (2015) estimate a reduction of approximately 6% on the global dust AOD when accounting for spatially varying mineralogy in the Community Atmosphere Model (CAM-5). Particle morphology and minerology may also present a general bias in AOD retrievals as well as the models. Simplified particle shape modeling during retrieval has been shown to cause underestimation of AOD from space-based retrievals and overestimation from ground-based observations (Kalashnikova and
- 20 Sokolik, 2002). Similarly, strongly absorbing dust can result in underestimation of the AOD, although improvements in MODIS Collection 6 have been shown to alleviate this (Hsu et al., 2013). The impact on the observational estimate of dust AOD will be dependent upon the specific assumptions made by the MODIS and MISR retrievals, both of which take particle non-sphericity into account but using different methodologies (see Sections 2.1 and 2.2 and references therein). Finally, potential biases exist via erroneous filtering of thick dust plumes during the retrieval (Baddock et al., 2016)."

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Specific comments

2, 8-9: please add a reference here.

Added

2, 14-16: Why PM2.5 in particular? You do not discriminate the size in your product.

Good point, thank you. We now just discuss in terms of PM

2, 24: It would be useful to mention already here what is the general strategy of the work, and why you will use all of the following data from observations or model. Maybe add a table or a brief description in the text, so that the reader can already have a better idea of the role of each type of data in this paper.

We have elaborated on the usage of the data products in the introduction to the data description:

"To derive an estimate of dust AOD we make use of AOD retrievals from three satellite instruments as well as surface-based sun photometers to provide a 'ground-truth' for correcting the satellite retrievals. We use in combination with four global aerosol models that provide information on a range of estimates for the non-dust aerosol AOD and the spatial distribution of dust aerosol (see Section 3 for a full description of the methodology)."

3, 15: the usage of the angstrom exponent is not clear, please rephrase.

Rephrased as follows:

- 45 "The wavelength-dependence of the AOD, described by the angstrom exponent (Ångström, 1964) between the AOD at 440 and at 870 nm, is used to distinguish AOD dominated by coarse aerosol that is indicated by a lower angstrom exponent than for fine aerosol (e.g. O'Neill et al., 2001; Reid et al., 1999)."
 - 4, 24: this sentence is not clear; also the reference is missing from the list.
- 50 Rephrased and reference added

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8,20-24: How is your central estimate derived? Is it the mean of the distribution derived from the set of all possible combinations of models and satellite data depicted in Figure 4? Also, please describe more in detail how all the combinations were constructed in the previous section.

We have clarified the methodology in the closing paragraph of the methodology description as follows:

"This process is repeated for all combinations of the 3 satellite instruments, 4 model estimates for non-dust, and 4 model regional-to-global scaling factors; this produces 48 realizations, 16 per satellite instrument, each with an uncertainty estimate.

We use the kernel density estimation method (Silverman, 1986) with a Gaussian kernel and standard smoothing to determine a probability density function for the global dust AOD based on the 48 realizations."

And clarified the description of Figure 3 in Section 4.1:

15 Figure 3 summarizes our observationally-constrained global dust AOD estimate, averaged over the 2004 – 2008 period, for the combination of all data and for each of the satellite instruments individually.

20 Comments from Paul Ginoux

This is a very nice work, which will provide a better constrained mean dust load and optical depth. Still, I wonder about some biases related to satellite data in general, and MODIS Deep Blue in particular. The authors note a lack of bias in MODIS AOD based on the scatter plot of daily values at AERONET sites. However, these sites are characterize by different aerosol environments and surface albedo. Uncertainties related to satellite retrieved AOD between sites will be different. In Figure 3 of Ginoux et al. (Rev. Geophys., 2012), you will notice very different biases between regions. For example Australia is biased high, while Africa is slightly biased low. Although this study was done with Collection 5.1, similar results are obtained with Collection 6, but with much more reduced bias in Australia. My point is that there is very little information we can extract from your Figure 2. A better approach would be to also plot seasonal variation at dusty sites (e.g. Tamanrasset, Birdsville, Solar Village. Dunhuang, etc.).

Thank you for your comments on this work. The simplistic bias correction method that we applied is certainly a source of uncertainty. In response to the review comments we have revised the methodology to apply bias corrections that are specific to each region. We have revised the bias correction of the satellite data to be regional, rather than global and to incorporate Marine Aerosol Network (MAN) daily AOD in relevant regions. We assess an uncertainty to the bias correction by calculating the standard deviation in bias corrections for each year with sufficient (>100) coincident satellite and AERONET retrievals. There are still significant uncertainties resulting in this methodology as the AERONET sites will not represent the entire region, but we believe this goes some way to addressing your concerns and improving the correction applied to each region.

40 (We did also attempt a bias correction that used the lag-correlation of the AOD from GEOS-Chem to propagate the bias correction to regions surrounding the AERONET site. However, this did not take into account the surface reflectance influence on satellite retrieval bias (which would not share the same lag-correlation). We did not believe there was enough justification for using this approach without also accounting for the influence of surface reflectance, which was out of the scope of this project).

I am also concerned about your method of temporal average of observations. If you consider only days with retrievals you will have a high bias, as you discard all days with dust being washout and rainout (low dust). This will be also true for AERONET data. But, it is uncelar which method you are using.

- 5 Apologies that this was unclear in the paper. We did indeed look at this impact. Firstly, we sample the models to the satellite retrievals and compare the resulting dust AOD with and without sampling. Globally, sampling increased the modeled dust AOD by <1% for MODIS Aqua and Terra and 1-2% when sampling to MISR. Even on a regional basis the sampling bias is less than a few percent. This is obviously only sampling the 'model world' so may not represent reality. If we use the MERRA reanalysis meteorological fields to compare the model dust AOD with and without cloudy regions (masking columns containing grid boxes with >50% cloud cover) we still don't see a large impact (2%), other than in the Gulf Of Guinea where cloud cover is persistent. Intuitively, we would expect there to be a bias between in-cloud and clear-sky dust but this suggests it is more balanced than expected over the timescales considered, probably through many compensating factors.
- We have added the following to highlight this potential bias and the work we did to test the effect by masking the model with satellite data and with MERRA cloud cover (pg11):
- "We calculate the modeled global dust AOD with and without masking to match the MODIS and MISR sampling, testing whether sampling affects the derived global dust AOD. We find negligible (<1%) changes in the modelled global dust AOD when sampling to the MODIS instruments and an increase of 1 2% when sampling to MISR. Therefore, we determine that sampling frequency is sufficient to represent the AOD in the regions considered. Furthermore, because the masking effectively removes cloudy regions, the very small change in the modelled global dust AOD indicates that there is no obvious bias in the global dust AOD when including regions within cloudy air masses, relative to clear-sky only. We also calculated GEOS-Chem global dust AOD after masking columns that have >50% cloud cover in any grid box, based on MERRA reanalysis. This causes the global dust AOD to increase by 2%, relative to when no masking is used, indicating that the difference between clear-sky and all-sky dust AOD is small. However, we acknowledge that poor representation of clouds in the reanalysis meteorology or potential satellite misclassification of heavy dust loading as cloud (Darmenov and Sokolik, 2009) could lead to a stronger perceived relationship between dust loading in cloudy and clear sky conditions."
- 30 None of the models simulate dust from agricultural regions or with dynamic vegetation. Their contribution is highly uncertain but may affect your results regionally.
 - This is certainly true. It will not be an issue inside the regions considered (as we do not use model dust AOD here) but it may contribute to errors in model dust in other regions that will affect the global scaling. We have added the following caveat to highlight this uncertainty:
 - "Finally, dust emissions schemes currently used in the models are unlikely to reproduce emissions where vegetation cover is variable and will not represent dust from agricultural regions (Ginoux et al., 2012). Therefore, it is expected that the tuned emissions in models will overestimate emissions from large, permanent dust sources to compensate and partially explain the bias towards African emissions."
- Finally, you are most likely using MODIS quality flag 3 (QA=3) aerosol products, as advised by Sayer et al. (2013). However, it is not a good choice over dust sources as clearly shown in Figure 1 of Baddock et al. (Geophys. res. Lett., 2015). This choice of QA=3 may induce a low bias, if you use all days rather than just days with QA=3. On the other hand, if you divide the sum of all valid AOD by the number of days with QA=3, you will again create a high bias. In fact, it may be very high in some areas. Take a look at the factor 10 difference of frequencies between QA=1 and QA=3 in Figure 1 of Baddock et al. (2015). Hopefully this will help improve your results. Paul Ginoux.
- Yes, the Baddock et al. work is very interesting. I spoke with Rob Levy about this, but unfortunately isolating QA=1 is not possible with the Level-3 product; that provides Quality-Assurance weighted AOD (giving a weighting of 1.0 to QA=1, 2.0 to

QA=2 and 3.0 to QA=3 data) and standard AOD using no weighting but excluding QA=0 data. The merged ocean-DarkTarget-DeepBlue Level-3 product that we use has QA=3 data over land and QA=1-3 data over ocean. However, we have included reference to the Baddock et al. paper to highlight this possibility and to make sure people using Level-2 data in future studies consider the findings of that work. Thank you.

Addition in the MODIS description:

"The merged Level-3 product uses QA=3 data over land and QA=1-3 data over ocean, where higher quality data is given commensurate weighting. Baddock et al. (2016) show that correlation between the frequency of high AOD and dust source location is actually improved when using only QA=1 data. For data to be considered QA>1 the standard deviation in AOD between 1km retrievals must remain below a threshold of 0.18. Therefore, some legitimate dust-influenced retrievals over source may be discarded when using the Level-3 merged product. However, this is a trade off in terms of improving the quality of the retrieval away from source regions."

Comments from Natalie Mahowald

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This is a potentially really important paper, with a sound methodology, for the most part. The issues come with the error analysis, which appears to substantially underestimate the errors. The paper also fails to provide context with previous studies. If these issues are fixed, the paper is likely to be extremely influential.

Thank you. We hope that the additions we have made in response to your comments and those of the referees have improved the paper significantly.

The main errors associated with knowing the dust aod come from: 1. errors with the retrieval algorithm, 2) spatial and temporal hetereogeneity in dust distribution, 3) spatial and temporal variability in dust composition and/or shape, 4) errors in detecting dust versus other aerosols or clouds. The authors seem to deal fairly well with the 4th of these, but seem to underestimate the errors in the other three. Please discuss the issues with the retrievals and all the problems with the retrieval algorithms. Are the algorithms making the same assumptions about dust properties? That would then add another error, which will be difficult to assess by just comparing different datasets. For example, if they assume all dust is one optical property, or spherical, or at particular altitudes, etc. Please describe these sources of errors.

We agree that the uncertainty has been underestimated. We now incorporate the retrieval uncertainty in the first stage of the bootstrapping process. When bootstrapping to create a seasonal mean AOD with standard deviation, we include the instrument uncertainty on the daily AOD, and combine the errors for each day used in the seasonal mean AOD. This does not alter the mean, but increases the standard deviation of the AOD at a specific location. However, we find that the final global dust AOD uncertainty increases by <5% because the uncertainty is dwarfed by other factors, primarily the uncertainty on the new regional AERONET bias correction and the uncertainty on the satellite seasonal AOD derived from bootstrapping (Table 2). We recognize that region-specific retrieval biases may exist and will be unaccounted for. This is now acknowledged in Section 4.4, a new section for unaccounted uncertainties.

The retrieval algorithm assumptions will affect how well spatial and temporal hetereogeneity in the dust distribution, composition and shape are accounted for. We now discuss the retrievals in more detail in Sections 2.1 and 2.2 for MODIS and MISR to show differences in assumptions:

"The MODIS retrieval algorithm uses a look-up table of surface reflectance for a set of simulated aerosol properties to determine the AOD that best represents the observed reflectance. For the Deep Blue retrieval, the most relevant to this study over dust-influenced regions, the assumed optical properties of the dust aerosol have a single-scattering albedo (SSA) between 0.87 and 1.0 for the look-up tables at 412 nm and 490 nm and a refractive index of 1.55 – 0.0i (at 670 nm). The Mie calculation uses an effective phase function, derived from comparison of the Sea-Viewing Wide Field-of-View Sensor (SeaWIFS)

instrument retrievals with AERONET, over the ocean to account for non-sphericity. Different locations and loading conditions trigger changes in the wavelengths used in the retrieval, more information can be found in Hsu et al. (2004, 2013)"

- "The MISR retrieval algorithm uses simulated TOA radiances using properties for eight particle types to determine the AOD.

 5 The optical properties of the two aerosol particle types corresponding to dust are calculated using the discrete dipole approximation and the T-matrix technique to account for particle non-sphericity (Kalashnikova et al., 2005; Martonchik et al., 2009)."
- The broad bias correction of satellite-retrieved AOD to AERONET may account for some of the biases in the retrieval, although this correction is very uncertain and we are neglecting that the AERONET retrieval is not perfect. We now account for the large uncertainty in the AERONET bias correction, propagating that through to the global dust AOD estimate.
- In the comparison of the MODIS, MISR and aeronet, what is the rms error? This error represents a combination of the spatial and temporal variability as well as errors in the retrieval algorithms, and needs to propagate into the error in your final estimate.

 5 As it stands, only the mean bias propagates into your error estimate, which will underestimate your errors. If I look at Moon et al., 2015, the error bar on individual retrievals in MISR are at least 30%: how can you claim smaller error than that in your results? You seem to be assuming that these errors will average out, but this seems unlikely and this assumption would have to be justified.
- We wouldn't necessarily expect the retrieval uncertainty to be similar to the 30% of Moon et al. as we are averaging over longer timescales and larger regions so should be able to beat down the uncertainty. However, as before there may be unaccounted biases. As mentioned above, we have revised the bias correction of the satellite data to be regional, rather than global and have added the histograms with statistics of the AERONET-satellite retrieval comparisons in the supplementary materials (Figures S1-S3). We now assess an uncertainty to the bias correction by calculating the standard deviation in bias
 corrections for each year with sufficient (>100) coincident satellite and AERONET retrievals. This propagates the uncertainty in the bias correction through the analysis and now accounts for half of the uncertainty in the global dust AOD (Table 2).
- You include a comparison of AOD across all sites in the world, with all types of aerosols. How does this comparison over just dusty regions compare? Is it better or worse, please explain.
 - Our regional comparison of satellite and AERONET AOD should address this concern. We have included histograms of the daily AOD comparisons within each region in Figure SI S3.
- 35 Dust is not homogeneous in chemical composition, size and thus optical properties, but the retrieval algorithms assume that they are. You should explicitly discuss this point, and you could bound the error from mineralogy using Scanza et al., 2015, which suggest for the CAM5, the impact of spatially varying optical properties depending on mineralogy is 0.002 out of 0.033 aerosol optical depth or about a 6% error (1 sigma). Then it would seem you would need to add all these errors to the total estimated error, without letting them cancel each other, and then it seems likely that you will get a reasonable value.
 - Thank you. We now include reference to Scanza et al. and discuss the added uncertainties from mineralogy and morphology (among other factors) in Section 4.4. However, it is not clear if the difference in global dust AOD when including mineralogy in CAM-5 (a decrease from 0.033 to 0.031) from Scanza et al. (2015) would bias our observational estimate higher or lower. Combining this error with our current estimate yields only a small increase; therefore, we simply highlight this uncertainty in the extra Section 4.4, added to discuss the uncertainties and biases that may not be captured by this work.
- The last comment is to consider how this estimate differs from previous model/data comparisons (e.g. Cakmur et al., 2007; Albani et al., 2014 or Balkanski et al. 2007). There are two main differences. Here the primary spatial and temporal variability relationships come from the satellite remote sensing data vs. model results in those papers. And secondly, because the first two

papers include comparisons to concentration and deposition data. To understand how important the second is, please provide a comparison of your 'constrained' AOD-implied concentration and deposition to available datasets. This can be done very simply, but just using, for example, the GEOSCHEM dust AOD to deposition to surface concentration relationships, and your inferred AOD at that grid box. That will allow you to do a very simple comparison and show that indeed, your approach is (probably) fairly consistent with the other datasets. It probably won't be completely consistent, since none of the models seem to be able to match the AOD, concentration and deposition data the same time. This information could be added to the supplemental material and referenced briefly in the text.

Thank you for this suggestion. We have produced an estimate of the dust deposition following the method you suggest above and have included comparison with deposition network data in Figures S10 and S11. We note that the result is strongly dependent upon the model used (GEOS-Chem). Also, we are producing regional dust AOD estimates that are not conducive to comparison with clustered deposition measurements. Indeed, the correlation is only slightly better than for the GEOS-Chem model, and not significantly so — in part owing to the reliance on the model AOD distribution and the model relationship between dust AOD and dust deposition. For this reason, it is not easy to compare with previous model studies. We have made sure to reference this work so that the reader can dig deeper into the underlying assumptions in the models and how they affect the model representation of concentration and deposition as well as the AOD.

An observationally-constrained estimate of global dust aerosol optical depth

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Abstract. The role of mineral dust in climate and ecosystems has been largely quantified using global climate and chemistry model simulations of dust emission, transport, and deposition. Global climate and chemistry models routinely simulate dust emission, transport, and deposition to provide estimates of the role that dust plays in climate and ecosystems. However, differences between these model simulations are substantial, with estimates of global dust aerosol optical depth (AOD) that vary by over a factor of 5. Here we develop an observationally-based estimate of the global dust AOD, using multiple satellite platforms, in-situ AOD observations and four state-of-the-science global models over 2004 - 2008. We estimate that the global dust AOD at 550 nm is 0.0303 ± 0.0056 (21σ), higher than the AeroCom model median (0.023) and substantially narrowing the uncertainty. The methodology used provides regional, seasonal dust AOD and the associated statistical uncertainty for key dust regions around the globe with which model dust schemes can be evaluated. Exploring the regional and seasonal differences in dust AOD between our observationally-based estimate and the four models in this study, we find that emissions in Africa are often overrepresented at the expense of Asian and Middle-Eastern emissions, and that dust removal appears to be too rapid in most models.

1 Introduction

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Mineral dust is a key component of aerosol, affecting climate through interaction with radiation, clouds and snowpack, human health through contribution to particulate matter (PM_{2.5}), and ecosystem health through nutrient transport and deposition. The direct radiative effect (DRE) of dust contributes ~30% of the total aerosol global mean DRE (Heald et al., 2014); however, there is significant uncertainty in the radiative forcing of dust, estimated to be anywhere between -0.3 and +0.1 Wm⁻² (Boucher et al., 2013), owing to large uncertainties in the anthropogenically-driven changes in dust (Ginoux et al., 2012; Heald and Spracklen, 2015), and the particle morphology and absorption properties (e.g. Balkanski et al., 2007; Mishra et al., 2008), and the dust size distribution (Kok, 2011; Kok et al., in review)(e.g. Balkanski et al., 2007; Mishra et al., 2008).

Dust concentrations are often highest in remote regions that are sparsely-monitored, leading to further uncertainty on the atmospheric burden and the associated radiative effects.

Dust aerosol can be transported far downwind of desert source regions, having a significant impact on the surface PM255 thousands of kilometers downwind (Prospero, 2007; Prospero et al., 2014; Zhang et al., 2013). This poses a significant health concern through cases of premature mortality from respiratory and cardiovascular disease that are attributed to aerosol exposure (Lim et al., 2012). Studies attempting to quantify the global premature mortality from aerosol exposure (e.g. van Donkelaar et al., 2006; Evans et al., 2013) highlight the strong contribution of dust to PM255 across large regions of Africa, Asia and the Middle East. Because of the lack of surface monitoring in dust influenced regions, those studies rely on satellite observations of aerosol optical depth (AOD), a measure of the column-integrated aerosol that is critical for understanding the radiative effect. Relating the AOD to surface PM256 requires information on the vertical distribution and aerosol speciation, generally obtained from models, which can introduce considerable uncertainty (Ford and Heald, 2015). Limited observations of global dust aerosol hinder our ability to estimate the full extent of the climate and air quality impacts of mineral dust.

To simulate the global dust aerosol-cycle, models must be able to predict the vertical dust flux from suitable regions and represent the evolution of the particle size distribution while the dust is transported and deposited out of the atmosphere (e.g. Kok et al., 2012). The AeroCom project, an intercomparison and evaluation of different aerosol models, provides a detailed evaluation of dust aerosol simulations from multiple models (Huneeus et al., 2011). There is a considerable spread in global dust AOD estimates from models ranging from 0.010 to 0.053 (yielding a mean of 0.028 ± 0.011) with and anan AeroCom "median model"—median estimate of 0.023 and standard deviation of 0.011. The uncertainty in the AOD highlights the underlying uncertainties in emissions, size distributions, lifetime, and optical properties. Even over the well-studied, most productive dust region of West Africa, climate models struggle to represent dust emission and their year-to-year changes (Evan et al., 2014).

An observationally-constrained estimate of dust AOD can thus provide a suitable valuable metric to holistically evaluate model dust emission, transport, and deposition, thereby helping constrain both the direct radiative effect and the role of dust in adverse health effects from exposure to PM_{2.5}. Here we derive such a metric, with consideration for the sources of uncertainty, and use it to highlight seasons and regions in which current global models deviate from the observations.

2 Data Description

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In this studyTo derive an estimate of dust AOD we make use of AOD retrievals from three satellite instruments as well-as surface-based sun photometers to provide a 'ground-truth' for correcting the satellite retrievals. We use in combination with four global aerosol models tohat provide information on range of estimates for the non-dust aerosol AOD and the spatial distribution of dust aerosol (see Section 3 for a full description of the methodology). We use observational data and model simulations over the 5-year period between 2004 and 2008, except when calculating biases between satellite and surface-based observations, for which we leverage a longer dataset between 20032 and 20132. Below we give a brief description of each instrument and model, and the products used.

2.1 Moderate Resolution Imaging Spectroradiometer (MODIS)

Two MODIS instruments are in sun-synchronous orbit aboard the Terra and Aqua platforms, making equatorial overpasses at 10.30am and 2.30pm local time (LT), respectively. Radiance measurements are made across 36 bands between 0.4 and 14 microns, with 7 channels used to retrieve the AOD at 550 nm. The wide swath (2330 km) allows almost daily coverage of the globe by both instruments at a native resolution of 500 m at nadir (2 km at swath edge), for the aerosol-relevant bands, with AOD reported at approximately 10 km x 10 km resolution (Level-2 product). The Collection 6 MODIS data includes a merged AOD product that combines retrievals over ocean, vegetated land surface (Dark Target), and bright land surface (Deep Blue) to maximize global coverage. The retrieved AOD (τ) is estimated to be accurate to $\pm 0.03 \pm 0.05 \tau$ over ocean (Remer et al., 2005), $\pm 0.05 \pm 0.15\tau$ over dark land surfaces (Levy et al., 2010) and $\pm 0.05 \pm 0.20\tau$ over bright surfaces (Hsu et al., 2006; Sayer et al., 2013). The quality-assured (OA) Level-2 AOD retrievals are aggregated on a daily basis onto a 1° x 1° grid (Level-3) with statistics, including cloud fraction and standard deviation. Throughout this study we use the Level-3 product. The merged Level-3 product uses QA=3 data over land and QA=1-3 data over ocean, where higher quality data is given commensurate weighting. Baddock et al. (2016) show that correlation between the frequency of high AOD and dust source location is actually improved when using only OA=1 data. For data to be considered OA>1 the standard deviation in AOD between 1km retrievals must remain below a threshold of 0.18. Therefore, some legitimate dust-influenced retrievals over source may be discarded when using the Level-3 merged product. However, this is a trade off in terms of improving the quality of the retrieval away from source regions. The MODIS retrieval algorithm uses a look-up table of surface reflectance for a set of simulated aerosol properties to determine the AOD that best represents the observed reflectance. For the Deep Blue retrieval, the most relevant to this study over dust-influenced regions, the assumed optical properties of the dust aerosol have a single-scattering albedo (SSA) between 0.87 and 1.0 for the look-up tables at 412 nm and 490 nm and a refractive index of 1.55 - 0.0i (at 670 nm). The Mie calculation uses an effective phase function, derived from comparison of the Sea-Viewing Wide Field-of-View Sensor (SeaWIFS) instrument retrievals with AERONET, over the ocean to account for non-sphericity. Different locations and loading conditions trigger changes in the wavelengths used in the retrieval, more information can be found in Hsu et al. (2004, 2013)

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2.2 Multi-angle Imaging Spectro-Radiometer (MISR)

The MISR instrument, aboard the Terra satellite platform, measures radiance over 9 camera angles with an equatorial overpass at 10.30am LT. The relatively narrow swath width (400 km) results in global coverage every 9 days, compared with 1 - 2 days by MODIS. MISR provides AOD at four wavelengths (446nm, 558nm, 672nm, 867nm) with about three-quarters of retrievals falling within 0.20 τ (but no less than 0.05) 0.05 or 0.20 τ of AERONET observations (we assume an instrument uncertainty of \pm 0.05 \pm 0.20 τ throughout this study) and reliable retrieval over bright desert surfaces (Kahn et al., 2010; Martonchik et al., 1998, 2004). In this study, we use the Level-3 daily 0.5° x 0.5° resolution gridded AOD product. The MISR retrieval algorithm uses simulated TOA radiances using properties for eight particle types to determine the AOD. The optical properties of the two aerosol particle types corresponding to dust assume a refractive index of 1.51 – 6.5x10 4 i and SSA between 0.971 and 0.994 (at 672 nm). The extinction is calculated using the discrete dipole approximation and the T-matrix technique to account for particle non-sphericity (Kalashnikova et al., 2005; Martonchik et al., 2009).

2.3 Aerosol Robotic Network (AERONET)

AERONET consists of a global network of Cimel Electronique CE-318 sun photometers, which reports AOD with a high-degree of accuracy leading to estimated errors of ~0.01 - 0.02 (Eck et al., 1999; Holben et al., 1998). Direct sun measurements are made every 15 minutes at 340, 380, 440, 500, 675, 870, 940 and 1020 nm and AOD is retrieved at all but the 940 nm channel, which is used to provide total column water vapor. We use Level 2.0 data that has been screened for clouds (Smirnov et al., 2000), and use+The wavelength-dependence of the AOD, described by the angstrom exponent (Ångström, 1964) between the AOD at 440 and at 870 nm, is used to distinguish AOD dominated by coarse aerosol that is indicated by a lower angstrom exponent than for fine aerosol (e.g. O'Neill et al., 2001; Reid et al., 1999). Sun photometer measurements made from aboard ship cruises as part of the AERONET Marine Aerosol Network (MAN; Smirnov et al., 2011) are incorporated into the AERONET analysis in this work.

2.4 GEOS-Chem

We use the GEOS-Chem global chemical transport model (v9-01-01; http://www.geos-chem.org) to simulate the coupled oxidant-aerosol chemistry of the troposphere at a resolution of 2.5° by 2.0° over 47 vertical levels following the specifications used in (Heald et al., 2014). The oxidant-aerosol simulation includes H₂SO₄-HNO₃-NH₃ aerosol thermodynamics described by ISORROPIA II (Fountoukis and Nenes, 2007) and coupled with an O₃-NO_x-hydrocarbon chemical mechanism (Park et al., 2004, 2006). The aerosol simulation also includes carbonaceous aerosols (Park et al., 2003; Pye et al., 2010; Pye and Seinfeld, 2010), mineral dust (Fairlie et al., 2007; Ridley et al., 2012), and sea salt (Alexander et al., 2005). Aerosol mass is transported in 4 size bins (0.1–1.0, 1.0–1.8, 1.8–3.0, and 3.0–6.0 µm radius) for dust, two for sea-salt and one for each of the other species.

The model is driven by assimilated meteorology from the NASA Modern-Era Retrospective analysis for Research and Applications (MERRA), which provides winds, precipitation, could cover etc. at 1-hourly and 3-hourly temporal resolution.

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Dust emissions are generated using the DEAD scheme (Zender, 2003) with the GOCART source function (Ginoux et al., 2001; Prospero et al., 2002) and a fixed soil clay fraction of 0.2. We follow Ridley et al. (2013) by using a probability distribution of sub-grid scale winds, generated from 0.5° x 0.67° MERRA 10-m winds, rather than the average wind speed when calculating dust uplift. Biomass burning emissions are provided by the Global Fire Emissions Database version 3 (GFEDv3; van der Werf et al., 2010). Anthropogenic emissions are provided by the Emissions Database for Global Atmospheric Research (EDGAR) v3.2 inventory (Olivier, 2001) for SO_x, NO_x, and CO which is superseded by the National Emissions Inventory (NEI99; http://www.epa.gov/ttn/chief/net/1999inventory.html) over the United States and Streets et al. (2003, 2006) over Asia (van Donkelaar et al., 2008). Sea salt emissions follow Gong (2003) with added dependence on sea surface temperature (Jaeglé et al., 2011). AOD at 550nm is calculated online assuming lognormal size distributions of externally mixed aerosols and is a function of the local relative humidity to account for hygroscopic growth (Martin et al., 2003). Aerosol optical properties are based on the Global Aerosol Data Set (GADS) (Hess et al., 1998a) with modifications to the size distribution based on field observations (Drury et al., 2010; Jaeglé et al., 2011) and improvements to the UV/visible refractive indices of dust (Sinyuk et al., 2003).

2.5 CESM

15 The Community Earth System Model (CESM), version 1.1 (Hurrell, 2013), is used in this study following the specifications described in (Kok et al., (2014b). The atmospheric component of the model, the Community Atmospheric Model version 4 (CAM4), is run at 2.5° x 1.9° resolution and is driven by ERA-Interim reanalysis meteorology (Dee et al., 2011) with free-running dynamics. CAM4 simulates aerosol as bulk species from the Model for OZone And Related chemical Tracers (MOZART) chemistry package (Lamarque et al., 2012), including sulfate, ammonium, ammonium nitrate, black carbon, organic carbon and secondary organic aerosol. Emissions of these species are prescribed by the AeroCom specifications (Neale et al., 2010). Sea salt is emitted and transported in four size bins and is calculated from 10 m wind speed (Mahowald et al., 2006). Dust emission in the Community Land Module version 4 (CLM4) is traditionally based on the DEAD dust scheme (Zender, 2003) with some minor modifications (Mahowald et al., 2006, 2010). Here we use the new dust emission model developed in (Kok et al., 2014a), which generates a vertical dust flux with no prescribed source function, and accounts for the exponential increase in dust flux with increasing soil erodibility. This dust emission model both better reproduces smallscale dust emission measurements (Kok et al., 2014a) and its implementation in CESM results in improved agreement against AERONET measurements in dusty regions (Kok et al., 2014b). Here we use an updated version of the dust emission model that generates a vertical dust flux with no prescribed source function, and that accounts for the exponential increase in dust flux with increasing soil crodibility (Kok et al., 2014a, 2014b). Dust is emitted into four size bins (0.1-1.0, 1.0-2.5, 2.5-5.0, and 5.0-10 µm diameter), and the fraction emitted into each bin is independent of wind speed, as shown by measurements (Kok, 2011b), and distributed following brittle fragmentation theory (Kok, 2011a). All aerosols are assumed externally mixed. and The aerosol optical properties are based on GADS (Hess et al., 1998a) with improvements to the dust optical properties

described in (Albani et al., 2014). And prescribed Assumed size distributions for bulk aerosolthat can be found in (Emmons et al., 2010). Emmons et al., (2010).

2.6 WRF-Chem

The quasi-global configuration of the WRF-Chem (version 3.5.1) model is used in this study, described in detail in Huet al. (2016). The simulation uses the MOSAIC (Model for Simulation Aerosol Interactions and Chemistry) aerosol module (Zaveri et al., 2008) with the CBM-Z (carbon bond mechanism) photochemical mechanism (Zaveri and Peters, 1999). A sectional approach is used to represent aerosol size distributions with eight discrete size bins and all major aerosol components including sulfate (SO₄⁻²), nitrate (NO₃⁻), ammonium (NH₄⁺), black carbon (BC), organic matter (OM), sea-salt, methanesulfonic acid, and mineral dust are simulated. The MOSAIC aerosol scheme includes physical and chemical processes of nucleation, condensation, coagulation, aqueous phase chemistry, and water uptake by aerosols. The model is run at a resolution of 1° x 1° (between 180° W-180° E and 67.5° S-77.5° N) with 35 vertical layers up to 50hPa (Hu et al., 2016). The modeled u and v wind components and temperature in the free atmosphere above the planetary boundary layer are nudged towards NCEP/FNL reanalyses on 6-hourly time steps (Stauffer and Seaman, 1990). Biomass burning emissions are derived from GFEDv3. Anthropogenic emissions are provided by the REanalysis of the TROpospheric (RETRO) chemical composition inventories (http://retro.enes.org/index.shtml) except over East Asia, where emissions are taken from the inventory developed for the INTEX-B mission in 2006 (Zhang et al., 2009), updated with SO₂ and carbonaceous emissions from Lu et al. (2011), and the United States, where the National Emissions Inventory (NEI) for 2005 are used. Sea salt emissions are based on (Gong, (2003), with added emission dependence on sea surface temperature (Jaeglé et al., 2011). Dust emission fluxes are calculated with the GOCART dust emissions scheme (Ginoux et al., 2001) and partitioned into the MOSAIC size bins based on brittle fragment theory (Kok, 2011a). Aerosol optical properties are computed as a function of wavelength for each model grid box. Aerosols are assumed internally mixed (a volumetric mean refractive index) in each bin. The Optical Properties of Aerosols and Clouds (OPAC) dataset (Hess et al., 1998b) is used for the shortwave (SW) and longwave (LW) refractive indices of aerosols, except that a constant value of 1.53 + 0.003i is used for the SW refractive index of dust following Zhao et al. (2010, 2011). A detailed description of the aerosol optical properties calculated in WRF-Chem can be found in (Fast et al., 2006) and (Barnard et al., 2010). The optical properties and direct radiative forcing of individual aerosol species in the atmosphere are diagnosed following the methodology described in Zhao et al. (2013).

2.7 MERRAero

The NASA Global Modeling and Assimilation Office (GMAO) GEOS-5 Earth system model can be run in a configuration that assimilates meteorological and aerosol properties retrieved from NASA Earth observing satellite platforms (Rienecker et al., 2011). The resulting aerosol simulation is termed MERRAero. The simulation is run at a resolution of 0.5° x 0.625° providing speciated AOD with 3-hourly temporal resolution. The aerosol processes are based on the Goddard Chemistry, Aerosol, Radiation and Transport model (GOCART; Chin et al., 2002) with coupling of chemistry and climate

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(Colarco et al., 2010). Dust, sulfate, organic carbon, black carbon and sea salt are simulated as external mixtures. Dust aerosol is partitioned into 8 size bins between 0.1 and 10 µm particle radius, and sea salt aerosol is partitioned into 5 size bins between 0.03 and 10 µm dry radius; all other aerosol is transported in a single size bin per species. Emissions of fossil fuels and biofuel follow the GOCART model (Chin et al., 2002) with updates in the U.S. following Park et al. (2003). SO₂ emissions are from the EDGAR-4.1 inventory with altered injection profiles (Buchard et al., 2014) and biomass burning emissions are supplied from the NASA Quick Fire Emission Dataset (OFED) Version 2.1. Sea salt aerosol production follows Gong (2003) with added dependence on sea surface temperature (Jaeglé et al., 2011). The aerosol optical properties follow GADS, but with modifications to reduce the absorption of dust at short wavelengths (Sinyuk et al., 2003), and extinction is calculated following Mie theory assuming spherical particles (Colarco et al., 2010). MERRAero differs from the other three models used in that the model assimilates AOD information from the MODIS instruments. The assimilation process is explained in detail in Buchard et al. (2016), here we give a brief description. The MODIS reflectances are cloud screened and converted to AOD using a neural net framework. The error covariance between the 2D MODIS AOD and the model AOD is used to generate 3D aerosol mass increments. Using a Local Displacement Ensemble (LDE) methodology, ensembles of isotropic displacements in aerosol mass around a central grid box are weighted based upon the reduction in the error. Different aerosol species can be perturbed in each vertical layer, e.g. to allow a plume to be shifted to better match the MODIS AOD, and therefore aerosol mass can vary independently for each species.

3 Methodology

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3.1 Derivation of dust AOD

Our aim is to provide seasonal dust AOD estimates, both global and regional, that are as independent from modeled dustestimates as possible. The methodology and development of associated uncertainty estimates is described in detail below, but
the general methodology is as follows: We rely primarily on satellite retrievals of AOD, which we bias-correct using dustdominated AERONET AOD retrievals. In order to partition the retrieved AOD in dusty regions between the component due
to dust and the component due to other aerosols, we use simulated estimates of non-dust AOD with several global models in
15 regions that are identified as contributing significantly to the global dust AOD. We rely primarily on satellite retrievals of
AOD, with correction by dust-dominated AERONET AOD retrievals. Model seasonal estimates of AOD from non-dust species
are used to extract the dust AOD in regions that are identified as contributing significantly to the global dust AOD. These
regions are defined such that they account for the majority of dust AOD, based on model estimates, and are shown in Figure
1. Finally, model dust AOD is used to estimate the fraction of dust AOD that is outside of the 15 dust-dominated regions,
thereby relate the regional dust AOD to the global dust AOD, providing global seasonal dust AOD estimates between 2004
and 2008, to give the multi-year seasonal averages. Our methodology accounts for a large number ofmany uncertainties,
including the satellite retrieval error, estimation of seasonal mean AOD, bias correction, modelled non-dust AOD and global

scaling factors. We discuss potential biases that are not accounted for the weaknesses of our methodology, and possible unaccounted for biases, in Section 4.4.

We aggregate Ddaily AOD data from MISR and both MODIS instruments (Aqua and Terra) are aggregated onto a 2° x 2.5° grid and averaged over 3-month periods to increase coverage and provide a consistent grid between model and observations. AWe use bootstrapping method (Efron and Gong, 1983) is used to estimate the random uncertainty in the seasonally-averaged AOD due to sampling uncertainty within each grid box. This is achieved by randomly sampling (with replacement) the grid box daily AOD n times, where n is the number of days with a retrieval in that 3-month period, and the mean calculated. This is repeated to provide build a probability distribution of the seasonal AOD for each grid box. We find that a log-normal distribution is a good approximation to the resulting seasonal AOD uncertainty distribution, so we retain the mean and standard deviation of this distribution as the mean and uncertainty on the seasonal $log_{10}(AOD)$ for each grid box.

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Although the bootstrapping method quantifies the random error in each grid box' seasonal AOD, it does not quantify or correct the systematic error (bias) in the AOD, Therefore, we use AERONET AOD as ground-truth to apply a bias correction to the satellite-retrieved AOD, with a focus on dust influenced regions. AERONET hourly AOD (interpolated to 550nm) is used to produce a morning (10am - noon, LT) and afternoon (1pm - 3pm, LT) average to compare with daily retrievals from aboard the Terra and Aqua satellites, respectively. We compare these at the native satellite retrieval resolution and choose a two-hour window to both cover the approximate range of the overpass times and to maximize the number of coincident AERONET and satellite AOD retrievals. We use all AERONET sites within the regions defined in Figure 1 and aggregate all data in each region for the comparison. For the regions encompassing ocean we also use AOD measurements from the AERONET Marine Aerosol Network (MAN) where available (see Figure 1). Note that this analysis is performed at the location of all the AERONET sites in Figure 1, not solely those within the 15 dust dominated regions we have identified in Figure 1. To better isolate cases in which dust influence likely dominates, we only consider AERONET retrievals when the angstrom exponent (AE) is less than 0.4 (e.g. Muller et al., 2003; Prasad and Singh, 2007). This removes observations when the AOD is dominated by fine mode aerosols that may skew the bias correction to regions strongly affected by anthropogenic acrosol and not dust (see Figure 1 for the location of the AERONET sites). We generate histograms of the daily log₁₀(AOD) from AERONET and each satellite instrument using data between 2003 and 2013 (see Figures S1 - S3) and present the Sstatistics of the bias and linear regression between AERONET and satellite retrievals are calculated for each region in Table 1for the seasonal (3-monthly) climatology between 2002 and 2012 and used to scale the slope and y intercept of the satellite AOD to match that of the co-located AERONET data (see Figure 2). Although 2004 - 2008 is the main period of study in this research we use 12+ years of AERONET and satellite retrievals to maximize the amount of data and better characterize the biases. More than 100 days of co-located data available are required for the bias correction to be applied to a region; a criterion usually met, except for MISR in some Asian deserts. The standard deviation of the bias correction is derived from each pair of daily log₁₀(AOD) in a region, and is used to propagate this uncertainty into the global dust AOD estimate (see below). If

not enough data is available, we apply a bias correction of 1.0 (i.e. no bias) with a standard deviation of $\pm 50\%$ to represent the uncertainty.

On average across all sites, both MODIS instruments show a slight high very little mean annual bias in AOD relative to AERONET retrievals with AE<0.4 (+1+8% for Terra; -2+4% for Aqua; see Figure 2Table 1), although there is considerable variability between regions and on a day-to-day basis (indicated by the correlation coefficient, r, in Table 1). The correlation between MODIS and AERONET is good (r=0.83 for both), and similar for MISR and AERONET (r=0.84). MISR is also biased high relative to AERONET (+137%), owing primarily to retrievals when AOD<0.5, but also exhibitings a low bias for AOD>1.0, consistent with previous comparisons, e.g. Moon et al. (2015). Splitting the data by season does not yield qualitatively different results; however, it reduces the number of data points in some regions enough to make comparison unreliable. Therefore, we apply an annual bias correction per region. There is no clear seasonality in the bias between the satellite and AERONET AOD for any of the instruments. Initially, we implemented a site-dependent bias correction using the spatial covariance of the AOD to propagate the bias correction away from AERONET sites; however, strong bias corrections that could not be confirmed often propagated from polluted regions into remote locations. Therefore, a single correction per satellite instrument is used globally owing to the sparse AERONET data in the regions most affected by dust. The bias correction has a negligible moderate impact on the average global dust AOD, decreasing it by 10% (1%), but and bringing the individual satellite instruments into closer agreement. However, the large uncertainty on the bias correction (see Table 1) is a major causes a 9% decrease insource of the uncertainty on the global dust AOD about that average as the satellite AOD retrievals are brought into closer alignment (see Table 21).

Although dust aerosol is often the main contributor to the AOD in the regions shown in Figure 1, other aerosol species can make a significant contribution and need to be accounted for to extract dust AOD from the satellite retrievals of AOD. We use GEOS-Chem, CESM, WRF-Chem and MERRAERO MERRAERO to provide non-dust AOD; using multiple models provides an estimate of the variability in the non-dust portion of the AOD resulting from uncertainty in aerosol emissions and formation mechanisms. Anthropogenic aerosol is generally well characterized by global models, especially on seasonal timescales, and have been regularly evaluated against observations, particularly in the Northern Hemisphere (e.g. Hu et al., 2016; Leibensperger et al., 2012; Liu et al., 2012; Mann et al., 2014). We focus on regions in which the dust AOD often dominates to reduce potential errors from biases in modeled non-dust AOD. Biomass burning aerosol concentrations are inherently uncertain because of the challenges in determining burned area and emissions factors (French et al., 2004; van der Werf et al., 2006). Despite considerable evaluation against observations the resulting biomass burning AOD is sometimes underrepresented (Matichuk et al., 2007; Reddington et al., 2016); therefore, we treat regions affected by biomass burning emissions with caution. In the regions analyzedanalysed, here the AOD is predominantly driven by dust aerosoldust aerosol plays a key role and often dominates in the spring and summer, limiting the influence of the model non-dust AOD. Exceptions to this are in South America, South Africa, and Australia, that have a minimal impact on the global dust AOD, and the Gulf of Guinea, where significant biomass burning aerosol is present (we consider results with and without these regions, see Table

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1). In addition, most regions considered in this study are inland and therefore sea salt aerosol will have a limited impact. Figure 23 displays the climatology of non-dust AOD and dust AOD for each model used, averaged over 2004 - 2008.

Figure 3 displays the climatology of non-dust AOD and dust AOD for each model used, averaged over 2004 - 2008.

For each 2° x 2.5° grid box, i, within the 15 regions we apply the AERONET-derived bias correction, α , to the seasonal satellite AOD, τ^{obs} , and subtract the model non-dust AOD, τ^{model}_{nd} , to provide an estimate of the regional dust AOD, τ^{reg}_{d} (Eqn. 1). We allow negative values of τ^{reg}_{d} so as not to introduce a positive bias. The uncertainty distribution for each of these three variables, (bias correction, satellite $log_{10}(AOD)$, and model non-dust $log_{10}(AOD)$); is sampled and the average dust AOD is calculated for each region. This process is repeated multiple times to yield a stable distribution of seasonal dust AOD ($\frac{1}{2}$ 00 times is sufficient for a robust average) for each of the regions between 2004 and 2008. For a single iteration of the dust AOD calculation we use the same random sampling (sampling the same number of sigma from the mean) for all grid boxes, thereby assuming the worst case scenario that the uncertainty is correlated spatially. If we use a different sampling of the uncertainty distribution for each grid box, the uncertainty on the global dust AOD drops by approximately a factor of 8.

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$$\tau_{d}^{reg} = \frac{1}{N} \sum_{i}^{N} \alpha_{i} \tau_{i}^{obs} - \tau_{nd,i}^{model} \tag{1}$$

The regional dust AOD, τ_d^{reg} , for the 15 regions is weighted by surface area, A^{reg} , summed, and scaled by the surface area of the Earth, A_E , to give the total regional contribution to the global dust AOD (Eqn. 2). To obtain the globally-averaged dust AOD, τ_d^{glob} , we calculate the ratio, β , between the modeled dust AOD across all regions and the modeled global dust AOD (Eqn. 3).

$$\tau_d^{glob} = \beta \frac{1}{A_E} \sum_r^{N^{reg}} A^{reg} \tau_d^{reg}, \tag{2}$$

$$\beta = \frac{\tau_d^{glob,model}}{\frac{1}{A_r} \sum_r^{Nreg} A^{reg} \tau_d^{reg,model}}$$
(3)

This allows the satellite estimate within the regions to be scaled to a global dust AOD estimate. This is the only element of our analysis that relies upon simulated dust AOD. The 15 regions account for between 83% and 95% of the global dust AOD, depending on the model, so the model influence is limited and using multiple models provides an estimate of the uncertainty this introduces into our analysis (see Table 1). This process is repeated for different_all_combinations of the 3 satellite instruments, and4 model estimates for non-dust, and 4 model regional-to-global scaling factors; this produces leading to 48 realizations, 16 per satellite instrument, each with an uncertainty estimate. We use the kernel density estimation method (Silverman, 1986) with a Gaussian kernel and standard smoothing to determine a probability density function for the global dust AOD based on the 48 realizations.

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4 Results

4.1 The observationally-constrained global dust AOD

Figure 4 summarizes our observationally constrained global dust AOD estimate, averaged over the 2004 2008 period. The global dust AOD for the AeroCom models in Huneeus et al. (2011) is also displayed in Figure 34 with the associated probability density function generated using the kernel density estimation method. Our observational estimate of the global dust AOD is centered around an average of $0.033 \cdot 030 \pm 0.0056$ (12 σ), and is thus much more narrowly constrained than the AeroCom estimate of 0.028 ± 0.011. All but 3 of the 48Over three-quarters (77%) of the realizations ensemble members (94%) fall above the AeroCom model mean global dust AOD; however, the broadness of the AeroCom model distribution implies that a global dust AOD greater than 0.035 would be required for statistically significant disagreement at the 95% confidence level (i.e. p < 0.05; in this case p = 0.6348). Relative to the dust AOD from the four models used in this study (see Figure 32), two all lie within 1\sigma of the observational estimate and two below. The average global dust AOD estimates from each satellite instrument are remarkably similar (MODIS Aqua: 0.030 ± 0.00433 , MODIS Terra: 0.030 ± 0.00433 , MISR: 0.030 ± 0.0062). This is partially owing to the AERONET bias correction: that decreases the AOD from all satellite instruments and brings them into closer agreement. however, even without bias correction the global dust AOD estimates are similar (MODIS Aqua: 0.031, MODIS Terra: 0.033, MISR: 0.035), indicating that the observationally constrained estimate is not heavily dependent upon the AERONET correction applied. The AERONET bias correction suggests that the satellite AOD is generally biased high in dusty regions, based on the available data for comparison in the regions of interest (see Figures S1 to S3). . This is partially owing to the AERONET bias correction; however, even without bias correction the global dust AOD estimates are similar (MODIS Aqua: 0.031, MODIS Terra: 0.033, MISR: 0.035), indicating that the observationally-constrained estimate is not heavily dependent upon the AERONET correction applied. On an annual basis, the observationally-constrained global dust AOD varies between 0.02831 and 0.0326, with good agreement in the interannual variability in dust AOD derived from the three instruments (Figure 54). The dust AOD is similar for years between 2004 and 2006 before increasing in 2007 and peaking in 2008, largely driven by a sharp increase across the Middle East (Yu et al., 2015). The AeroCom model simulations are representative of the year 2000; therefore, some of the difference between the global dust AOD in this study and that from the AeroCom study may derive from the interannual variability. However, the lowest annual global dust AOD, in 2005, still for each year equals or exceeds the AeroCom mean and median. Figure 4-3 and Figure 5-4 suggest that the global dust AOD aerosol-from models in this study is inis general agreement with the observational AOD constraints; whereas the models from the AeroCom study show more diversity. Hy underestimated in models, We note that the global dust AOD masks important regional differences that are discussed in Section 4.3. Furthermore, considerable uncertainty remains on the dust loading despite the similarities in global dust AOD as a result of compensating differences in dust emission, optics and aerosol size distribution assumptions (e.g. Albani et al., 2014; Balkanski et al., 2007; Cakmur et al., 2006). either via mass concentration or the representation of dust extinction. Indeed, a A follow-on study indicates that both the abundance and extinction efficiency of dust are underestimated in models and better constrains these factors to improve estimates of the dust impact on global

climate indicates that both these factors are underestimated in models (Kok et al., personal communication, manuscript in preparationunder review). However, the uncertainty in the observational estimate does encompass several of the models.

4.2 Uncertainties in the observational estimate of global dust AOD

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Table 1 summarizes the uncertainties considered in this study, both in terms of potential bias to the global dust AOD and the contribution to the standard deviation of the estimate (0.005). The latter is quantified by assessing the reduction in the spread of the global dust AOD PDF when the uncertainty for a factor is omitted. The leading uncertainty arises from the AERONET bias correction (α, Eqn. 1). The bias correction yields a decrease in the global dust AOD of 10% and brings the estimates from each satellite instrument into close agreement, but the uncertainty ofn the bias correction accounts for over half of the ultimate uncertainty on the global dust AOD. The instrument retrieval errors contribute 5% of the uncertainty, whereas estimating the seasonal satellite AOD from a limited number of retrievalsestimate that is based on a limited number of daily AOD retrievals within a season contributes 13% of the total uncertainty (±41%). The difference in regional-to-global dust AOD scaling from models (±19%) and the difference in non-dust AOD from models (±19%) both-yield ±6% and ±8% uncertainty, respectively, on the estimated global dust AODeontribute to the uncertainty. The latter uncertainty is primarily a consequence of higher non-dust AOD in MERRAero than the other three models and therefore a lower estimate of dust AOD. The uncertainty from non-dust AOD may not be symmetrical about the mean and is discussed further in Section 4.4 and in Supplementary Materials. The regional-to-global scaling factor (β , Eqn. equation 3) is strongly dependent upon the dust lifetime within the model and ranges from 1.20 to 1.45, a lower scaling factor indicative of less dust far from source and therefore a shorter dust lifetime. The uncertainty from the regional-to-global scaling may not be symmetrical about the mean if the model dust lifetime estimates are biased low, as analysis of dust outflow into the mid-Atlantic suggests (see later discussion). In that ease, the true regional to global scaling and the global dust AOD would be closer to the upper end of our estimate. We find that the AERONET bias correction (α, equation 1) plays a minor role in changing the global dust AOD (<1%) and reduces the uncertainty by bringing the different satellite estimates into closer agreement, as expected.

Other factors that are <u>explored</u>, <u>but</u> not encompassed by the uncertainty estimate on the global dust AOD₂ are the impact of spatial and temporal sampling biases in the satellite data (e.g. overpass timing and frequency, regions of persistent cloud, high latitudes), cloud filtering of satellite AOD retrievals, and inclusion of the Gulf of Guinea region. These are also included in Table 1 and discussed below.

Satellite retrieval of AOD is only possible in clear sky conditions and at locations that fall within the satellite swath; therefore, the observed dust AOD will not take into account the effect of dust present before or after the satellite overpass, and in the presence of clouds. We assess the impact of this sampling bias by processing the AOD from the 4 models in the same way as the satellite-retrieved AOD, including masking the daily AOD data where no satellite retrieval is available. By comparing the modeled dust AOD with and without masking, we determine that the impact of satellite sampling_upon the global dust AOD estimate is minimal, < 1% for the MODIS instruments and +1.3% for MISR. Masking does however increase the uncertainty in the dust AOD estimate by 7% when sampling is based on MODIS and 50% when sampling to the sparser

MISR retrievals. Because the masking effectively removes cloudy regions, the very small change in the modelled global dust AOD indicates that there is no obvious bias in the global dust AOD when including regions within cloudy air masses, relative to clear-sky only. We also calculated GEOS-Chem global dust AOD after masking columns that have >50% cloud cover in any grid box, based on MERRA reanalysis. This causes the global dust AOD to increase by 2%, relative to when no masking is used, indicating that the difference between clear-sky and all-sky dust AOD is small. However, we acknowledge that poor representation of clouds in the reanalysis meteorology or potential satellite misclassification of heavy dust loading as cloud (Darmenov and Sokolik, 2009) could both-lead to a stronger perceived relationship between dust loading in cloudy and clear sky conditions.

The sun-synchronous orbit of the Terra and Aqua satellites results in overpass at similar morning and afternoon local time, respectively, each day. Therefore, a significant daily cycle in the AOD would create a bias in the inferred daily AOD. For all dust-influenced AERONET sites, we compare the 10.00 - 12.00 LT and 14.00 - 16.00 LT AOD to the daily AOD (calculated from all available retrievals within the daytime) between 2002 and 2012. We find that, on the days with AE > 0.4 at the AERONET sites used in this study, the AOD during the morning and the afternoon are closely related to the daily AOD, deviating by < 2% on average. This is in agreement with Smirnov et al., (2002) that found AOD varied diurnally by less than 10% at dust-influenced AERONET sites. When the satellite retrieved AOD is bias corrected to the daily AOD, rather than the AOD at time of overpass (as done here), we find that the dust AOD is 4% lower (see Table 1).

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By filtering MODIS daily AOD 1° x 1° retrievals that contain more than 80% cloudy Level-2 pixels we find that the AOD drops considerably in the Mid-Atlantic, Gulf of Guinea and the Arabian Sea (Figure 56). This leads to significantly different estimates for the dust AOD in certain regions (Figure 67). The largest impact is seen in the Mid-Atlantic where the dust AOD declines by 40% on average when filtering for clouds. The models also decrease when the equivalent masking is applied, but only by 20% on average in this region. This suggests that the filtering preferentially removes higher dust AOD cases, but the association of high dust AOD with cloudy regions is stronger in the observations than in the model. Similarly, reductions in dust AOD of up to 30% in winter and spring are produced by cloud filtering in the Gulf of Guinea. In the southern part of the Arabian Peninsula and the Arabian Sea the summertime peak in dust AOD is decreased by 30% by filtering pixels with more than 80% cloud cover. Cloud filtering of Level-2 retrievals is generally considered conservative in Collection 6 and misclassification is more common for thin cirrus than cumulus cloud decks (Levy et al., 2013; Remer et al., 2012). Therefore, removal of large regions in which high dust loading is associated with cumulus and stratus clouds may introduce an erroneous negative bias. It is also possible that high AOD retrieval in cloudy regions is the result of hygroscopicity and 3-D cloud effects (Koren et al., 2007; Marshak et al., 2008; Quaas et al., 2010). Indeed, it has been shown in studies using AERONET that AOD can increase dramatically between clouds and may be mistakenly screened as cloud (Eck et al., 2014). While this is a legitimate AOD enhancement, we cannot expect the global models with >100 km resolution using assimilated meteorology to reproduce enhancements from near-cloud hygroscopic growth or 3-D cloud effects on scattering. The observational-estimate of dust AOD provided in our analysis does not include the extra cloud filtering: www rely on the screening provided as part of the MODIS retrieval, rather than arbitrarily filtering the cloud-cleared product. However, the AERONET bias correction does decrease the AOD substantially, especially in the Mid-Atlantic (up to a 20% decrease), and so may partially account for the higher AOD associated with cloudy regions. However, as the enhanced AOD in cloudy regions is unlikely to be captured by models, we have made available the cloud-filtered global dust AOD estimate for all regions.

The regions defined around the South American, Southern African and Australian deserts and the outflow cover relatively large areas that are only intermittently affected by dust (see Figure 1). This may increase the likelihood of misattribution of non-dust AOD as dust AOD. We find that including those regions in the analysis does not have a significant impact on the global dust AOD, increasing it by 2<1%, although it causes an 68% increase in the uncertainty. In contrast, including the Gulf of Guinea region increases the dust AOD by +76% and increases the uncertainty by 169%. The dust AOD in the Gulf of Guinea region is consistently higher in the observational estimate than the models owing to a combination of persistent cloud cover, high biomass burning emissions in winter that are not always captured by the models, and a lack of dust towards the equator in the models that may result from too efficient convective wet removal. To prevent an artificially high bias in the global dust AOD, we do not explicitly evaluate the Gulf of Guinea region in our estimates beyond the assessment of uncertainty in Table 1. This region is still accounted for in the global dust AOD via the regional-to-global dust AOD scaling (the 14 remaining regions account for 77% - 87% of the global dust AOD, depending on the model).

4.3 Comparison of modeled and observed regional dust AOD

Model dust emissions are often tuned to a specific annual global emission mass (Fairlie et al., 2007; Huneeus et al., 2011)* or scaled to a global AOD inferred from assimilations (Mahowald et al., 2006; Rasch et al., 2001). The annual global dust AOD derived from the models in this study show encouragingly similar interannual variability to the observationally-constrained estimates, even though the global dust AOD is generally underestimated (see Figure 45). However, tuning the models globally will not necessarily produce the right spatial and seasonal distribution. Here we use the observational constraints developed in this study to highlight regional and seasonal discrepancies between models and observations in an effort to isolate potential errors that affect multiple models. We compare the interannual variability globally and the seasonal dust AOD aggregated over broad regions for each of the models with the observational estimates from each satellite instrument (Figure 78a). We also compare the climatological seasonal dust AOD from each model with the range of the observational dust AOD for each region (Figure 78b). We provide regional disaggregation of these results in Figures 89 and 9 and summarize the seasonal observational dust AOD for each region in Table 340.

Broadly, in Figure 78 we see that the models, except MERRAero, overestimate the amount of dust AOD over Africa with respect to the satellite estimates. The models generally over-emphasize winter or spring dust at the expense of summer. This is especially the case for GEOS-Chem (highlighted in Ridley et al., 2014; see Fig. S4 therein) and for CESM, and likely a consequence of the lack of convectively-driven dust emissions that will be somewhat alleviated by new parameterizations (e.g. Pantillon et al., 2016). Switching the dust scheme in GEOS-Chem to a new parameterization that does not rely on an explicit source function (Kok et al., 2014a, 2014b) does not alleviate the seasonality issue in Africa, suggesting that the poor performance relative to the other models is likely the result of meteorology rather than the dust parameterization. Isolating the

dust AOD in sub-regions, we find that the models overestimate dust in the North Africa, West Africa, and Atlas MountainBodele/Sudan regions, while better matching but underestimate the dust AOD in the mid-Atlantic outflow region, although there is significant variability between the four models (see Figure 89 and 910 for the sub-regions).

In the models, dust AOD over North Africa is greater than observed and dust AOD over the mid-Atlantic is often lower than observed (see Figure 89), even when extra cloud filtering of the satellite retrievals is included. This yields a ratio of the dust AOD over Africa to that over the Mid-Atlantic of $3.12.46 \pm 0.2531$ for the models and 2.3042 ± 0.1628 based on observations (2.62 ± 0.16 and 1.63 ± 0.08 , respectively, with cloud filtering applied). The predominant direction of long-range transport of dust is across the Atlantic; therefore, the models are likely to be removing African dust too rapidly during transport. This is unlikely to be the result of too much dust mass concentrated at large particle sizes that sediment out rapidly, based on comparison between observed and modeled size distributions (Kok, 2011a). Instead, it may stem from the vertical distribution and mixing in the planetary boundary layer that can lead to increased dry and wet removal through proximity to the surface and co-location with precipitating clouds, respectively. Indeed, the choice of boundary layer mixing scheme can have a significant impact on long-range dust transport (Jin et al., 2015). The GEOS-Chem model dust lifetime over the Atlantic was shown to be 25-50% shorter than inferred from MODIS and primarily controlled by wet removal, that dominates over dry deposition in the mid-Atlantic region (Ridley et al., 2012). It is unclear whether this bias is connected to a poor representation of the Saharan Air Layer (SAL) at present model resolution or an unidentified source of systematic bias. Higher resolution simulations will be required to capture the structure of the SAL, which can act as a conveyor for dust across the Atlantic. Excessive removal of dust will bias modeled dust lifetime low and result in a conservative observational dust AOD estimate because of the regional-to-global scaling employed in this study. The range of model dust lifetimes results in 13% to 23% of the global dust AOD coming from regions outside of those considered explicitly in this study. This constitutes a $\pm 56\%$ (0.00186) uncertainty in the observational global dust AOD estimate (Table 1); therefore, based on the comparison of dust AOD acrossin the mid-Atlantic it is plausible that the actual global dust AOD is towards the upper limit of this uncertainty bound. While the model underestimation representation of transport and deposition of mid-Atlantic dust may not be a major factor in the global dust AOD, it could have important implications for the simulation of hurricane genesis and nutrient deposition in the Amazon.

The models consistently underestimate AOD over Asian desert regions throughout most seasons (Figure 8). The low bias is present across all models and in all seasons except fall, when dust AOD is relatively low. The greatest divergence between models and observations occurs in spring AOD peak at the Taklamakan desert in spring and in summer peak in the Thar desertsummer, located between India and Pakistanwhen dust emissions are at their peak. Only CESM and MERRAero capture the seasonality in the Thar region. Enhanced summertime c-oarse mode AOD retrieved at AERONET sites in Karachi and Jaipur, located on either side of the Thar desert, indicates that the models are likely missing dust emissions rather than the observational estimate being biased high. However, there is some discrepancy between the MODIS and MISR dust AOD during winter and spring. Some of the increase in MODIS dust AOD is present south of the Taklamakan desert, on the raised Tibetan Plateau. Although dust can be transported out of the basin onto the Tibetan Plateau (Chen et al., 2013) the magnitude

of the AOD reported by MODIS is unexpected; therefore, we expect the MISR dust AOD to better represent this region. The low bias in modeled dust AOD is less pronounced in the Gobi desert, where re is also considerable spread between observations in the Gobi Desert; however, the models are all biased low, except for spring when GEOS-Chem and WRF-Chem appear to capture the observed seasonal spring peak in dust AOD. However, Figure 8 indicates that there is considerable uncertainty between the observational estimates in the Gobi Desert,—Seasonality is well captured by most models in the Thar desert, located between India and Pakistan, despite the general lack of dust AOD relative to the MODIS retrieved dust observations (Figure 9).

In the Middle East there is a slight low bias in the models relative to the observational dust AOD, through a combination of a the model dust AOD is biasedsubstantial low bias in the Southern Middle East region, and a slightly low-high bias in the Northern Middle East and Kyzyl Kum regions relative to observations. We find general agreement between the modeled and observed seasonality, with few systematic biases across all models spring peak in the Northern Middle East region and summer peaks in the Southern Middle East and Kyzyl Kum regions. However, all but MERRAero overemphasize summer dust at the expense of winter in the Kyzyl Kum region. CESM produces too much dust in summer, relative to other seasons, driven by high dust AOD between the Southern Middle East and Kyzyl Kum regions (the Gulf of Oman) that is present, but weaker, in the satellite observations.

Considering the southern hemispheric regions, our analysis indicates that the simulated dust AOD is comparable in Australia and lower than observed in South Africa and South America. However, the uncertainty in the observational dust AOD is too large to draw quantitative conclusions about the model representation of dust in those regions.

Throughout the comparison we find that MERRAero generally provides better dust AOD seasonal agreementthat agrees better with the observational estimates, both in seasonality and magnitude, relative tothan the other models. This is expected as the MERRAero simulation involves assimilation of MODIS AOD retrievals (Buchard et al., 2015) and is therefore not independent from the observations to which we are comparing. Furthermore, MERRAero is also produced at a higher resolution than the other models (0.625° x 0.5°) which may further contribute to better representation of dust emissions owing to more spatially resolved surface winds. However, the magnitude of the global dust AOD provided by the MERRAero simulation is the lowest of the four models (0.027) and is 0.003 lower than the observationally-constrained estimate presented here (only just within the 12σ uncertainty bound) on the observationally constrained dust AOD estimate. The total global AOD for all species in MERRAero is 5% - 15% higher than the other models, while dust accounts for a smaller fraction of the AOD in MERRAero (20%) than the other models (24% - 26%). This can be interpreted in two ways: either the contribution of dust to the total AOD is conservative in MERRAero, or the observationally-constrained estimates are biased high owing to a persistent low bias of non-dust AOD in 3-three of the 4-four models.

For this reason, we present the observational estimate with a 2σ uncertainty to encompass the range of realistic global dust AOD estimates.

4.4 Discussion of the remaining uncertainties

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We endeavoured to account for the uncertainties and biases involved in estimating the global dust AOD from observations; however, eertain-uncertainties remain that are difficult to quantify within this study. Potential sources of bias stem from (1) the model non-dust AOD, (2) the model regional-to-global AOD scaling, (3) treatment of particle morphology and mineralogy in models and in the satellite retrievals. These may present additional biases in the observational estimate and contribute to the discrepancies between models and observations.

We use multiple models to represent the uncertainty in non-dust AOD. However, It is possible that the non-dust AOD in all models may be systematically biased high or low, which would bias the observational estimate of the dust AOD low or high, respectively. Comparison between modeled and observed AOD at the AERONET sites and MAN ship locations does suggest a low bias in the modeled total AOD in some of the regions considered, although there is no clear systematic bias in the models (see Figures S5 – S9). Comparison of model and AERONET AOD in low and high dust cases (using the model dust AOD to discriminate) suggests that two of the models are biased high and two biased low (Figure S4). Overall, the ensemble of models appears to underestimate the non-dust AOD; correcting this results in a 7% decrease in the global dust AOD estimate (0.028). However, the uncertainties involved in this method are such that we do not include the bias correction in our final estimate (see Supplementary Materials).

Modeled dust AOD is used as a scaling factor to determine the global dust AOD from the regional observational estimates. We use multiple models to represent the uncertainty, but there may be a systematic bias present, rather than the ±6% uncertainty presented (Table 2). If the over-zealous removal of dust in models, highlighted in the mid-Atlantic, is a global phenomenon then the models would predict too much dust in the source regions relative to downwind and yield a low regional-to-global scaling factor. Similarly, Finally, ddust emissions schemes currently used in the models are unlikely to reproduce emissions where vegetation cover is variable and will not represent dust from agricultural regions (Ginoux et al., 2012). If those emissions are substantial, then it is possible that tuned emissions in models overestimate emissions from large, permanent dust sources to compensate for the lack of agricultural emissions, which could partially explain model bias towards African emissions.

Some of the discrepancy between the dust AOD from models and observations is likely born out of simplifications in representing particle morphology and minerology and the resulting impact on the AOD. The models in this study assume a globally fixed refractive index for dust and either spherical or spheroid particle shapes. We do not quantify the uncertainty from mineralogy and morphology here; however, several studies have shown the influence of refractive index and shape upon the derived optical and radiative properties (e.g. Balkanski et al., 2007; Kalashnikova and Sokolik, 2004; Scanza et al., 2015). Scanza et al. (2015) estimate a reduction of approximately 6% on the global dust AOD when accounting for spatially varying mineralogy in the Community Atmosphere Model (CAM-5). -Particle morphology and minerology may also present a general bias in AOD retrievals as well as the models. Simplified particle shape modeling during retrieval has been shown to cause

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underestimation of AOD from space-based retrievals and overestimation from ground-based observations (Kalashnikova and Sokolik, 2002). Similarly, strongly absorbing dust can result in underestimation of the AOD, although improvements in MODIS Collection 6 have been shown to alleviate this [Hsu et al., 2013). The impact on the observational estimate of dust AOD will be dependent upon the specific assumptions made by the MODIS and MISR retrievals, both of which take particle non-sphericity into account but using different methodologies (see Sections 2.1 and 2.2 and references therein). Finally, potential biases exist via erroneous filtering of thick dust plumes during the retrieval (Baddock et al., 2016),

Finally, dust emissions schemes currently used in the models are unlikely to reproduce emissions where vegetation cover is variable and will not represent dust from agricultural regions (Ginoux et al., 2012). If those emissions are substantial, then it is possible that tuned emissions in models overestimate emissions from large, permanent dust sources to compensate for the lack of agricultural emissions, which could partially explain model bias towards African emissions.

5 Conclusions

To provide an observational constraint for the global dust AOD we use three satellite retrievals of AOD over a 5-year-eperiod, use AERONET observations to correct biases in the satellite retrievals, and use speciated aerosol AOD from four global chemical transport models to separate the contributions of dust and non-dust AOD. Throughout the analysis we use bootstrapping to retain a robust estimate of the uncertainty on the dust AOD. We determine the global dust AOD to be $0.03\underline{0}^3 \pm 0.00\underline{5}6$ ($2\underline{1}\sigma$), with nearly three-quarters (73%)all but 3 of the satellite-model combinations-ensemble members in this study yielding a larger dust AOD than the median of the 15 AeroCom models ($0.02\underline{8}9 \pm 0.011$) and all combinations greater than the AeroCom model median (0.023). The observational estimate narrows the likely range of dust AOD greatly by half from that presented by the model estimates. The observational dust AOD is presented constructed as seasonal averages for 5 years (2004 - 2008) across 15 regions, providing a dataset with which the broad performance of model dust schemes can be evaluated (summarized in Table 3, with further data available by request to the author).

AllTwo-of the four models used in this study are within the one standard deviation uncertainty of the global mean observational estimate. However, it is essential to evaluate models on regional and seasonal scales, at which we find considerable differences, the other two-fall below the average. Using the regional and seasonal estimates of dust AOD, we highlight three-four general discrepancies between the models and observations: (1) the dust AOD across most of North Africa is overestimated in the models; (2) the Asian and some-Middle-Eastern deserts are underrepresented overall, (3) modeled seasonality varies considerably between models but generally overestimates winter and spring dust at the expense of summer in Africa, and overestimate fall dust at the expense of spring in Asian deserts, and (4) removal of dust exported across the Atlantic appears to be too strong in the models, which may indicate a systematic underestimation of dust lifetimes. We have used the observationally-constrained estimate of dust AOD to isolate specific regions in which the models disagree with the observations. However, the underlying mechanisms for the discrepancies are unclear and may be driven by the assumed physical characteristics of the surface, by the representation of surface wind, by the subsequent transport and deposition, or

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likely a combination of all factors. Further research in the areas highlighted in this work is expected to improve model simulations, and hence future estimates of the radiative, human health, and biosphere interactions of mineral dust.

-Acknowledgements

This work was supported by NASA under grant NN14AP38G. J.F.K. acknowledges support from the National Science

5 Foundation (NSF) under grant 1552519. Chun Zhao is supported by the U.S. Department of Energy (DOE) as part of the Regional & Global Climate Modeling (RGCM) program.

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Tables

Region						•			
	AERONET bias correction and correlation coefficient								
	MODIS (A	Aqua)	MODIS (7	Terra)	MISR,				
<u> </u>	NMB.	<u>r</u> ,	NMB.	<u>r</u> ,	NMB.	<u>r</u> 4			
Mid-Atlantic	0.82 ± 0.07	0.93	0.86 ± 0.06	0.93	0.84 ± 0.04	0.98			
N. Africa	0.91 ± 0.22	0.79	1.01 ± 0.12	0.80	0.99 ± 0.09	0.83			
Gulf of Guinea	0.90 ± 0.10	0.84	1.01 ± 0.11	0.83	0.92 ± 0.10	0.82			
W. Coast	1.04 ± 0.17	0.80	1.04 ± 0.19	0.83	0.94 ± 0.10	0.84			
Mali/Niger	0.98 ± 0.17	0.86	0.95 ± 0.17	0.87	0.93 ± 0.12	0.86			
Bodele/Sudan	1.01 ± 0.19	0.68	0.99 ± 0.19	0.76	0.91 ± 0.07	0.76			
N. Mid-East	0.81 ± 0.10	0.75	0.86 ± 0.12	0.73	0.95 ± 0.08	0.74			
S. Mid-East	1.01 ± 0.11	0.77	1.06 ± 0.11	0.78	0.88 ± 0.06	<u>0.8</u> €			
Kyzyl Kum	1.02 ± 0.29	0.83	1.05 ± 0.22	0.82	1.19 ± 0.10	0.86			
Thar	1.03 ± 0.12	0.84	1.04 ± 0.15	0.84	1.28 ± 0.14	0.74			
Taklamakan	0.82 ± 0.17	0.66	0.98 ± 0.21	0.79	0.77 ± 0.16	0.44			
Gobi	0.98 ± 0.41	0.54	0.90 ± 0.42	0.45	0.66 ± 0.28	0.71			
S. America	0.85 ± 0.15	0.43	0.95 ± 0.22	0.18	0.56 ± 0.15	0.24			
S. Africa	1.44 ± 0.23	0.73	1.71 ± 0.27	0.74	1.08 ± 0.11	0.89			
Australia	1.02 ± 0.32	0.42	1.01 ± 0.28	0.43	0.92 ± 0.10	0.80			

Table 1 – Bias corrections applied to satellite AOD retrievals in each of the regions (see Figure 1) based on comparison with AERONET daily AOD between 2003 and 2013 (see Figures S1 – S3)

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	Relative bias in global	Relative contribution to	
Source of uncertainty	dust AOD	uncertainty	
Instrument retrieval uncertainty	<1%	+5%	
Satellite retrieval of seasonal AOD	≤1%	+ <u>13</u> 41%	
Model non-dust AOD	±5 <u>8</u> % <u>*</u>	+ <u>196</u> %	
Model regional-to-global scaling	±5 <u>6</u> %	+194%	
	<1 <u>-10</u> %	<u>+56%</u> -12%**	
AERONET bias correction	(<u>-4</u> + 6 %, <u>-10</u> + 1 % - <u>16</u> 8%)	<u>(+64%, +73%, +50%)</u> (3%,	
(MODIS Aqua, MODIS Terra, MISR)		2%, <1%)	
Satellite retrieval spatial sampling	<1%		
(MODIS Aqua, MODIS Terra, MISR)	(<1%, <1%, +1.3%)	(+7%, +7%, +50%)	
Satellite retrieval diurnal sampling	-4%	+2%	
(MODIS Aqua, MODIS Terra, MISR)	(-5%, -1%, -2%)	(-1%, +2%, +3%)	
Cloud filtering (>80%)	-13% <u>**</u>	<1%	
Inclusion of S.H. desert regions	<1 <u>2</u> %	+86%	
Inclusion of Gulf of Guinea	+ <u>76</u> % <u>**</u>	+169%	

Table 24 – Each source of uncertainty is assessed in terms of the impact upon the global dust AOD mean and standard deviation and any bias. The sign of the relative uncertainty indicates whether the uncertainty yields a bias about the average or is assumed symmetrical about the average. For the model non-dust AOD and regional-to-global scaling the bias is defined as the difference between the upper and lower estimate of the global dust AOD when the source of uncertainty is isolated. Italicized uncertainties are explored but not incorporated into the global dust AOD uncertainty estimate. Values here are for correlated errors between neighboring 2×2.5 degree grid cells; assuming errors within a region are uncorrelated (i.e. a different number of sigma from the mean for each grid cell in an iteration) yields ~8x smaller uncertainty.

* May not be symmetrical about the mean (see Supplementary Materials, Figure S4)

3 ** Acts to reduce the uncertainty by this amount, bringing satellite retrieval estimates closer together.** Relative to global dust AOD without AERONET bias correction

Region	DJF		MAM		<u>JJA</u>		SON		4
<u>Asia</u>	0.114	±0.017	0.237	±0.017	0.191	±0.027	0.094	±0.021	4
Mid-East	0.119	±0.012	0.201	±0.017	0.250	±0.021	0.129	±0.014	4
Africa	0.167	±0.007	0.291	±0.012	0.298	±0.017	0.196	±0.015	4
N. Africa	0.118	±0.011	0.219	±0.010	0.207	±0.016	0.151	±0.016	4
Mid-Atlantic	0.064	±0.013	0.106	±0.008	0.143	±0.005	0.084	±0.006	4
Mali/Niger	0.257	±0.019	0.441	<u>±0.022</u>	0.462	±0.044	0.277	±0.023	4
Bodele/Sudan	0.191	±0.006	0.339	±0.023	0.310	±0.018	0.212	±0.021	4

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W. Coast	0.180	±0.010	0.250	<u>±0.019</u>	0.365	±0.016	0.233 <u>±0.022</u>
S. Mid-East	0.123	±0.018	0.204	<u>+0.021</u>	0.330	±0.044	0.150 ±0.020
Kyzyl Kum	0.115	±0.017	0.176	±0.026	0.154	±0.034	<u>0.101</u> <u>±0.018</u>
N. Mid-East	0.112	±0.011	0.223	±0.011	0.164	±0.015	0.113 ±0.019
Thar	0.130	±0.029	0.238	±0.033	0.319	±0.029	0.135 ±0.037
Gobi	0.093	±0.022	0.192	±0.022	0.102	±0.035	0.047 <u>±</u> 0.021
Taklamakan	<u>0.119</u>	±0.013	0.275	±0.027	0.171	±0.026	<u>0.104</u> <u>+0.011</u>
S. Africa	0.097	±0.023	0.073	±0.022	0.059	±0.021	<u>0.114</u> <u>±0.040</u>
Australia	0.022	±0.016	0.008	±0.009	-0.005	±0.008	0.001 ±0.023
S. America	0.020	±0.017	0.000	±0.013	-0.012	±0.013	0.017 ±0.013

Table 3 – Observational estimates of the seasonal dust AOD in each region of Figure 1, averaged over 2004 - 2008. The first three rows show the seasonal dust AOD for the broad regions (grouped by color in Figure 1; Africa does not include the Gulf of Guinea region)

Figures

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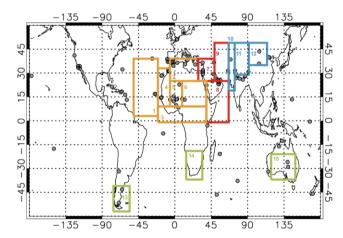


Figure 1 – The 15 regions considered explicitly in this study are defined. Regions are grouped into African (orange), Middle Eastern (red), Asian (blue), and Southern Hemispheric (green). AERONET sites used to bias correct satellite AOD are indicated with (gray circles). The regions are identified as (1) Mid-Atlantic, (2) N. Africa, (3) Gulf of Guinea, (4) W. Coast, (5) Atlas Mountains Mali/Niger, (6) Bodele Depression and Sudan region, (7) N. Mid-East, (8) S. Mid-East, (9) Kyzyl Kum, (10) Thar, (11) Taklamakan, (12) Gobi, (13) S. America, (14) S. Africa, and (15) Australia.

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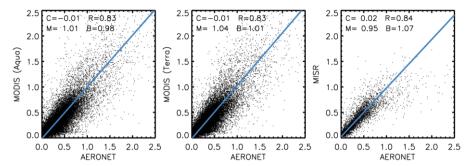
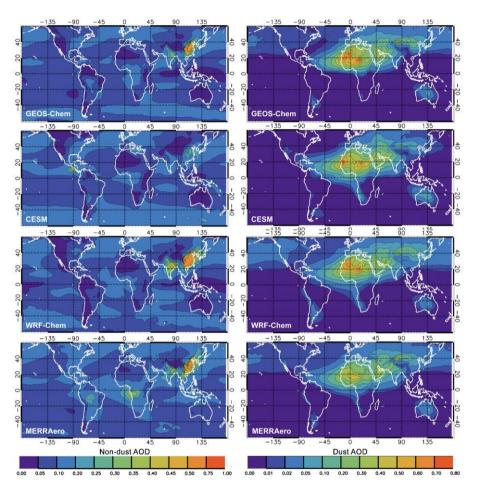
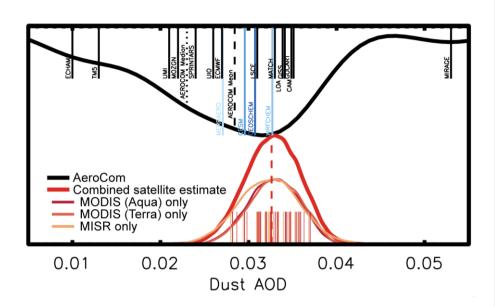


Figure 2—Comparison of daily AOD from collocated AERONET and satellite retrievals. Only days when AERONET angstrom exponent is < 0.4 are used to focus on dust-influenced measurements. The linear regression is shown for each, along with the slope (M), intercept (C), correlation coefficient (R) and bias (B). For MISR, two regressions are performed, one for AOD<0.5 and one for AOD>0.5.



 $Figure \ \ {\it 23-Annual\ } non-dust\ AOD\ (left)\ and\ dust\ AOD\ (right)\ at\ 550\ nm\ for\ the\ four\ models\ used\ in\ this\ study.\ Data\ is\ averaged\ over\ 2004-2008.$



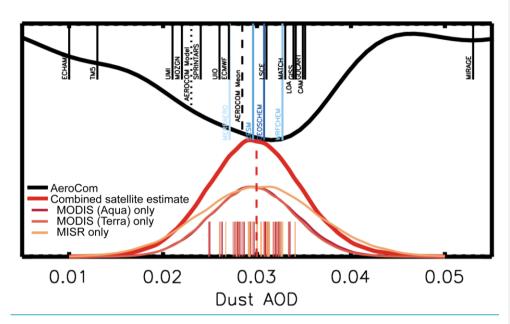
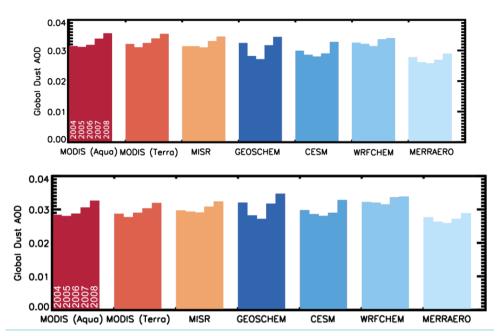


Figure 34 – The global dust AOD adapted from Huneeus et al. (2011) for 14 AeroCom models (vertical black lines) and the associated PDF (solid black line), mean (dashed black line) and the AeroCom median model (dotted black line) are shown along with the global dust AOD from the four models used in this study (vertical blue lines). The PDF of the observationally-constrained dust AOD estimate of this study (red) with the associated mean (dashed red line) is shown on the bottom axis. The PDF of the observationally-constrained dust AOD derived from each of the satellite instruments is shown (red hues) with the individual ensemble members (vertical red hue lines).



 $Figure \ \underline{45} - Annual\ global\ dust\ AOD\ for\ 2004 - 2008\ derived\ from\ the\ three\ satellite\ instruments\ (red\ hues)\ and\ from\ the\ four\ models\ (blue\ hues).$ The annual dust AOD\ from\ the\ satellite\ instruments\ is\ an\ average\ of\ the\ ensemble\ members\ for\ that\ instrument.

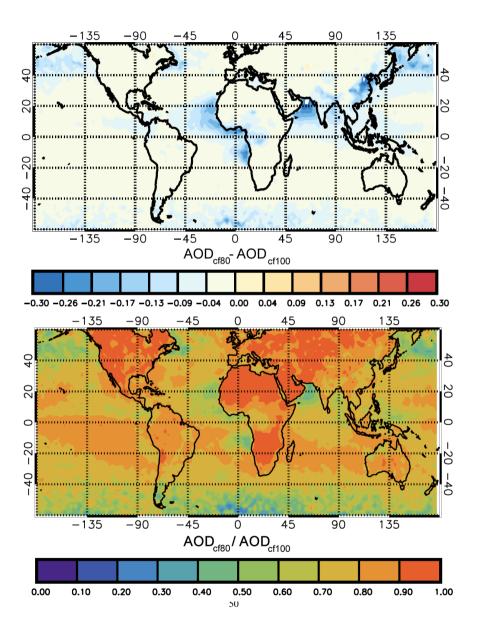


Figure $\underline{56}$ – The absolute change (top) and fractional change (bottom) in the annual MODIS Aqua AOD (averaged over $\underline{2004}$ - $\underline{2008}$) when applying a filter to remove any Level-3 data that contains more than 80% cloudy Level-2 pixels.

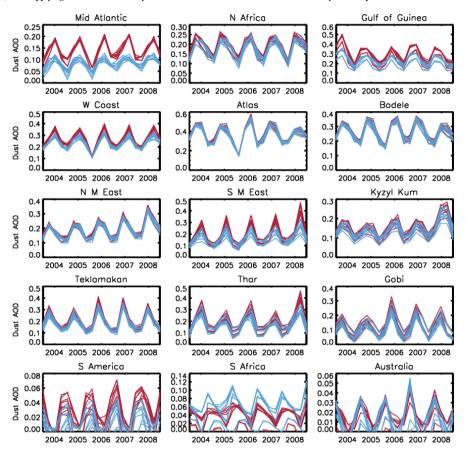
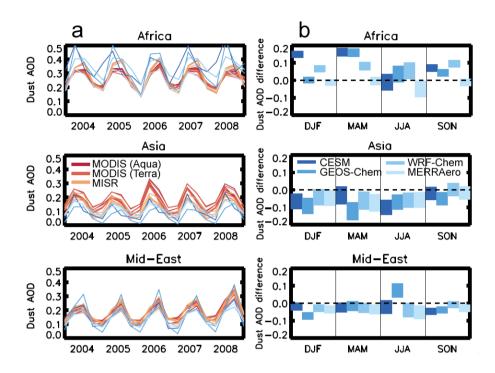


Figure 67 – Observational dust AOD from MODIS Aqua and Terra with (red) and without (blue) filtering of 1° x 1° daily regions with over 80% cloud cover. Each line corresponds to a different combination of satellite and model when calculating the dust AOD, indicating the uncertainty. Results are shown without any bias correction from AERONET.



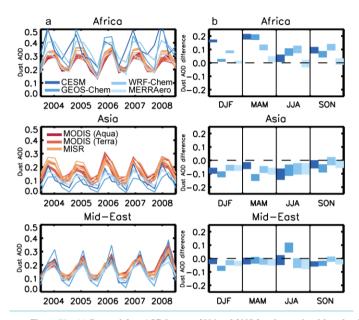
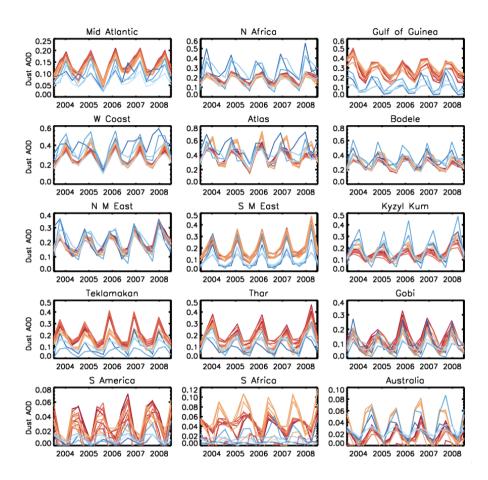


Figure 78 – (a) Seasonal dust AOD between 2004 and 2008 for observational-based estimates (red hues; each of the 12 lines represents a different ensemble member) and for the models (blue hues). (b) To isolate the seasonality, the difference between model and observational-based seasonal dust AOD, averaged over 2004-2008, is shown. Bar thickness indicates the range of the observational-based estimates for each season, deviation from zero (dashed line) indicates the bias in model seasonal dust AOD relative to the observations. The regions are based on area-weighted averages over the subset of regions defined in Figure 1, except Africa, which does not include the Mid-Atlantic region (shown separately in Figure 8 and 9 with other sub-regions).



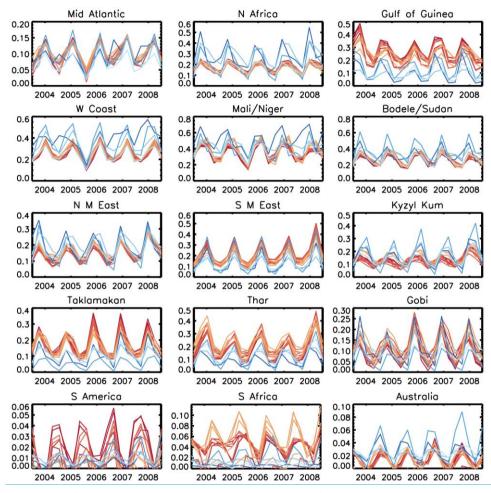
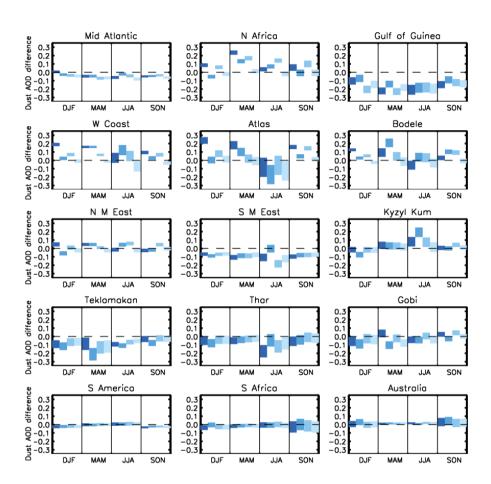
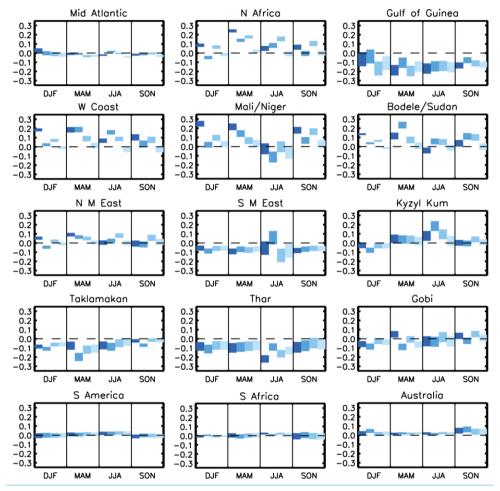


Figure 89 – Same as Figure 78a but for each individual region (in Figure 1). Observational dust AOD is shown for multiple realizations of MODIS Aqua, MODIS Terra and MISR (dark to light red). Models are GEOS-Chem, CESM, WRF-Chem and MERRAero (dark to light blue).





 $Figure \ \underline{940} - same \ as \ Figure \ \underline{78} b \ but \ for \ individual \ regions \ (in \ Figure \ 1). \ Models \ are \ GEOS-Chem, \ CESM, \ WRF-Chem \ and \ MERRAero \ (dark \ to \ light \ blue).$