Thank you for your comments. Each of your comments are followed by my replies (shown in Italics).

-Brian Magi

Anonymous Referee 1

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Specific comments:

P13440, L15 Is the estimate that 26-27% of the aerosol absorption is due to OC really that precise?

This is the median value based on Figures 4b and 5b. The interquartile range (based on the regression) is 24-36%. I changed the text to simply read '~26%'.

P13441, L2 Perhaps being overly picky, but the word "we" is used, which implies a group decision even though this is a single author paper. It may be worth stating somewhere if Kirchstetter and Gao are in agreement with the correction that was derived.

I agree that 'we' reads strange. I wanted to avoid the passive voice, but looking at past publications, it seems to be the norm in single author studies. I changed the text to reflect this and minimize 'we'. Kirchstetter and Gao are mentioned in the acknowledgements as having read the paper and offered comments.

P13441, L9-10 The statement that an analysis of regional haze was precluded until now may need some qualifying. There was a "distributions" paper and a GEOS CHEM-measurement comparison paper that dealt with the regional haze by Sinha et al. Schmid et al discussed the regional haze as did Magi to some extent. So it may help to make this sentence a little more specific. Then at some point it may be worth noting if the conclusions of the Sinha et al papers and the original Gao et al paper are affected by the correction.

I meant that considering the carbon and ionic species from K2003 and G2003 together in a synthesized picture of the regional haze was not possible until we corrected the miscalibrated flow meters. I changed the statement to reflect this distinction. I agree that pointing out how my results affect past studies was an important oversight. For the most part, mass concentrations from G2003 and K2003 from regional haze have not been used since both studies offered more analysis of smoke plume samples and were cautious with the regional haze samples. I added a paragraph at the end of Section 2.3 to address the impact on past studies, but here is a (much) longer summary:

**K2003 results are not affected; they in fact list the specific values of OC and BC that I used in my study in their Table 1 with the exceptions being that I used corrected values from the simultaneous filter pairs (Flight 1833, 1229 UTC, and Flight 1835, 620 UTC in K2003 Table 1), and I do not use the filters listed for Flights 1837 and 1839 in K2003 Table 1 (they are over Namibia). My correction adds a second filter for flight 1833 and corrects the second value listed in K2003 Table 1 for flight 1835 from 20.1 to 9.1 (for TC) and similarly for OC and BC.*
**G2003 Figure 7 (described in Paragraph 19) suggest that the median contribution of inorganics was 30%, whereas our range in Figure 2 suggests 17-46%. These conclusions are consistent since G2003 was presenting results in the relative sense. By that I mean that G2003 relate values of SO4 and NO3 to PM2.5 that are all incorrect by the same fraction (my flow meter correction). G2003 are fairly cautious about their conclusions, and focus mainly on smoke plume samples and analysis. Values in G2003 Table 2 are very similar to those in our Table 2. I don't know the exact partitioning G2003 used, but like the samples in K2003, I used exactly the same data set to begin with. G2003 Table 2 'Background' values should be replaced by values I present in Table 4.*
**Sinha et al (Distributions of trace gases and aerosols..., 2003) Table 2 is affected. Namely, comparing my Table 2 and Table 4 gives a rough idea of how Sinha's Table 2 values are more like the pre-correction values in my Table 2 (Starting with Total particle mass and going down). The Zambia and Botswana sectors in S2003, for example, might be similar to what I called the tropics and the PM2.5 was about 21 ug/m3, while the S2003 South Africa and Mozambique sectors (~extratropics) had PM2.5 of 30 ug/m3. These are coarse comparisons, but consistent with my Table 2. My Table 4 provides important new information that updates the values. Some of the basic conclusions S2003 reach need to revised based on my study. For example, their conclusion that SO4 was higher in South Africa+Mozambique than in Zambia+Botswana is not what I have shown and discussed in my study. Overall, however, S2003 is a presentation of measurements that sets up the transport and chem studies by S2004.*
**The GEOS-CHEM study by Sinha et al (Transport of biomass burning emissions..., 2004) is not affected since they used BB emissions from Duncan et al. (2003) which have nothing to do with the aerosol mass concentrations from K2003 and G2003. I believe GEOS-CHEM in that era uses CO as a proxy for aerosol emissions, but S2004 is primarily a chemistry study.*

P13441 The aircraft sampled lots of fresh plumes, but those samples are evidently not included in this analysis. That should probably be clarified somewhere near the beginning. Also, the author largely ignores any SOA that may occur.

Plume samples are excluded, and we added this in Section 1 (namely, that K2003, G2003, and E2003 already presented these results). SOA is not explicitly characterized by the filter measurements.

P13441 It may be worth putting in the introduction that the tropical portion of the region studied is impacted mostly by biomass burning and the extratropical part has relatively more industry and desert-dust sources.

I added a statement specifically saying this (and referring to Piketh et al 1999 and Bond et al 2007) in Section 2.1 (Background). The sentence reads 'Since the tropics of southern Africa are more impacted by fires (Figure 1), and the extratropics are relatively more impacted by industrial and dust sources (Piketh et al., 1999; Bond et al., 2007), we would expect differences in aerosol properties (such as chemical composition, size, or number concentration) between samples characteristic of the tropics and the extratropics.'

P13443, L25-26 Its not immediately clear how an increase in particle number would increase the SSA. Further Table 6 lists just one value for MSC and MAC for BC and OC and the tropical aerosol has a higher OC/BC ratio (implication of Fig 2) so that seems to suggest a higher SSA for tropics. It doesn't seem consistent.

*I meant to say that the *higher AOD* is mostly due to change in N; the lower SSA is not related to N per se (except for the fact that most of that N increase in the tropics is related to an increase in carbonaceous aerosol mass. Table 6 contains evidence showing that the 'unifying' characteristic of the tropical and extratropical haze is the MSC and MAC. This makes sense because OC and BC are essentially from the same source. SO4 and NO3, on the other hand, have multiple sources and therefore are not as regionally uniform (hence the MLR results in Table 6 are much more uncertain than OC and BC). Referring to Table 6, I started Section 4.2 with 'The unifying characteristic of...'. Regarding the OC/BC ratio, Figure 2 is a % apportionment of total mass. A rough understanding of OC/BC ratio for extratropics vs tropics can be deduced from Table 5 (ie. 4.3 vs 5.0), but a more precise understanding of the ratio can only be gleaned from the data that together make up the median values in Table 5. I calculated this and the median OC/BC for the extratropics is 4.4 (3.7-6.8 IQR) and for the tropics is 4.2 (3.9-12.0 IQR). I also changed the last sentence of Section 2.1 to read 'The lower SSA in the tropics is due to a change in the overall chemical composition of the aerosol. We discuss both the evidence supporting this assertion and the chemical species that contributed to the increase in AOD in the tropics in Section 3.'*

L13443, L28 Can "shape of the size distribution" be defined in a more precise fashion?

The shape of the size distribution in terms of the angstrom exponent (alpha, Table 1) is not different. Alpha = 1.7-1.9 (extratropics) and 1.8-1.9 (tropics), implying a tendency towards a higher number of smaller particles in the tropics.

P13444, L17 I think that normally a flow meter reading should not depend on whether other flow meters are in use and perhaps that is why the word "defective" is used on line 28. Can the author give a simple reason why this problem could occur (e.g. broken meter, incorrect wiring?).

I changed the word 'defective' to 'miscalibrated' to be consistent with the previous paragraph and Section title. I was unable to determine the reason, but through simple tests, the calibration coefficients of the flow meters depended on the number of flow meters in use.

Section 2.3 General The correction is based on a limited amount of data, but the results of the correction seem logical at a first reading. I was on the same aircraft and saw higher CO and other pollutants expected to correlate with aerosol in the tropics. However, it might be worth clarifying (if possible) what evidence supports the conclusions of the paper in addition to the corrected data. In other words there is probably plenty of evidence that the conclusions of the paper are sound even without using the "corrected" Gao data. And is there any impact on Sinha et al distributions paper and the GEOS-CHEM modeling?

The conclusions I arrived at are based on the corrected data and are consistent with the various observations and measurements throughout the text. As is clear from Table 2 and Table 1, there is a major discrepancy between changes in aerosol optical properties and aerosol mass concentrations from filter measurements described in G2003. Perhaps I am misunderstanding your comment, but I don't see how the conclusions of the paper are sound without the Gao data. They are quite the opposite. I replied to your comment about the Sinha papers above (your comment about P13441, L9-10).

P13448, L19 Here and through out is assumed that NH_4^+ is the +1 counter ion. In fact couldn't K^+ be the counter ion as well? It doesn't matter for mass calculations since NH_4 and K are within one mass unit of each other. Is it also immaterial for the optical properties though? Or can K^+ be ruled out even though it is often used as an indicator of biomass burning (BB) and can be some 4% of fresh BB aerosol.

K^+ could indeed be the other part of the molecule. Gao et al. (2003) reported results of K but not NH_4. For regional haze, I did some analysis of the potential for K to be the counter ion to NO_3 and SO_4, but I decided not to include it in the text I initially submitted. Here is why: Median concentrations of K (after correction for the miscalibrated flow meters) were 0.15 and 0.64 ug/m3 for the extratropics and tropics, respectively. The IQRs were 0.12-0.20 and 0.51-0.88, if you're wondering. Converting the median mass concentrations to moles (using 39.0983 g/mol) results in median values are about 4 and 16 nmol/m3. There are about 41 and 78 nmol/m3 of SO_4 and NO_3 (based on Table 5 median). This implies that most (94% and 86% in the extratropics and tropics) of the SO_4 and NO_3 ions are not found in the form of K_2SO_4 and KNO_3 in the regional haze. From this, I inferred that NH_4 was the dominant counter ion to SO_4 and NO_3 and did not consider K in the mass calculations. In terms of the impact of this assumption, the mass of the molecular forms of SO_4 and NO_3 would be underestimated by 7 and 22% for the extratropics and tropics, respectively. This does not significantly affect the mass apportionment (ie. the values are still within the IQR in Table 5). The optics based conclusions should not be affected either. Tang 1997 (JGR, v102, no. D2, p. 1883-1893) shows in their Table 2 that the partial ionic refractions of K and NH_4 are 3.21 and 5.01, respectively. Considered alone, NH_4 is about 1.6 times as refractive per mol, but this does not account for density differences, among other things. The real refractive indices for K_2SO_4 and KNO_3 are less than those of NH_42SO_4 and NH_4NO_3, but a weighted average of the refractive indices (using 94% and 86% as above) would result in refractive indices that are only marginally less than what we assumed based on the simplifying assumption that NH_4 is the only counter ion. I added a sentence to the beginning of Section 3.2 stating this assumption.

P13448, L24-25 Not critical but Aiken et al also have a paper supporting the higher OM/OC ratios for BB aerosol.

I am not sure about which study you are referring to, but Aiken et al, Env Sci Tech, 2008 states (p 4483) that OM/OC for BB aerosol is 1.56-1.70 for lodgepole pine and sagebrush, pointing out that these are lower than similar values reported in Turpin and Lim.

P13449, L11-13 The data presented seem to indicate an important role for SVOC, but that is not clarified or developed.

I did not intend to analyze this beyond what E2003 already did. Namely, they pointed out that SVOC 'which could not be measured by conventional filter samplers, averaged 38% of the total fine particulate mass in aged aerosols from savanna fires'. Values in our Table 5 suggest median contributions of 11-33%, which are different than E2003 because we use a larger sample size of PM2.5 mass concentrations (ie. those from G2003 and corrected by the method I described). The SVOC analysis and conclusions did not change since E2003 - I added a sentence saying this at the end of Section 3.1.

P13449, L20 Standard temperature should be specified (273 or 298?).

I added that STP = 273K, 1013hPa in Section 3.3. Thanks.

P13450 Somewhere in this section the author may want to specify if there is an assumption of an internally or externally mixed aerosol and if it makes any difference.
Also, I can only find optical properties listed for nitrate and sulfate so the properties are evidently independent of the counterion. If that is the case it may be worth stating somewhere at this point in the text.

I assumed an external mixture and added this assumption in the beginning of Section 3.3. The MLR technique would not work for an internal mixture. I could find no evidence suggesting that the molecular forms of NO3 and SO4 absorb significant amounts of visible radiation, whether the counter ion is K or NH4. Also, see my response to your comment for P13448, L19 above.

P13452, L 3 and 12 and P13453, L6 The word "concentrated" or something like it is more appropriate than the word "intense" which could be interpreted as a description of fire behavior.

Agreed. I changed the wording. Thanks.

General comment on this section: The southern Africa gyre and the "river of smoke" are responsible for at least some mixing between the tropical and extratropical regions investigated here. This would cause a little blurring of the distinction between the regions.

I assume you meant Section 4.1. I offered some justification for the distinction in the case of the filter data based on the back-trajectory analysis and described in Section 2.1. The distinction is not perfect, but it helps to explain the significant differences in Table 5 PM2.5, OC, and BC. Given the trajectories, I asserted that the dominant source of the samples could be characterized as 'tropical' or 'extratropical' in origin. It is a simplification, but one that is supported by ample evidence (filters, back traj, AOD, SSA, number concentrations, fire occurrence).

P13455, L1 Was the value of MAC(BC) 7.5 +-1.2 m2/g from Bond et al restricted to a fuel type or region? I ask because a detailed study by Martins et al of smoke in the Amazon suggested a value of 12+-4 m2/g.

I am assuming Martins et al, JGR, 1998 (SCAR-B). 12+/-4 does not tell the whole story. Martins et al 1998 state that the full range of MAC_BC is 5-58 m2/g (5-21 m2/g if high K/BC were excluded) . Bond and Bergstrom 2006 included Martins et al 1998 in their review and synthesis, and their work is an attempt to arrive at a single representative value (and range) for MACBC. From Bond and Bergstrom 2006 Section 9.1: 'we suggest a value of 7.5+/-1.2 m2/g for the MAC of fresh light-absorbing carbon. During a particle's lifetime, this value may increase due to coating and decrease due to particle coagulation and aggregate collapse'. The main problem with this value is discussed in their Figure 9.

P13456, L1-2 Not a critical comment, but this could be more precise, since "OC" is not an actual molecule that absorbs light, but a measure of the mass of C found in organic molecules. The chromophores that absorb light are quite often functional groups that contain O, H, or N.

Good point. I changed this to say 'there is no evidence suggesting that OM is more absorbing than OC '.

P13456, L15-17 You could leave out "the plume" since smoke can likely age even after it is no longer in a coherent plume. This is sort of why chosing a smoke age appropriate for models is tough. What age and how reproducible are the aging effects no-one knows.

Changed. Thanks.

Table 1: I expected N2/N1 to crudely correlate with C/F, but it doesn't. Not sure if that matters or not.

C/F is calculated as the ratio of coarse mode (1000-3000 nm diameter) volume to fine mode (100 to 1000 nm) volume. N1 and N2 are number concentrations and also have much different lower limits on particle size (3 nm vs 100 nm).

Table 2 header should perhaps signal that these values are "uncorrected" or "precorrected"

Changed. Thanks.