

Interactive comment on “The composition and variability of atmospheric aerosol over Southeast Asia during 2008” by W. Trivitayanurak et al.

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Response to reviewer comments of “The composition and variability of atmospheric aerosol over Southeast Asia during 2008,” by W. Trivitayanurak et al

We thank one anonymous reviewer and A. R. Mackenzie for providing useful comments that have ultimately improved the manuscript. Below we address all comments (denoted in italics) individually.

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1 Anonymous review

The abstract could use at least 1 or 2 sentences of general motivation/background information for those readers lacking a predisposed interest in aerosol in Borneo.

Point well taken. We have added:

Our focus on Southeast Asia reflects the importance of this region as a source of reactive organic gases and aerosols from natural forests, biomass burning, and food and fuel crops.

..the abstract contains only one sentence regarding model skill. Focusing on unexplained biases could actually be of greater interest to the broader aerosol community than details of the model budget. . .

We have revised the manuscript to address the reviewer comments:

We use MODIS aerosol optical depths (AOD) data and the model to put the July campaign into a longer temporal perspective. We find that Borneo is where the model has the least skill at reproducing the data, where the model has a negative bias of 76% and only captures 14% of the observed variability. This model performance reflects the small-scale island-marine environment and the mix of aerosol species, with the model showing more skill at reproducing observed AOD over larger continental regions such as China where AOD is dominated by one aerosol type. The model shows that AOD over Borneo is approximately even split between organic and sulphate aerosol with sea salt representing 10-20% during May-September; we find a similar breakdown over continental Southeast Asia but with less sea salt aerosol and more dust aerosol.

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In contrast, East China AOD is determined mainly by sulphate aerosol and a seasonal source of dust aerosol, as expected. Realistic sensitivity runs, designed to test our underlying assumptions about emissions and chemistry over Borneo, show that model AOD is most sensitive to isoprene emissions and organic gas-phase partitioning but all fail to improve significantly upon the control model calculation. This emphasises the multi-faceted dimension of the problem and the need for concurrent and coordinated development of BVOC emissions, and BVOC chemistry and organic aerosol formation mechanisms

Last line: "that the model". . . what aspect of the model?

We refer to the model calculation of AOD. See revised abstract above.

Re partitioning between SO₂ and SO₄: the authors are shying away from the more challenging, and interesting, aspects of the study.

The focus of the paper is about organic aerosol. We used the EANET measurements, in the absence of other sulphate data over Southeast Asia and in particular Borneo, to provide constraints on the model sulphur budget over our broader study region. We agree that the partitioning between SO₂ and SO₄ is interesting but we feel it is better addressed in a separate study where appropriate space can be given to that subject.

A table of the statistical analysis (bias, error) for the model/aircraft data would be informative

We have included a new table in the manuscript that summarises the statistical analysis.

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Would it make a difference if comparisons were made to other MODIS product, such as the Deep Blue AOD retrievals, or those wherein the GEOS-Chem model is used for the retrieval? Or are the authors otherwise accounting for assumptions made regarding aerosol properties and distributions in the MODIS retrieval vs their model?

We suspect it would make a difference to our analysis if we used another MODIS AOD product but without such a major undertaking we are cautious about whether it would improve or degrade our model bias. Certainly based on previous studies of satellite observations of AOD, where retrievals are informed by model aerosol speciation, particularly over regions where we know the most about aerosol speciation (e.g., North America), the model-data comparison is improved. But this assumes that the model is unbiased. For Borneo, where we have aircraft measurements during OP3/ACES, the model has a bias that could degrade the measurement comparison. Again, this looks like analysis that could form the basis of another substantial paper.

Are the OC and SO₄ extinctions anti-correlated in the FT? Also if one were to normalize these plots by the value of the aerosol concentrations, which species would make the largest contribution to extinction relative to its mass concentration? Would that help guide investigation of which sources of error could likely be leading to the model bias?

OC and SO₄ extinctions appear to be anti-correlated in the FT but depends on the upwind sources. Robinson et al (2011) explained that the air en-route to Borneo in the FT showing an anti-correlation of organics and SO₄ because as the organics get replenished from the underlying forest, there is a net depletion of SO₄ (marine origin) via deposition.

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Aerosol mass extinction (eq 2 in the manuscript) is a function of relative humidity (RH) and aerosol optical properties (see Figure below). At low RH, black carbon makes the largest contribution to extinction relative to its mass concentration. At high RH, hygroscopicity and accumulation-mode sizes play a larger role in the aerosol mass extinction, making accumulation-mode sea salt, sulphate, and organics the species with highest aerosol mass extinction.

Insert Figure. Caption: Aerosol mass extinction (m²/g) as a function relative humidity (%) for sea salt in accumulation (SSa) and coarse (SSc) modes, sulphate (SO₄), black carbon (BC), organic carbon (OC), and dust at 0.7, 1.5, 2.5, and 4 microns (Dust1, Dust2, Dust3, and Dust4, respectively).

In comparing Figs 5 and 6, why does SO₄ seem to play a greater role in AOD than in extinction?

There are details missing in the description, which we have now addressed. While the model extinction calculation is for Danum Valley region, or more accurately the local area defined by the nested model resolution of 0.5x0.67 degrees that includes the flight tracks, the AOD calculation is for the larger Borneo Island which reflects a spatial scale compatible with MODIS data availability. The other difference is that the Danum Valley extinction calculation is at the nested grid resolution while the AOD calculation is at the 2x2.5 degree resolution to allow us to compare the control run with sensitivity runs. The difference between the AOD and extinction is largely due to the regional definition rather than the resolution.

“only days when AOD is relatively low” Does this description hold true for the SO₄

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extinction peaking near day 50, where it seems like the total extinction is also high?

This is generally true except for day 50, it seems. We have toned down this offending statement.

“that chamber yields do not represent atmosphere formation.” This statement seems overly broad and I urge the authors to consider a more carefully worded conclusion. There are many studies of chamber yields under varying NO_x levels, particle acidity, aqueous content, organic ageing, photochemical state, etc. The present modelling study has ruled which ones, precisely? In general, the conclusions here regarding SOA could better placed in context with recent advances in this field regarding the importance of these processes, most of which are not included in the present modelling study (and thus the persistent bias in estimated SOA concentrations is perhaps anticipated).

We have included the following sentence in section 4.3.3:

Significant isoprene SOA concentrations were detected over the Danum Valley ground measurement site during OP3/ACES (Robinson et al, 2011b) so low sensitivity of organic aerosol to changes in isoprene emissions suggest that SOA yields used in our current model are not indicative to atmospheric formation.

I found it odd that none of the results of the sensitivity runs were compared to the in situ observations, only to the MODIS AOD, and encourage the authors to revisit comparisons to the former during during the sensitivity calculations. Is there even a mechanism by which the model could match both the AOD and still retain reasonably agreement with the profiles measured from the BAe-146 flights, or are the implications

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of these two data sets in opposition?

We have addressed this oversight in the revised paper. The improvement we see in AOD in the PAR simulation is achieved at the cost of a positive model bias in organic aerosol observed along the OP3 aircraft flight tracks. Doubling biomass burning emissions significantly reduces the model bias against the aircraft data but results in only a modest improvement in AOD. Increasing anthropogenic and natural source of sulphur decreases the model AOD error over Borneo but results in an increased positive model bias along the aircraft flight tracks. In the SEA simulation, for which we increase isoprene emissions, increases the model bias of isoprene and isoprene oxidation products and modestly decreases the model bias in organic aerosol relative to the aircraft data. This last result confirms the shortcomings of the SOA yields in the model.

1.1 Technical Corrections

p22046: Does this, and all subsequent analysis, refer to only the MODIS finemode fraction?

No, we don't use the fine mode aerosol product in the comparison. The only use of fine-mode AOD to support the calculation in the DMS sensitivity run is clearly stated as "fine-mode AOD".

All other comments have been addressed.

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2 Review from A. R. MacKenzie

The discussion of the MEGAN implementation here does not describe how variation in land use – particularly oil palm plantation vs rainforest – was taken into account. Because oil palm has extremely large isoprene emissions and a different terpene emission speciation, the details of the land-use map may be important for interpreting model-measurement difference in section 4.2.1.. See MacKenzie et al.

This reviewer is correct that incorrectly describing the oil-palm/natural forest boundaries over Borneo may contribute to the model bias. We use a MEGAN implementation that includes elevated isoprene emissions over Danum Valley, however our nested grid resolution (0.5x0.67o) is not sufficient to describe the sharp spatial variations in land-use observed during OP3/ACES.

If a similar regional comparison of the GEOS-Chem model against MODIS AOD has been carried out for Amazonia or central Africa, it would be useful to report how the current comparison relates.

Ridley et al (2011) compares GEOS-Chem model AODs against MODIS observations over Africa, with a focus on North African dust. In comparison to our study, Ridley et al (2011) show that the central Africa forested region is overlaid with dust and biomass burning aerosol, with only a small contribution from biogenic organic aerosol. The study also shows that GEOS-Chem has a negative AOD bias over central Africa during SON when dust aerosol sources are low. Although only qualitative, this result suggests that low organic aerosol concentrations are also found over central Africa, another major forest region.

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2.1 Minor Comments

Text, references have been changed accordingly to reviewer comments.

3 References

Ridley, D. A., C. L. Heald, and B. J. Ford (2011), North African dust export and deposition: A satellite and model perspective, *J. Geophys. Res.*, doi:10.1029/2011JD016794, in press.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 11, 22033, 2011.

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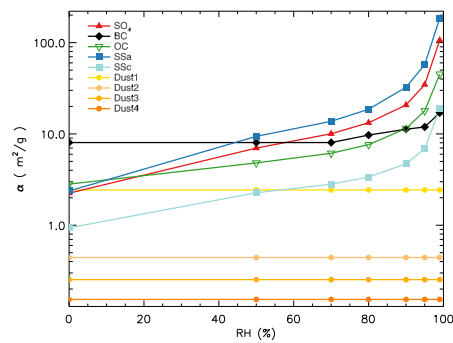


Fig. 1.

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