Response to Anonymous Referee #1 Received and published: 24 October 2014

· Authors' responses appear as bullet points

Summary

Comments on "An overview of regional and local characteristics of aerosols in South Africa using satellite, ground, and modeling data" by Hershey et al. all refer to the PDF of the paper at http://www.atmos-chem-phys-discuss.net/14/24701/2014/acpd-14-24701-2014.pdf . The authors present a useful comparison between satellite aerosol optical depth (AOD) and surface particulate matter (PM2.5 and PM10) concentrations, but suffers from a lack of clarity in the discussion and in the figures. For these reasons, I think the paper requires Major Revisions before being published. I do think the core message – that column AOD from satellite data is not a useful proxy for surface PM2.5 in South Africa – is a publishable finding that builds on a body of research that stemmed from or led to the SAFARI 1992 and 2000 field campaigns.

General

Previous research:

The connection to previous research, such as the SAFARI 1992 and 2000 field campaigns, needs to be much clearer. For example, there are a number of studies of aerosol properties from the SAFARI 2000 JGR Special Issue that aren't not discussed in this paper, yet this paper states in the title that it is an "Overview." I included many relevant SAFARI 2000 papers below for reference and consideration (Maenhaut et al 1996; Magi et al 2003; Magi, 2009; Matichuk et al 2007; Reid et al 2005; Schmid et al 2003; Sinha et al 2003, etc.). In my thinking, an Overview implies that there is a body of work that can be referred to and that the current paper will talk about that and how new results fit into that body of work.

 The authors greatly appreciate the additional references to work from SAFARI-92 and -2000 that provide context for the results presented here. We have incorporated the references mentioned by the reviewer (in addition to several others), and have clarified the connections between results presented here and results from results cited from the SAFARI campaigns. Throughout the results and discussion section, we have expanded references to SAFARI publications to better place our results in the context of previous work in the region.

Writing seems too dense at times and I make a number of suggestions of places to separate the ideas in a more logical way. Namely, please avoid long paragraphs.

• Writing has been clarified throughout to highlight the main points more succinctly. All paragraphs longer than 20 or so lines have been divided into multiple paragraphs.

Figures need work to be acceptable for publication. I have many comments below.

 All figure suggestions have been adopted, and figures significantly revised. The authors thank the reviewer for comments that have made the figures much more effective in communicating our main results.

Specific

ABSTRACT:

Seems excessively long. Could you pare this down?

· Abstract has been rewritten to be substantially shorter and clearer

p. 24704, line 5: The first and second sentences should be heavily supported by citations to literature that substantiates these assertions.

• The statement was unnecessary and heavy support of citations would have been cumbersome in the text. The statement has been removed.

p. 24707, Section 2: What about AERONET? It seems that discussions such as Eck et al. JGR 2003 and Queface et al. Atmos. Env. 2011 address issues of surface AOD vs satellite AOD.

 Great idea, and it's something we've explored. We have looked closely at AERONET vs satellite comparisons for the Johannesburg area, and the results are quite interesting. AOD is consistently higher from AERONET, suggesting that surface AOD measurements are able to capture the boundary layer aerosol, but that satellites miss it. Ängström exponent shows very poor agreement - especially in winter - with surface measurements consistently higher during winter. This also suggests that ground measurements see BL aerosol that satellites miss. A full discussion of these results will appear in a separate manuscript, and our focus here remains on satellite AOD/Ängström/UVAI versus ground PM concentrations.

p. 24712, lines 1-2: Need a citation for this sentence. A peer-reviewed citation for the next paragraph would be useful too.

• The paragraph has been shortened considerably, and a reference has been included.

p. 24713, line 25: Figure 2 does not show fire counts.

• The text referred to a previous version of the figure, and has been edited to reflect integrated fire radiative power.

p. 24714, paragraph from line 1 to 25 is enormous. Can this be presented any differently? Seems there are multiple ideas in the same paragraph.

• Agreed. The paragraph has been divided into 3 separate parts.

p. 24714, line 26: Domestic burning has no effect on column aerosol? But aren't you referring to AOD that are not necessarily measured in domestic burning areas? Do you have column AOD measurements from the surface in an area with high levels of domestic burning that would support this claim (through a closure study)? I don't understand.

• This statement is out of place. Later in the text is an extensive discussion of agreement between satellite AOD and ground-based PM concentrations. We have removed the statement in question and have left the conversation about satellite vs ground measurements until later in the text.

p. 24715, line 9: I don't understand this discussion of correlation with CWV, or at least I thought I did until the last 2 sentences which sound like they aren't related to

CWV. Could you clarify? Are your findings consistent with past hygroscopicity studies of biomass burning particles by Chan et al. (2005), Reid et al. (2005), Magi and Hobbs (2003)?

- The last 2 sentences were out of place due to a formatting error. This has been corrected.
- The authors appreciate the context of Chan et al. (2005), Reid et al. (2005), and Magi and Hobbs (2003), but unfortunately the lack of correlation between CWV and AOD does not allow for any conclusions to be drawn about the hygroscopicity of biomass burning aerosols.

p. 24712, line 25: This correlation between Terra MODIS, Aqua MODIS, and MISR of r > 0.7 between AOD550 and AOD555 could be falsely read to sound like AOD dependence on wavelength is driving that relatively low r. You need to make clear that the first sentence of this paragraph is driving that low r value. MISR and MODIS are fundamentally different AOD measurements. I don't really understand why it is necessary to worry about AOD550 or AOD555. I doubt that just making a conversion to 550 or 555 using an assumed Angstrom exponent will have any significant effect on your analysis, and this would alleviate the unwieldy use of two marginally different wavelength AOD retrievals. My request would be, then, adjust AOD550 to AOD555 via Angstrom exponent or vice versa and then move forward. Many studies have done this in the past (for example, Schmid et al 2003).

• Excellent point. We have converted all MISR AOD555 values to AOD550, using Ängström as suggested. None of the AOD averages changed, nor did the correlations. We have added a line at the beginning of the AOD section stating that AOD is AOD550, and have clarified the paragraph in question.

p. 24716, line 18-19: Not a hotspot for particulates? This seems to contradict the point that surface air quality is quite bad at times, but that satellite AOD does not show this.

• Yes, this statement was unclear. It has been revised to state clearly that while NOx column is high, AOD is low, and that this is in contrast to other megacities.

p. 24722, much of text: Seems you should be referencing Magi (2009) here.

• I don't think Magi et al. (2009) fits in with the ground-based PM results on p. 24722. I think the reviewer *may* have meant 24714, where BB impacts on AOD are discussed. We did neglect to include that reference at two points in that section, and have added it.

p. 24723, lines 10-25: Very interesting, but I would caution against antecdotal evidence like this in a science paper. You do not cite evidence supporting this such as emissions factors from the cook stove (for example, Bond et al 2004), or taxi density patterns, and you have not conclusively made the case that power plants and/or industry are not contributing in some way. Please minimize this discussion, or temper the discussion to ensuring the reader that you have not gathered evidence to verify this hypothesis.

• We have shortened this discussion substantially, and have made it clear that it is a qualitative result based on anecdotal evidence of commercial activity. We have also referenced Bond et al. (2004) to support the statement that open flames emit more particulates.

p. 24724-24725: This is a very large paragraph and hard to follow, but it is critically important to the findings. I think there is material for about 5 paragraphs in there and you should break this apart accordingly. A more substantive comment: Do other studies (other parts of the world or South Africa) find something like this negative correlation

between surface PM and column AOD? Very interesting finding!

• The paragraph has been divided into 5 (as suggested) and clarified to highlight the main points. We have also noted that this is the first study (to our knowledge) to show such categorical and generalized disagreement between remotely-sensed aerosol parameters and PM concentrations at the ground.

p. 24727, line 11: Citation for NO2 assertion? You said this megacity idea before and cited a couple of studies, but please cite again here.

· This entire statement has been removed, as it is unnecessary and unclear

p. 24727, line 12: Why say satellite data suggest area is not a "major regional source of particulates" when you also say that satellite data is next to useless to prove this? Are you trying to point out this discrepancy? If so, please clarify the text.

This entire statement has been removed, as it is unnecessary and unclear

p. 24727, line 21: Citation for "previous studies"?

Citation added

Fig. 1: Really nice figure. Darken up the lines. It's very faint.

• Figure has been revised with darker lines

Fig. 2: Why FRP? Why not MODIS fire counts since you aren't really directly talking about the fire characteristic like high temperature combustion vs low temperature combustion?

 Very good question, and we went back and forth about whether to use fire counts or FRP. In the end we concluded that FRP was more relevant to total particulate emissions, so we went with that. That's the nutshell explanation. To expound, fires are detected during every overpass i.e. approximately twice daily. So the fire count contains no information about the history of a fire pixel or its burn duration, it is simply a snapshot at the time of the overpass. The fire pixel is approximately 1sqkm (the resolution of the MODIS imagery used for FIRMS and they say that in normal conditions, a fire pixel will be identified if the fire is larger than 1000 m2, however no information is derived about the distribution of the burn within the fire pixel. They also say that in clear conditions (low smoke, flaming fire, homogeneous land surface, near nadir) that a fire will be flagged even if it is as small as 100 m2. Once a fire pixel is flagged, it is saturated i.e. you cannot tell if there are 2 separate fires burning or one larger one. The issue of intensity factors into the detection limits also: however the limit is effectively a function of both size and intensity and even then it is not a hard boundary. This reference has a figure which shows the relationship: Giglio, L., Descloitres, J., Justice, C. O. and Kaufman, Y. 2003. An enhanced contextual fire detection algorithm for MODIS. Remote Sensing of Environment 87:273-282. doi: 10.1016/S0034-4257(03)00184-6. Regarding multiple small fires in a pixel: if they are numerous within a pixel there is a higher chance that they will be detected and once detected, they will be in the archive that we have used. I think this is one of the benefits of using the FRP since we can say that as long as it is detected, then FRP should be a reasonable proxy for particulate emissions at least to a first order.

Fig. 9: I don't understand this figure. Please elaborate in the caption or the text

The figure describes the frequency with which air mass arriving in Gauteng originates at a
particular location in the previous 3 days. So essentially it is just showing where air in Gauteng
province is typically coming from during each season. We have added the following text to the
caption: "Colors correspond to percentage frequency with which air mass arriving in Gauteng
originates at a particular location within the previous 3 days."

Fig. 10: Hard to tell what the lines in the bottom row correspond to in the legend

• The bottom row has been remade with different color lines for each type. It should be easy to distinguish now.

Fig. 11: This figure is very busy and hard to follow. Please break it apart into multiple figures so it's easier to read, or digest this so I know what to see. Lines in bottom row of plots hard to read with respect to the legend. Units on y-axis should be confirmed – says "normalized" but has physical units.

• The figure has been split into 3 separate figures, which are referenced in their respective places in the text. Thanks for catching the units - units have been removed and only the "Normalized" axis label appears.

Fig. 12: this seems really interesting and highlights an important result. Can you modify the figure so that it is easier to read? do any of those correlation coefficients indicate a statistically significant fit?

• The figure has been edited so that the correlation coefficients are larger and easier to read. Only relationships with Ängström exponent (except at Industrial sites) are statistically significant. The statistical significance of relationships has been noted in the text.

Fig. 13: Nice figure, but I can't tell which line corresponds to the legend. Why not produce this figure for the other seasons?

• The figure has been remade with new line types that are easy to distinguish. We made the figure for only Winter because the result is the same for all seasons (satellites pass over during minima in ground PM concentrations). Producing it for all the seasons would be redundant, and we feel it might be cumbersome. If the reviewer feels it is necessary for completeness, we may reconsider; for now we have left it with just Winter.

REFERENCES

Bond, T. C., D. G. Streets, K. F. Yarber, S. M. Nelson, J.-H. Woo, and Z. Klimont, 2004: A technology-based global inventory of black and organic carbon emissions from combustion. Journal of Geophysical Research: Atmospheres, 109, D14203.

Chan, M. N., M. Y. Choi, N. L. Ng, and C. K. Chan, 2005: Hygroscopicity of Water Soluble Organic Compounds in Atmospheric Aerosols:âAL' Amino Acids and Biomass Burning Derived Organic Species. Environmental Science & Technology, 39, 1555-1562

Maenhaut, W., I. Salma, J. Cafmeyer, H. J. Annegarn, and M. O. Andreae, 1996: Regional atmospheric aerosol composition and sources in the eastern Transvaal, South Africa, and impact of biomass burning. Journal of Geophysical Research: Atmospheres, 101, 23631-23650.

Magi, B. I., and P. V. Hobbs, 2003: Effects of humidity on aerosols in southern Africa during the biomass burning season. Journal of Geophysical Research: Atmospheres, 108, 8495.

Magi, B. I., P. V. Hobbs, B. Schmid, and J. Redemann, 2003: Vertical profiles of light scattering, light absorption, and single scattering albedo during the dry, biomass burning season in southern Africa and comparisons of in situ and remote sensing measurements of aerosol optical depths. Journal of Geophysical Research: Atmospheres, 108,

Magi, B. I., 2009: Chemical apportionment of southern African aerosol mass and optical depth. Atmospheric Chemistry and Physics, 9, 7643-7655.

Matichuk, R. I., P. R. Colarco, J. A. Smith, and O. B. Toon, 2007: Modeling the transport and optical properties of smoke aerosols from African savanna fires during the Southern African Regional Science Initiative campaign (SAFARI 2000). Journal of Geophysical Research: Atmospheres, 112, D08203.

Reid, J. S., R. Koppmann, T. F. Eck, and D. P. Eleuterio, 2005: A review of biomass burning emissions part II: intensive physical properties of biomass burning particles. Atmospheric Chemistry and Physics, 5, 799-825.

Schmid, B., and Coauthors, 2003: Coordinated airborne, spaceborne, and groundbased measurements of massive thick aerosol layers during the dry season in southern Africa. Journal of Geophysical Research: Atmospheres, 108, 8496.

Sinha, P., P. V. Hobbs, R. J. Yokelson, D. R. Blake, S. Gao, and T. W. Kirchstetter, 2003: Distributions of trace gases and aerosols during the dry biomass burning season in southern Africa. Journal of Geophysical Research: Atmospheres, 108, 4536. Interactive comment on Atmos. Chem. Phys. Discuss., 14, 24701, 2014.

Anonymous Referee #2 Received and published: 9 December 2014

Authors' responses appear as bullet points

Summary

Review report on "An overview of regional and local characteristics of aerosols in South Africa using satellite, ground, and modeling data" The authors present an overview of particulate air quality across some areas of areas of South Africa using satellite and ground-based data. The authors built their conclusion on level 3 data which is coarser than level 2 data. So primarily I suggest using level 2 data which has better resolution and average data on about 75-100 km and compare the results. Also the results and discussion section needs to be improved as it too many details from literature and sometimes discussions are not in concurrent with figures. Accordingly I suggest that the manuscript can be published with major correction. Here are some points which are needed to be fixed.

- Regarding level 2 versus level 3 data, the only conclusion we draw that might possibly be different with level 2 data is the poor agreement between satellite AOD and ground PM based on spatial resolution. Use of level 2 data would require a complete re-analysis of satellite data and ground-satellite correlations, which we have not done for the following reasons:
 - First, the discrepancy between satellite aerosol data and ground-based PM concentrations is primarily due to vertical inhomogeneity with stratified aerosol layers aloft (previous studies cited in the text). Level 2 data makes no difference here.
 - Second, the localized gradients between sites of different types occur on orders of 10 km or less. Level 2 data are still an order of magnitude coarser than these gradients.
 - Finally, we did perform an abbreviated analysis of AOD and Ängström exponent with level 2 data, and calculated correlation coefficients with ground-based PM concentrations. Level 2 data displayed no better agreement than level 3 data, and annual means were no different with level 2 data.
 - So while theoretically it may make sense to use level 2 data, a complete re-analysis of our data is not warranted. We have added a note in the text that we did look at level 2 data, but that agreement was no different.
- The abstract and results and discussion have been abbreviated substantially, and our main conclusions have been stated more clearly. Where there was unnecessary detail, it has been removed. The paper should now read more succinctly. Discussion of figures has been improved to make sure it is aligned properly with figure mention, and figure labeling has been improved.

General

Page 24702 Line 8, do you mean AOD from MODIS Aqua and Terra ?, please clarify

• Yes, and this has been clarified.

Page 24702 Line 9, the same for Ängström Exponent do you mean MODIS Aqua and Terra?

Please clarify

• Yes, and this has been clarified.

Page 24702 Line 25, too much details, I would place put the sites description somewhere else rather than abstract

• This has been shortened substantially, and the descriptions are left to the results section. We only name the types in the abstract, as necessary to present main findings that follow.

Page 24702 Line 28, the statement "PM10 concentrations in." is too long and not clear. Split it and make it clear

• The statement has been split and clarified.

Page 24703 Line 11 instead of "- and underscore" change it to "which reflects.."

Change made.

Page 24703 Line 13, make this statement shorter as it is too long" These results from the urban/ industrial Gauteng area quantitatively conïn Arm ...", summarize.

This has been summarized and clarified.

In general the abstract is too long and has many details that should be removed, I suggest rewrite the abstract in a more proper way.

• The abstract has been substantially shortened, and details beyond the main conclusions of the paper have been removed. It now reads clearer and more succinct.

Please identify the objective of research at the end of introduction section I a clear way.

• A clear statement of objective has been added to the end of the introduction.

Page 24707 Line 27, please rephrase "MODIS data included daily. . .", it has something missing..l

This has been rephrased and clarified

Page 24708 Line 4 The same of "Data from MISR included.." it has something missing, rephrase.

This has been rephrased and clarified

Page 24707 Line 27, why data till 2009?, Giovanni has aerosol MODIS data till Dec 1, 2014.

When we started analysis, data were available until July 2012. As we are interested primarily
in describing seasonal trends typical of column aerosol above the major metropolitan areas of
South Africa (as opposed to the most recent data), we chose data from the last complete
decade available (2000-2010). Adding the additional 18 months is does not change any
results or conclusions, and our choice of exact start end end dates to the decade was a matter
of preference.

Page 24708 Line 10, what is the source of GOCART data?

• Data were obtained from Giovanni, and this has now been noted in the text.

Page 24708 Line 18, please provide the URL of FIRMS data that you used in the study.

The URL has been included in the methods section

Page 24710 Line 14, add "there" after "In every region"

Change has been made

In the Results and discussion section, I do not understand why long introduction about Aerosol Optical Depth and other parameters, it looks like text book. I think that it should be shorter and cited to references if anybody wants to get more details.

• The discussion of AOD has been shortened substantially, and only a brief description of the parameter is presented to give context for measurements.

Also I suggest just start discussing the results and in the interpretation part you can use literature for discussion.

• This is a nice suggestion, and we tried to integrate the interpretation part into the results. Doing so, however, distracted from the first main conclusions presented in the results. The results flow significantly better, and are presented with better context, with the physical basis of the measurement presented before the main results. That said, the discussion of the physical basis of the satellite measurements have been substantially shortened, and flows better now.

Page 24712 Line 25, here you are talking about correlation, Is not shown? Why there are no correlation plots.

• The paper is already heavy in figures, and the correlation between different satellite platforms is a minor point that does not deem representation in a figure. Correlations are only presented to demonstrate that satellite parameters from different platforms, which appear correlated in figures, are indeed statistically well-correlated. A detailed comparison of satellite platforms is beyond the scope of this work.

Page 24713 Line 25, If you want to discuss Figure 2 after Figure 3, why you do not switch them?

• They have been switched.

I noticed that sometimes you write Fig. and sometimes it is Figure, please unify

• "Figure" is now consistently used at the first mention of a particular figure, and "Fig." is used thereafter, in accordance with ACP standards.

Page 24715 Line 9, Are you here talking about Figure 4? If yes please refer to it.

• Yes, and we have made reference.

Page 24725 Line 17, what do you mean by Terra and MISR? Terra is the satellite and MISR is the instrument.

• We are referring to MODIS-Terra and MISR instruments. This has been clarified in the text.

Where are the correlation plots of water vapor with aerosol parameters?

 We feel that inclusion of correlation plots would be cumbersome and would add more figures to an already figure-heavy paper. The main point of the paper is not to explore correlations between satellite platforms or provide a detailed analysis of CWV correlations. We present CWV as one possible explanation for the trends observed, and state correlation coefficients as support. If the reviewer feels that it is absolutely necessary for publication to include correlation plots, we are happy to oblige. But if the correlation coefficients themselves are sufficient to establish statistical significance, then we would prefer - for readability sake - not to include the plots.

You mentioned that spatial resolution of satellite data is a factor that prohibits satellite data to capture trends in ground PM concentration, so why do you not try level 2?, level 2 data has much better resolution than level 3 that you used in this study.

- As noted above, the only conclusion we draw that might possibly be different with level 2 data is the poor agreement between satellite AOD and ground PM based on spatial resolution. Use of level 2 data would require a complete re-analysis of satellite data and ground-satellite correlations, which we have not done for the following reasons:
 - First, the discrepancy between satellite aerosol data and ground-based PM concentrations is primarily due to vertical inhomogeneity with stratified aerosol layers aloft (previous studies cited in the text). Level 2 data makes no difference here.
 - Second, the localized gradients between sites of different types occur on orders of 10 km or less. Level 2 data are still an order of magnitude coarser than these gradients.
 - Finally, we did perform an abbreviated analysis of AOD and Ängström exponent with level 2 data, and calculated correlation coefficients with ground-based PM concentrations. Level 2 data displayed no better agreement than level 3 data, and annual means were no different with level 2 data.