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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterization of a large biogenic secondary organic aerosol event from eastern Canadian forests" *by* J. G. Slowik et al.

J. G. Slowik et al.

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Comment 1:

The paper points out that the agreement between the elevated measured OA and the AURAM model results contrasts with the reported gross under-prediction of measurements by models in polluted regions. However, what the paper doesn't discuss is that recent studies in tropical biogenic environments do show reasonable agreement between measurements and models (e.g. Chen et al., GRL, 2009), though not at such elevated concentrations as are reported here. It is also not discussed whether the model performs better in these environments or whether there are multiple compound-

ing errors. Capes et al. (2009) show that whilst there is good agreement between measurements and a global model over the sub Saharan African region during the wet season, steady state calculations show that the measurements are considerably higher than would be expected from the precursors. They use different yields and atmospherically relevant precursor concentrations and show that there is a great variability in these parameters and may give a false impression of the goodness of the agreement between measurements and global models. The authors need to tackle this question in the paper for their environment I think.

Response

We now note the agreement between measured and modelled biogenic SOA observed in pristine environments dominated by isoprene emissions in the AURAMS/measurement comparison, citing the Capes et al. and Chen et al. studies. The present study provides a consistent result in a region with a stronger monoterpene influence.

We cannot replicate the steady-state analysis of Capes et al. in this study, as our measurement site is significantly removed from the region of high monoterpene emissions driving the biogenic event. We do estimate an SOA yield from monoterpenes based on MCM modelling of the photochemical production of CO from monoterpenes (using α-pinene as a representative compound for monoterpenes). However, this analysis is complicated by potential CO (and SOA) production from isoprene. The AURAMS predictions incorporate estimates of regional monoterpene emissions and are in agreement with SOA concentrations during the biogenic period.

Comment 2:

There is new evidence (Kiendler-Scharr et al., 2009) that isoprene may inhibit SOA formation via terpenes by providing an OH scavenger. If this is an important mechanism then it will greatly reduce the SOA formed over tropical regions where isoprene emissions dominate those of terpenes, whereas in boreal regions, where terpene emis-

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sions dominate, one might expect a greater SOA formation. The results in this paper are consistent with this though the results from Hytialla are not. I am aware that the authors may not have been aware of the KS et al. results when preparing the ACPD paper but I would encourage them to include such a discussion and identify ways of testing this in future studies. I would also encourage a summary of current findings on field measurements in isoprene and monoterpene dominated environments and a discussion of the consistency and differences in the results.

Response

We now discuss the Kiendler-Scharr results in terms of the driving forces of the biogenic event. Specifically, the lack of large biogenic events corresponding to southerly winds and high temperature are likely the result of isoprene-dominated biogenic VOC emissions, leading to both lower SOA yields and possible OH scavenging. This contrasts with the higher biogenic SOA concentrations expected during periods of high temperature and northerly winds, bringing monoterpene emissions from the boreal forest.

Comment 3:

The authors stress the finding that AURAMS agrees with the measurements during the biogenic event is a novel result. As noted above, this has been observed a few times previously. However it remains an important finding. However the model description shows that a considerable amount of new yield data has been used. The changes that these yield data make to AURAMS output in a range of situations from those previously used by AURAMS and other models are not commented on or referenced, they need to be, this is important. Capes et al., (2009) show that such changes can be very large. Are the authors sure that the reason for the good agreement is because of the biogenic situation is different to the anthropogenic one or due to changes in yield data? Following up on this, I agree with an earlier reviewer that it would be very good to see how the simulation behaves compared to data in the polluted period.

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Response

We did not discuss the anthropogenic SOA in this paper because we wanted to keep the focus of the paper on describing the surprisingly large biogenic event using surface measurements, satellite data and model. In a subsequent paper, we will focus on model performance for anthropogenic events at sites in southern Ontario closer to anthropogenic emissions (BAQS-Met study in the Detroit-Windsor area, mid-June to mid-July 2007).

For the present study, Fig. 9 has been expanded to include all model and measurement data from the end of model spin-up (end of 3 June) to the end of the measurement campaign (15 June). Unfortunately, the end of the campaign prevents us from accomodating the reviewer's request to extend the figure past this date, though we agree this would be interesting. The expanded figure shows that AURAMS underpredicts the peak SOA in Toronto outflow during the June 8 event by a factor of 2 to 3; however, the relative contributions of fine-scale meteorology and SOA production to this discrepancy are uncertain.

We have finished an evaluation of AURAMS organic aerosol predictions against the STN measurement data in the eastern U.S. (urban sites, 24-hr samples, June-July 2007) (see attached Fig. 1).

We do see a negative bias but the model and measurement averages are within the standard deviation. This is a significantly improved bias over a prior comparison of the model with the IMPROVE data for the summer 2005 (northeastern U.S., rural sites, 24-hr samples, June-August) (see attached Fig. 2).

For this IMPROVE comparison, we used SOA yield data from Odum et al (1997) and Griffin et al. (1999) studies. Another key difference in the prior model was related to model chemical species lumping. Previously, monoterpenes were lumped with anthropogenic alkenes so the monoterpenes did not have ïĄąi and Ki values specific to monoterpenes. Numerous other studies in the literature have noted order of mag-

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nitude model-measurement discrepancies from prior SOA yield data (Hallquist et al., 2009 and Kanakidou et al. 2004 review papers)

For the BAQS-Met SOA time series, there are periods where AURAMS agrees quite well and periods where the model completely misses the urban plumes due to the complexity of the meteorology (e.g. lake breeze fronts).

Comment 4:

The AURAMS modelling shows the agreement for the biogenic period, and then the agreement is not good for the last part of that period. What it does not show is whether AURAMS does or does not do a good job during the polluted period. The data appear to show that biogenics are also playing a significant role during this part of the experiment, do the model simulations bear this out? What can be learnt by showing where the measurements and model disagree as well as showing that they agree well?

Response

This issue was discussed above in response to Comment 3. To summarize, Fig. 9 has been expanded to include all model and measurement data from the end of model spin-up (end of June 3) to the end of the measurement campaign (June 15). The expanded figure shows that AURAMS underpredicts the peak SOA in Toronto outflow during the June 8 event by a factor of 2 to 3; however, the relative contributions of fine-scale meteorology and SOA production to this discrepancy are uncertain. A detailed model/measurement comparison for anthropogenic events at sites in southern Ontario closer to anthropogenic emissions is underway (BAQS-Met study in the Detroit-Windsor area, mid-June to mid-July 2007) and will be the focus of a subsequent paper.

Comment 5:

The authors compare a 4 and 5 factor solution for the PMF analysis and conclude that as the third OOA factor retrieved from the 5 factor solution alters the OOA1 and OOA2 then it must be due to splitting and so reject the 5 factor solution. I am not sure that such

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a conclusion can be drawn at all. The mass spectrum of OOA3 in the supplementary material is indicative of a younger secondary organic aerosol – similar to those seen in laboratory chamber studies, whereas the OOA 2 in the 5 factor solution represents a higher 44/43 ratio and is indicative of a more processed OA. The time series shows that OOA3 is a relatively much larger contribution during the identified biogenic period than at other times – this at first sight is consistent with a closer source. PMF picks out factors which explain the variance, where one has multiple air masses demonstrating different ageing states of an organic aerosol during processing then different OOA can easily result. These are not splitting but may reflect different stages in the continuum of the ageing process. A boot strapping analysis should be performed on both the 4 and 5 factor solutions to see if the 5 factor solution is robust and a more rigorous discussion of why a four factor solution is used should be provided.

Response

The 5-factor solution is rejected primarily for two reasons. (1) The 5-factor solution does not significantly decrease the scaled residuals relative to the 4-factor solution (see Fig. 1). This indicates that the 5-factor and 4-factor solutions are similar in their ability to describe the dataset. (2) The 5-factor solution represents a subset of the data ("OOA") that can be described as a linear combination of two factors (OOA-1 and OOA-2) as a linear combination of three factors, in the absence of correlations with external tracer data that would suggest a physical meaning for this more complicated description. Note that the presence of a reasonable-looking OOA-3 spectrum does not indicate that the factor is "real;" similar behavior has been observed as a result of factor mixing/splitting in synthetic datasets [Ulbrich et al., 2009]. We cannot altogether rule out the possibility of meaningful factors in higher-order solutions, but given the lack of improvement in the solution and absence of tracer correlations, such solutions cannot be supported. This line of reasoning is now explictly stated in the manuscript.

Bootstrapping analysis has been performed on both the 4 and 5-factor solutions, and the results are summarized in the new Figs. S9 and S10. This analysis does not show

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large variation in either solution, and the 4-factor solution is selected according to the criteria described above.

Comment 6:

Specific points Abstract P 18114 line 1-2: Biogenic SOA was not measured in this study, OA was measured in this study and analysis is presented to show that this arises from biogenic sources.

Response

The revised manuscript reads: “Measurements of aerosol composition, volatile organic compounds, and CO are used to determine biogenic secondary organic aerosol (SOA) concentrations at a rural site 70 km north of Toronto. These biogenic SOA levels are many times higher than past observations and occur during a period of increasing temperatures and outflow from Northern Ontario and Quebec forests in early summer.”

Comment 7:

Page 18114 line17: Radiative forcing as used by the IPCC refers to the radiative flux changes induced by man-made perturbations and not to natural systems. In this sense the SOA presented here is not a radiative forcing but represents a cooling on the Earth System via increased scattering of short wave radiation.

Response

We now refer to “enhanced direct optical scattering” rather than radiative forcing.

Comment 8:

Page 18115 line 7-9: I do not understand this sentence – the authors seem to imply that not much work has been done studying SOA from biogenic environments because everyone was too busy studying polluted environments. This isn’t really the case: there have been many studies in Hyttialla for a number of years in northern Boreal forests and studies in the Amazon Basin from several years ago. Furthermore, there has been

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considerable interest in studying biogenic OA systems in chambers for a number of years, more so perhaps than polluted environments.

Response

The sentence has been deleted.

Comment 9:

Page 18115 Line 9: “significantly lower levels”: : :than those observed over polluted regions.

Response

We appreciate the suggestion and have modified the manuscript.

Comment 10:

Page 18115 line 10: It would be useful to separate these studies into tropical, subtropical and northern boreal for the benefit of the reader and possibly give some insight into whether these are isoprene or terpene dominated locations.

Response

The studies are now classified as suggested, and we have added the Chen et al. (GRL, 2009) study in the Amazon as an additional tropical study.

Comment 11:

Page 18117: Though the effect is only slight it would be useful to know whether the mass loadings presented are standardised to STP or whether they are based on the standard pressure at the time the AMS was calibrated?

Response

Mass loadings are based on the calibration pressure. This is now stated in the manuscript.

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Comment 12:

Page 18117 lines 26-28: There is no need to define the collection efficiency twice.

Response

The revised explanation of the collection efficiency due to bounce reads as follows: "An important consideration in the quantitative analysis of AMS data is the collection efficiency due to particle bounce. On impacting the vaporizer, some fraction of non-refractory particles bounces off the vaporizer surface instead of vaporizing. The detected number fraction is defined as the bounce collection efficiency (Eb), which depends primarily on particle phase [Matthew et al., 2008]."

Comment 13:

Page 18119 lines 7: why isn't the period on the 24th and 25th when C3H3+ dominant and the K is low also classified as a biogenic period?

Response

Criteria for determining periods of biogenic influence are discussed in detail in section 3.2 in terms of the data presented in Fig. 4. For the period of 24-25 May, Fig. 4 shows high sulfate loadings (indicative of long-range transport) and high acetylene concentrations (a long-lived anthropogenic tracer). As shown in Fig. 3, benzene and NOx are also somewhat elevated. These characteristics are consistent with anthropogenic influences. This interpretation is confirmed by back trajectories showing the airmass arriving from the south after passing over Toronto. We now note this period as an additional example of anthropogenic influence.

Comment 14:

Page 18123 lines It isn't really necessary for this paper but the authors should comment on the thermodynamics scheme used.

Response

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SOA yield equations are based on Raoult's law equilibrium for an ideal solution composed of both POA and SOA [Pankow 1994].

Reference: Pankow, J.F.: An absorption model of the gas/aerosol partitioning involved in the formation of secondary organic aerosol, Atmos. Environ., 28, 189-193, 1994.

Comment 15:

Page 18124 line 15: channel should be plural

Response

The typo has been corrected.

Comment 16:

Page 18126 lines 14-17: Though the correlation between HOA and NOx is good, there are a couple of periods when this is not the case. Are the authors able to determine what the cause of these differences might be? It appears that benzene is better correlated with HOA, yet the authors do not comment on this. Why might this be so?

Response

The temporal correlations of HOA with NOx and benzene are of similar quality (R2 = 0.41 and 0.43, respectively). Most of the variability between these three species is likely due to changes in emissions source profiles (e.g. the relative contributions of the Toronto urban outflow vs. highway emissions from the east/southeast). This is now noted in the manuscript.

Comment 17:

Page 18126 line 20: The correlation with levoglucosan refers to previous studies, this should be made clear.

Response

In the original manuscript, we provided a reference for the levoglucosan reference

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spectra (Schneider et al 2006, page 18126 line 21) and noted that all MS comparisons were performed with spectra from the AMS Spectral Database (page 18126 line 8).

Comment 18:

Page 18126 lines 20-24: I agree with previous reviewers, the correlations here are low and yet much is made of these correlations for determining the air masses. Some comment needs to be made and the justification be tightened up here.

Response

The comment refers to correlations between AMS BBOA and the biomass burning tracers potassium and acetonitrile. We agree that the correlations are low. In the present study, both tracers are known or suspected to be problematic. The issues with the AMS potassium measurement (competing electron impact/surface ionization pathways, interference from C3H3+) are discussed in detail in the manuscript. For acetonitrile, recent high resolution PTR-MS measurements in downtown Toronto have indicated an interference at m/z 42 from the C2H2O+ ion [A. Vlasenko, personal communication, 2009]. This likely influences the acetonitrile time series during periods of Toronto outflow, e.g. the elevated acetonitrile periods between 25 May and 6 June. As a result, we do not expect more than a qualitative correlation between BBOA and these tracers. The acetonitrile interference and expectation of qualitative correlations are now noted in the paper.

Comment 19:

Page 18129: It would be good to provide dates on the identified periods.

Response

We now provide dates for each case study period at the beginning of section 3.2, where they are first discussed. We also note that the periods are designated in Fig. 4.

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Comment 20:

Page 18129: The names given to the different periods in figure 5 are not mentioned earlier. They should be so that the link can be drectly established.

Response

As discussed above in response to Comment 19, the periods are now explicitly identified at the beginning of section 3.2.

Comment 21:

Page 18130 lines 7-9: Neither Yokelson et al or Reid et al measured the organic mass, rather they measured OC and converted to OM a neither used an AMS. Capes et al (2008) can be used also as they provide a direct measure of OM. However, both Capes et al and Yokelson et al are not from boreal forest fires and the OM/CO could be different, this needs to be commented on.

Response

We have added a new Table (Table 1) containing a more detailed comparison of delta_OM/delta_CO with other studies. We agree with the reviewer's comment regarding the Yokelson et al. and Reid et al. measurements; this was noted in the initial manuscript and is now contained in a footnote to Table 1 as follows: "Estimated from non-speciated PM measurements, assuming that OM accounts for 55% of PM [Reid et al., 2005]." The Capes et al. study is included in Table 1. As suggested, we note that none of the above studies measured boreal forest fire emissions. However, Table 1 also contains a study by Grieshop et al. (2009) including studies of fresh and processed emissions from yellow pine combustion, which may be a reasonable surrogate for boreal fires.

Reference: Grieshop, A.P., Logue, J.M., Donahue, N.M., and Robinson, A.L.: Laboratory investigation of photochemical oxidation of organic aerosol from wood fires 1: measurement and simulation of organic aerosol evolution, Atmos. Chem. Phys., 9,

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1263-1277, 2009.

Comment 22:

Page 18130 lines 22-24: The author claim that a tight correlation between OOA2 and CO means that the photochemical age of the air remains constant. I think what is meant is that the photochemical processing remains constant. I cannot see how a single point receptor receiving air travelling over a large areal extent of the boreal forest to the north can measure air parcels with a single photochemical age. Rather it is the footprint and the photochemical environment across the region that is likely to remain constant.

Response

We agree with the reviewer's suggested clarification, and now refer to the absence of changes to the emissions profile and photochemical environment.

Comment 23:

Page 18130 lines 20-28: The authors discuss that no other sources can produce such high OOA2/CO ratios and therefore the source must be biogenic. I think a more detailed discussion is needed here to rule out other sources as the NOx is non-negligible and the CO is high (whilst the latter is consistent with a biogenic source it also means other sources cannot be rule doubt). How well is the CO explained by the available biogenic that has been oxidised? This section implies that the reason for the high OOA2/CO is because the SOA is biogenic in origin. A likely explanation for the high OOA2 is that close to source the available condensable material has a high concentration and even if it is reasonably volatile a significant amount will be present in the particle phase. This will evaporate on dilution further from the source field. The high OOA/CO are a feature of the proximity and strength of the source not that the source is biogenic per se. This needs to be discussed.

Response

We have added a more detailed discussion of the potential influences of fresh or aged

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biomass burning sources in connection with the new Table 1.

As discussed in section 3.3.1, the observed relationship of photochemical CO to SOA suggests an SOA mass yield of 7.2 to 9.3%, assuming α-pinene-like characteristics for the SOA and CO precursors. This range is consistent with the literature, where reported SOA mass yields from α-pinene range from approximately 3 to 45%, although a direct comparison is complicated by the dependence of the yield on SOA mass loading (causing an increased yield), temperature, and high/low NOx regimes. The qualitative agreement between calculated and literature SOA mass yields indicates that the observed CO levels are reasonable for a photochemical source; this is now explicitly stated in the manuscript.

We agree that the partitioning and source proximity issues raised by the reviewer are important considerations. However, it is likely that the principal cause of the high biogenic OOA-2/CO is the lack of a significant source of primary CO. For the biogenic aerosol, the effects of partitioning/source proximity may be somewhat diminished by the fact that the biogenic aerosol originates from a relatively diffuse source compared to, for example, Toronto outflow. This is suggested by the additional scatter evident in the urban outflow relative the biogenic period in Fig. 5d. These issues are now discussed in the manuscript.

Comment 24:

Page 18131 lines 11-16: It isn't clear to me how the 600 ppb increase is obtained from figure 5. The intercept during the biogenic period looks like it intercepts the x axis at around 120 ppb. How is the background derived to obtain the secondary contribution to this number?

Response

First, we note for clarity that the CO increase is stated to be 60 ppb, rather than 600. The biogenic CO increase is estimated as the difference of the maximum value (180

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ppb) and the x-intercept (120 ppb). This is now explicitly stated in the manuscript.

Comment 25:

Page 18133 It appears that the BVOC are dominated by monoterpenes. Is this correct? What is the MT/ISOP emission ratio? Does this demonstrate that the precursors are dominated by MTs?

Response

A paper was published last year with isoprene (ISOP) and monoterpenes (ALKE) mass emissions averaged over a summertime 43-day period for eastern North America (Stroud et al., Atmos Environ, 42, 2008, Figure 2, see below as attached Fig. 3). For the Canadian boreal forest in Northern Ontario and Quebec, the isoprene and monoterpene mass emissions are quite comparable (within a factor of 2) while the yield data for monoterpenes is an order of magnitude larger than isoprene. The situation is quite different in the southeastern US where isoprene emissions are much larger than monoterpenes. The relative strength of the isoprene and monoterpene emissions are now mentioned in the manuscript and the Stroud et al. study is cited.

Comment 26:

Page 18134 point 2: I am unsure what is meant here. Do you mean that the OM/CO is significantly larger for the biogenic period compared with other periods in this study or than is reported in the literature?

Response

This discussion has been expanded in the manuscript. delta_OM/delta_CO for the biogenic period is significantly larger than that of the biomass burning and urban outflow case studies, as well as literature values for urban outflow and fresh biomass burning emissions. The biogenic delta_OM/delta_CO is also larger than literature values for aged biomass burning emissions, though this comparison is more complicated and is discussed in connection with the new Table 1.

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Comment 27:

Page 18135 line 6: Include the lifetime and references for acetonitrile and potassium.

Response

This information has been added to the manuscript. The atmospheric lifetime of acetonitrile is on the order of several months or longer (see for example Hamm and Warneck 1990; Singh et al. 2003). The lifetime of potassium is the same as that of the particle, which may vary considerably depending on transport and atmospheric processing, but is expected to be on the order of days to weeks.

References: Hamm, S., and Warneck, P.: The interhemispheric distribution and the budget of acetonitrile in the troposphere, j. Geophys. Res.-Atmos., 95, 20593-20606, 1990. Singh., H., Salas, L., Herlth, D., Kolyer, R., Czech, E., Vlezee, W., Li, Q., Jacob, D.J., Blake, D., Sachse, G., Harward, C.N., Fuelberg, H., Kiley, C.M., Zhao, Y.J., and Kondo, Y.: In situ measurements of HCN and CH3CN over the Pacific Ocean: Sources, sinks, and budgets, J. Geophys. Res., 108, 8795, doi:10.1029/2002JD003006, 2003.

Comment 28:

Page 18135 line 7: See Capes et al. (2008) for BBOA to OOA1 conversion time.

Response

Following Capes et al. (2008), we now note that while the lifetime of BBOA is less certain, the signal at the characteristic m/z 60 and 73 fragments has been observed to disappear as a biomass burning plume evolves over thousands of kilometers.

Comment 29:

Page 18135 point 2: This argument is circular in nature. The data are being used to test the AURAMS prediction, yet AURAMS is being used as evidence to interpret the data. The argument needs to be re-developed here.

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Response

The data and AURAMS represent two independent methods of estimating the biogenic SOA concentration. Comparison between the two methods provides a consistency check for both. This has been clarified in the manuscript.

Comment 30:

Page 18135 point 4: The data shown in figure 10c may not be as definitive as the authors state in the point. The back trajectories arriving at Egbert at 1800 on the 12th and 13th have come from a NE direction - close to the fire sources shown in figure 10a. However, figure 10a is for the period between 12th and 14th. What happens to the fires in the preceeding days? If these fires were present on the 10th then it is possible that long range transport of biomass burning air may influence Egbert. As previous studies (Capes et al 2008) the mass spectral fingerprint of BBOA looks like that of OOA after several days so this cannot be used to rule out biomass burning influence.

Response

We have added a new figure, Fig. 10d, to the manuscript, showing MODIS AOD for the boxed region of Fig. 10a, AERONET AOD at the Egbert site, and MODIS fire counts over the entire region of Fig. 10a for the period June 9 to June 16. Fire counts are neglibible until June 14. However, at this point, the wind abruptly shifts from north to south and Egbert samples the Toronto urban outflow; the fire emissions never reach the site.

Comment 31:

Page 18136 lines 21-22: Though lower temperatures tend to slow reaction rates and imply lower rates of photochemistry due to lower sun angle, they will favour partitioning – perhaps the authors should comment.

Response

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Here the discussion of lower temperatures refer to the monoterpene emissions rates, which increase exponentially with temperature, rather than SOA production processes (which, as the reviewer notes, have a more complicated temperature dependence). This has been clarified in the manuscript.

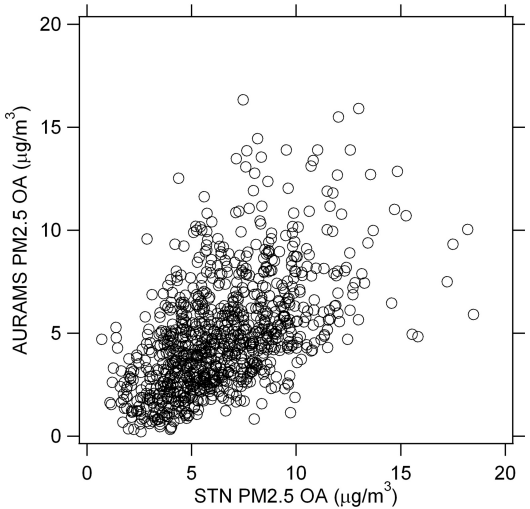
Comment 32:

Page 13186 line 29: Radiative impact not radiative forcing.

Response

This sentence has been removed from the paper. Where applicable, we now refer to a cooling/warming influence on climate.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 18113, 2009.



N=946 pts June-July 2007 STN	AURAMS V1.4.0
Model OA Average	4.8±2.7
Data OA Average	6.5±2.9
Slope	0.49±0.03
Intercept	1.7±0.2
Mean Bias	-1.7
RMSE	3.2
Correlation, R	0.51

Fig. 1. Comparison of AURAMS model and STN measurement data for eastern U.S.

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N=232 pts June-August IMPROVE	AURAMS V1.3.2
Model OA Average	0.24±0.17
Data OA Average	4.1±2.2
Slope	0.0235
Intercept	0.138
Mean Bias	-3.9
RMSE	4.5
Correlation, R	0.31

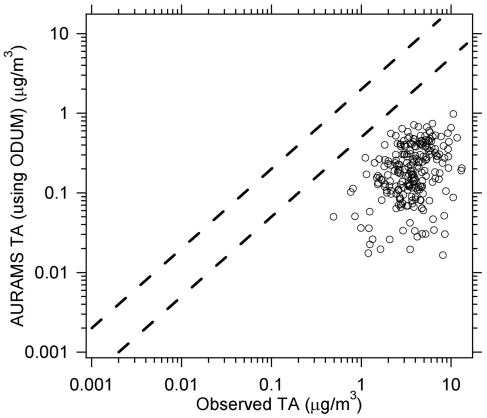

Fig. 2. Comparison of AURAMS model and IMPROVE measurement data for northeastern U.S.

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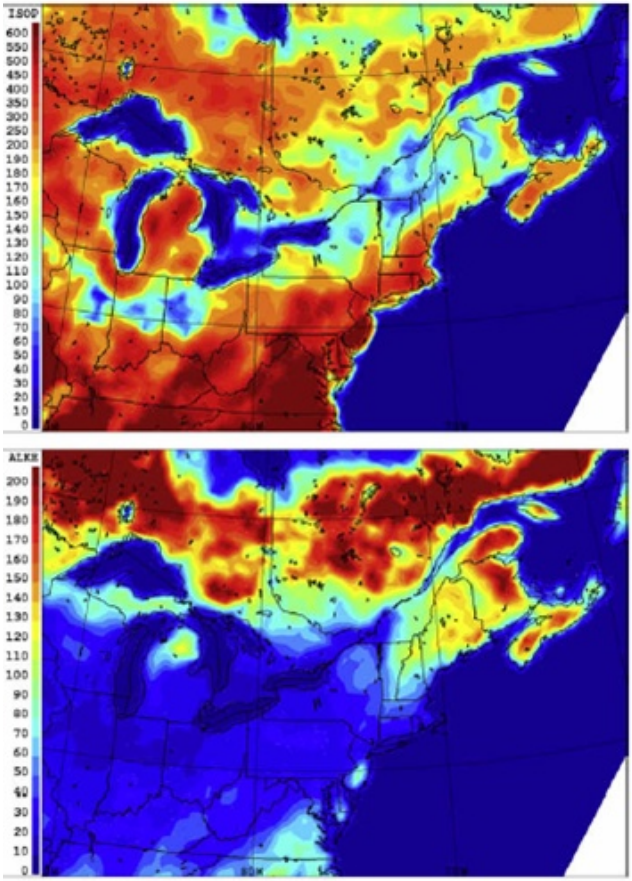

Fig. 3. Isoprene (ISOP, top) and monoterpene (ALKE, bottom) summertime emissions for eastern North America.

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