Response to Anonymous Referee #1 comments:

Specific comments

R1.1) Partly due to the length of the manuscript I may have missed some specific problems but here is at least a start. Page 30209, lines 4-5: "Dzepina et al. (2010) recently reported:" The sentence is difficult to understand, partly because the Dzepina-paper referred to is still not published (in review according to the reference list; and I have only been able to find an abstract for the paper); if I understand it correctly the Tsimpidi et al. (2010) mechanism does include SI-SOA and, if that is the case, the sentence does not make sense to me. In general I think the authors should try to limit the referencing of a paper "in review"; the Dzepina et al. (2010) paper is referred to three times in the manuscript.

The Dzepina et al. paper has now been published in Environ. Sci. Technol. and it can be downloaded from http://pubs.acs.org/doi/abs/10.1021/es103186f. Therefore we have kept the references to that paper as it does illustrate several points which are relevant to the present paper.

To clarify the specific point that is unclear to the reviewer, we have modified the text to read:

"However, Dzepina et al. (2010) recently reported that the Tsimpidi et al. (2010) A-V-SOA mechanism produces enough SOA to match the regional observations, and that a large SOA over-prediction is observed when SI-SOA is also implemented."

We note that the same SOA overprediction when implementing aging of A-V-SOA on top of SI-SOA is observed in Tsimpidi et al. (see their Figure 12) and Dzepina et al. (2011, see their Figure 1e).

R1.2) Chapter 2 Model description: Although I think this paper in general is far too long, the section about the introduction of semi/intermediate-volatility OA into the MOSAIC aerosol dynamics module is very short and very few details are given. If this really is the first time SI/SOA is coupled to the MOSAIC module it would be interesting to know some more details about how this was done (e.g., what extra modifications/assumptions are needed for the organic components, accommodation coefficients, impact on water uptake, etc); on page 30216, lines 22-27 you write: "The detailed treatment of OA has been fully integrated with the inorganic MOSAIC aerosol module in WRF-Chem, thus constituting a state-of-the-art representation of processes leading to organic and inorganic aerosol formation in the atmosphere. This detailed modeling approach in WRF-Chem is necessary to understand the suite of complex physical and chemical interactions between biogenic SOA formation, anthropogenic OA and inorganic species." But you give no details about how you have you have treated these "complex physical and chemical interactions" between the OA and the inorganic species. If the details are mostly technical and not interesting to the general reader they could be presented in Supplementary material to the article.

In the present version, the gas-particle partitioning of SOA species is treated as an instantaneous equilibrium process, while the gas-particle mass transfer of inorganic species is treated as a particle size-dependent dynamic process. We do not treat any chemical interaction between organic and inorganic species; however the organic and inorganic species are assumed to exist in

two separate phases within a given particle and are tied together in physical interactions such as dry deposition.
We have modified the description in the main text as:
"The detailed treatment of OA and the inorganic MOSAIC aerosol module in WRF-Chem, constitute a comprehensive representation of processes leading to organic and inorganic aerosol formation in the atmosphere. In the present version, the gas-particle partitioning of SOA species is treated as an instantaneous equilibrium process, while the gas-particle mass transfer of inorganic species is treated as a particle size-dependent dynamic process. Also, the organic and inorganic species are not allowed to interact with each other in the particle phase state and water uptake calculations. These assumptions will be relaxed in the future as experimentally derived parameterizations of complex physicochemical interactions between organic and inorganic species as well as dynamic condensation, evaporation, and reactive uptake of organic gases are implemented in the MOSAIC aerosol module."

R1.3) A general question regarding the comparisons of model concentrations to AMS-data: How are the size-distributed OA concentrations compared to the AMS-measurements? Are only the two smallest size bins included or also the third (0.625 – 2.5 um, dry diameter)? Is hygroscopic growth taken into account?

In these comparisons, the size bins 1-3 (0.039 - 2.5 µm dry diameter) are used for comparison to AMS data. The AMS at T0 used a dryer and the aircraft aerosol is effectively dried due to ram heating in the inlets. In WRF-Chem particle size bins denote the dry diameter. Note that AMS samples upto 1 um particles in diameter, while the first two size bins in WRF-Chem include particles upto 0.625 um dry diameter. In this work we chose to include the third size bin for comparison to AMS measurements. Fraction of mass in the third size (between 0.625 um and 2.5 um) was often less than 15% of total $PM_{2.5}$ OA mass, hence errors introduced are not significant.
We have added following lines to the main text:
"In WRF-Chem, the predicted OA in particle size bins 1-3 (0.039 - 2.5 µm dry diameter) is compared to AMS measurements. Fraction of mass in the third size bin (between 0.625 um and 2.5 um) was often less than 15% of total $PM_{2.5}$ OA mass."

R1.4) Page 30211, line 6: The SAPRC-99 reference is not correct. The link is not working. Please update to a correct reference for the mechanism. Also the comment in the reference is a bit strange: "last updated 30 March 2010"? As far as I understand the SAPRC-99 mechanism was replaced by SAPRC-07 in January 2010, so I wonder if there were any updates to SAPRC-99 in March 2010?

Corrected in the main text.

R1.5) Page 30213, lines 25-26: The anthropogenic emissions inventory includes traffic emissions and municipal trash burning. What about other anthropogenic emissions?

This sentence has been rewritten in the text to reflect other sources as:
"The anthropogenic emissions inventory includes traffic emissions and municipal trash burning as well as emissions from a wide range of point, and area sources".

R1.6) Page 30214, lines 1-4: "Also, municipal trash burning is expected to have similar OA spectra as fresh vehicular emissions dominated by hydrocarbon-like OA (HOA) with also some similarities with BBOA (Mohr et al., 2009); hence organic OA emissions from these sources are lumped together" What sources are lumped together? Trash-burning and vehicular emissions? Or Trashburning and BBOA?

This sentence has been corrected in the main text saying trash burning and vehicle emissions are lumped together. BBOA emitted from forest and agricultural fires is included in BBOA.

R1.7) Page 30214, lines 12-13: "Emission factors were taken from multiple sources (Andreae and Merlet , 2001; M. O. Andreae, personal communications, 2008)" This is not clear (and definitely not reproducible). I suggest that the details are specified in Supplementary Material.

The biomass emissions inventory was developed by Christine Wiedinmyer, see Wiedinmyer et al. (2006), cited in the manuscript for details of that methodology. Including details of how emission factors were calculated and used is beyond the scope of this work.

R1.8) Page 30216, lines 5-8: "Isoprene and terpene emissions calculated by the NEI emissions inventory for Mexico domain are removed to avoid double counting of biogenic emissions (already calculated by MEGAN model within the domain)." There is no reference to the NEI emissions inventory?

Yes, unfortunately there is no reference for the NEI emissions for Mexico. We have added a link that contains the available information on the inventory that was developed.

Added: http://mexiconei.blogspot.com/2007/01/national-emissions-inventory-now.html
to the text where NEI is first referred to.

R1.9) Why did you choose to use MEGAN emissions of isoprene also in the parts of the domain where you had another emission inventory that also included the anthropogenic isoprene? Was it impossible to combine the information in order to keep the, potentially important, anthropogenic isoprene emissions?

In order to avoid double counting we have chosen the widely used MEGAN model for biogenic emissions. Hodzic et al. (2009) quantified the influence of the anthropogenic isoprene and reported that B-V-SOA formed for this source is about 0.2 ug/m3 on average at T0 site in Mexico City during March 2006, which is a small contribution and does not affect the conclusions of our paper. As explained below the main reason for differences with BSOA predicted by Hodzic et al. is the use of much lower yields for biogenic precursors e.g. isoprene (factor of 3 lower values in this study for low NOx conditions, factor of 7 for high NOx conditions). In any case, the contribution of biogenic SOA (from anthropogenic and biogenic isoprene combined) is much smaller as compared to VBS SI-SOA as suggested by Hodzic et al. Since all modeling cases use the same isoprene emissions, excluding anthropogenic isoprene does not affect the main conclusions in the paper.

Recently, Hodzic et al. 2011 (Geosci. Model Dev. Disc., in press) has compared the BSOA formed by the parameterization used in Hodzic et al., 2009 based on the yields from Henze and Seinfeld, 2006 and Pun et al., 2006, and the parameterization used in the present study based on Lane/Tsimpidi, and have found a factor of 4-5 difference similar to our results. We have added following lines to the main text to explain these points:

"This also removes the anthropogenic isoprene emissions within the modeling domain. But, Hodzic et al. (2009) showed that contributions of SOA formed due to oxidation of anthropogenic isoprene precursors was low ~ 0.2 μg m^{-3} on an average during March 2006 at T0 site in Mexico City. As compared to Hodzic et al. (2009), this study predicts significantly lower V-SOA as discussed in the supporting information. This difference is chiefly due to lower yields for biogenic SOA precursors including isoprene in this study, as compared to parameterizations used by Hodzic et al. (2009) that were based on yields from Henze and Seinfield (2006) and Pun et al. (2006). However, V-SOA is expected to be a minor contributor to total SOA in the Mexico City region (Hodzic et al., 2010). Also, neglecting anthropogenic isoprene emissions do not affect the main objective of this paper (inter-comparison of different OA formulations), as all the formulations use the same isoprene emissions."

R1.10) 2.1.2.2 V-SOA formation You specify from where you have taken the SOA-yields for the AVOCs (Tsimpidi et al. and Hildebrandt et al.) but not for the BVOCs. I guess the ISOP, TERP and SESQ SOA yields were taken from Tsimpidi et al.?

Yes the isoprene, terpene and sesquiterpene yields are similar to Tsimpidi et al. and we have added this in the main text.

R1.11) Page 30219, lines 2-4: "The reaction rates of various VOC species with the OH radicals in Eq. (5) are already present within the SAPRC-99 mechanism, as a part of gas-phase chemistry" I can not find any SESQ reactions in SAPRC-99. What reaction rates were used for sesquiterpenes?

For sesquiterpenes we assumed same OH reaction rate as the terpenes. While reaction rate of sesquiterpenes with OH may be higher than terpenes, their emission rates are much lower than terpene species. We have added following lines to the main text:

"In this work, it is assumed that sesquiterpenes have the same OH reaction rate as the terpene species in SAPRC-99. While, sesquiterpenes may react much faster than terpenes, their emissions are significantly lower. Sesquiterpene concentrations within the modeling domain are atleast an order of magnitude lower than terpenes."

R1.12) Also, how did you treat SOA formation from ozonolysis? And what about reactions with NO3-radicals? Here you only mention reactions with OH-radicals.

In this work, we have only treated SOA formation due to OH reactions for V-SOA species. We recognize that this is a limitation and needs to be improved in the future on the basis of experimental data. We have added the following lines to the text to acknowledge this point:

"All SOA formation in this work is assumed to result from photo-chemical reaction with OH radicals. Reactions with O_3 and NO_3 radicals may also be important for SOA formation under certain conditions (e.g. Capouet et al., 2008), and they will be incorporated within WRF-Chem in the future."

R1.13) Chapter 2.2 Condensed 2-species OA mechanism Since this mechanism is constructed for computational efficiency I wonder why you have not neglected the 4 gas phase SI-SOA species, 4 of the 8 gas phase POA species and 16 of the 32 aerosol phase POA species (the low volatility species will remain in the aerosol phase under most atmospheric conditions and the high volatility species will be in the gas phase)?

We agree. Thanks for pointing this out. This will definitely reduce the computational cost further. In this work, we used the gas-particle partitioning code to decide the fraction of species in gas and particle phase as a function of its volatility. But assigning the low volatility species as non-volatile and the intermediate volatility species as volatile, will remove the need for solving gas-particle partitioning. This will also remove about 32 species as you point out and help reduce the advection costs. We will incorporate this in WRF-Chem in the future as you suggested.

R1.14) 2.2.2.2 V-SOA Page 30222, lines 25-26 (and Table 5): You state: "The SOA yields for alkane and olefin species are chosen to be equal to the yields corresponding to species with C* of1 ug m-3 in the 4-product VBS from Tsimpidi et al. (2010)." When I compare your Table 5 with the Tsimpidi's Table 2 it looks as if you have used the C*=1 ug m-3 values for the olefin species but not for the alkanes; for ALK4 andyou seem to use Tsimpidi0s values for the C*=10 ugm-3 species. Please correct the text and explain the reason for your different choices for the ALK and OLE species.

Done. We have added following lines to the text:
"For alkane species, since SOA yields corresponding to species with C^* of 1 ug m^{-3} are zero in Tsimpidi et al. (2010), yields from the next higher volatility species (C^* of 10 µg m^{-3}) are assigned to the lowest volatility species.

R1.15) Page 30223, lines 5-6: "to ensure atmospherically relevant conditions". Not a major point, but I would not say that you "ensure" atmospherically relevant conditions when using the smog chamber data from Ng et al. Relative humidities in the experiments are very low and even if you choose the experiments with the lowest aerosol generated it is still fairly high concentrations compared to the ambient atmosphere. Also, the Ng experiments only provide SOA yields for the OH-reaction. What do you assume for ozonolysis and reactions with NO3?

We agree. Low relative humidity experiments are not exactly representative of ambient conditions. We have removed the word "ensure" and modified the text as:
"All yields are chosen at lowest ΔM_0 values measured during the experiments, which are closer to ambient SOA concentrations."

As mentioned in response to R1.12, ozonolysis and NO3 reactions leading to SOA formation are not included.

R1.16) Page 30223, lines 9-11: Biogenic V-SOA is found to be a factor of 5 lower than what Hodzic et al. (2009) calculates and as a reason you give "mainly due to lower biogenic emissions in this work". But you have used MEGAN for biogenic emissions and Hodzic et al. also used the MEGAN model for biogenic emissions? So how can biogenic emissions differ so much that the difference in BSOA is a factor of five? I guess there must be other more important reasons? On page 30216 you state that you have neglected the Anthropogenic isoprene in your run (and that the anthropogenic isoprene could be a factor of 5 higher than the biogenic isoprene at the T0 site) but missing Anthropogenic isoprene should not have a significant impact on Biogenic SOA and you also quote that the largest contribution to biogenic SOA in the Mexico City basin is due to regional isoprene (>60%) produced upwind on the coastal area and advected into the city. It is also not clear if the factor of five difference in BSOA (compared to Hodzic et al.) is for all of the model domain or only at certain site(s).

The difference is due to the use of different B-V-SOA parameterizations which have several-fold differences in SOA yields, as discussed in response to comment R1.9 above.

R1.17) Page 30223, lines 23-24:"Also, the computational burden of handling the cloud processes would be unnecessarily excessive." This statement is a bit worrying; if you want to be able to use the model for larger scale modeling or longer time periods and/or wetter regions you really have to include wet deposition. How much would the computational burden increase by including wet deposition?

Wet deposition by itself is not that computationally expensive. However, in WRF-Chem wet deposition is currently coupled with cloud-aerosol interactions and requires parallel 'cloud-borne' species for many interstitial species in the aerosol model. This results in increasing the computational time by a factor of ~1.5. We first wanted to establish whether a 2-species VBS mechanism is a promising approach, before tackling issues on how to handle SOA with clouds. The improvements you suggested earlier would help us reduce 32 species and bypass the gas-particle partitioning code. This would also help improve the computational cost further, making the 2-species VBS more suitable for simulations including cloud processes. That's another reason why we have to extensively evaluate the 2-species VBS for similarity to the 9-species VBS in terms of both SOA mass and O:C ratios.

We added the following lines:
"Also, the computational burden of handling the cloud processes would be excessive in the 9-species VBS formulation due to the need to transport both interstitial and cloud-borne copies of each aerosol species. This would almost double the cost of the simulation. The condensed 2-species VBS formulation developed in this work is more suitable for complex cloud-aerosol interactions in WRF-Chem."

R1.18) Chapter 3.1

Page 30226, line 2 (and Figure 1): "Figure 1 shows total SOA surface concentrations. Is this correct? Does Figure 1 show only SOA (that is, not including POA)?

Yes that is correct. Figure 1 shows only SOA (not including POA).

R1.19) Page 30226, lines 22-26. The discussion about the B-V-SOA predictions is repeated in some more detail than above; see my comments above also for this part (if >60% of the BSOA is due to regional isoprene, produced upwind and advected into the city how can there be a factor of 5 lower BSOA at both T0 and T1 in this study compared to Hodzic et al. if the regional isoprene emissions are from MEGAN in both cases?). And, do you consider Anthropogenic isoprene as a source of Biogenic SOA? This is a bit strange I think and, if it is so, it needs to be pointed out in the text.

The difference is due to the use of different B-V-SOA parameterizations which have several-fold differences in SOA yields, as discussed in response to comment R1.9.

R1.20) Page 30227, lines 13-14: CO measurements are taken from a "nearby operational monitoring site" How near is this operational monitoring site? And does it have a similar character as the T0-site (closeness to major emission sources, geographical features etc). I find it a bit strange that you choose to almost exclusively present the model measurement comparisons for OA in terms of enhancement ratios with respect to CO (considering that you do not have CO measurements at T0 and the discussion in section 3.2.2 about the complicated interpretation of night-time CO data leading to "more uncertainties during night as compared to day-time"). I would have preferred to see more straightforward comparisons of modelled OA to measured OA.

The nearby site is within 3 km of T0 site and is still in the urban core, so it should have similar characteristics as T0. We have also taken the reviewer's point and present model measurement comparisons of OA directly without normalizing to CO throughout the manuscript.

R1.21) Page 30228, lines 18-20: "Model peaks do not result from night-time chemistry with O3 or NO3 radicals, since night-time chemistry leading to SOA formation is not represented in WRF-Chem." Does this mean that you do not include SOA formation from, e.g., ozonolysis of terpenes? Ozonolysis is estimated to be an important SOA-source (see, e.g., Capouet et al., JGR 2008). Why did you not include night-time SOA-chemistry in the model simulations?

As discussed in response to comment R1.12, ozonolysis and NO3 chemistry are not included, and this will be a subject for future work.

R1.22) Page 30229, line 7: "consistent with the underprediction of SOA/DCO in Fig. 3d". What do you mean by this? I guess this part of the sentence could just be skipped?

Removed. Again, note that we have removed normalization to CO in both main text and supplemental information.

R1.23) Page 30230, line 9 (and Figure 4): What is included in BBOA in Figure 4? Only primary BBOA or also oxidised BB-SI-SOA? Is BB-V-SOA included in the WRF-Chem model (and, if so, is it included in BBOA in Figure 4 or is it included in some of the other V-SOA fractions)?

In Table 1 we have explained that BBOA is biomass burning POA only. BB-SI-SOA is included as a part of total SOA in Figure 4. In our terminology we do not have BB-V-SOA, but we assume the reviewer means SOA from oxidation of VOCs emitted during biomass burning. If that's the case they are a part of A-V-SOA as discussed in Table 1, and they are a part of total SOA (not BBOA). However Hodzic et al. (2009) showed that BB-V-SOA is very small during MILAGRO, when using current inventories of VOC emissions from biomass burning.

R1.24) Page 30230, line 22: How do you handle the vertical emission distribution for BBOA (and other emissions)? Do you use some plume-rise calculation scheme for hot emissions?

We added the following lines to the text explaining this:
"In this work, fire emission injection heights are treated the same as was described in Fast et al. (2009), Section 3.3. Emissions from fires are distributed uniformly within ~300 m of the ground since insufficient information (e.g. fire temperature) was available to compute plume rise, and based on visual observations from aircraft of low plume rise for these fires (R. Yokelson, pers. comm., 2008)."

R1.25) Figure 4c: Observed concentrations are missing in the Figure. I definitely think that observed concentrations should be included here!

We disagree with the reviewer on this point. The point of Figure 4 is comparing model predictions at surface level, column burdens and vertical distributions. Comparison with measurements in done in Figure 3 and Table 3 in the revised paper.

R1.26) Page 30232, lines 15-17: You state that the agreement between case 3 and 2 is "very interesting" and still you do not show the results for case 3: : : If it really is very interesting you should perhaps consider illustrating these similarities (instead of a number of other less interesting plots)?

Point taken. We have included Case 3 in Figure 5a now.

R1.27) Page 30233, lines 1-16: I do not think that this section contains very interesting results and I do not see why it is included in the chapter about results at the T0-site (since it discusses results along flight tracks and not city center measurements). Also, it is difficult to see the details in Figures 5c and 5d (and if they are to be included I think you should change the x-axis to local time instead of UTC, or at least mention the time difference between local time and UTC in the figure caption).

We disagree with the reviewer on this point. This figure and discussions regarding O:C ratios on page 30233 are extremely relevant to the paper. O:C ratios are very relevant since they provide another measure to verify the performance of SOA treatments, rather than just using OA mass and its components. Relationships between O:C ratios and hygroscopicity (as discussed in the

introduction) have been found and could be useful in developing future relationships between SOA and CCN activation, and consequently aerosol indirect forcing. We believe it is useful to discuss all the O:C ratio comparisons in one location when it is introduced in section for the T0 surface site. There are only 2 other aircraft flights with this information. We have changed the X-axis to local time and also added following lines to the main text:

"In Figure 5, we evaluate modeled O:C ratio predictions in terms of 20-day diurnal variations at the T0 site, time averaged spatial variation across the modeling domain and along two C-130 flight transects in the atmosphere."

R1.28) Chapter 3.4 What particle size is evaluated at the T2 site (PM2.5? PM10?) Since the measurements at T2 are OC/EC measurements I do not understand why you first convert the observations to "TOTOA" using an assumed OM/OC ratio of 1.4? This just introduces a big, unnecessary, uncertainty into the comparison. You couldeasily compare your calculated OC concentrations to the observed OC. This needs to be changed. What OM/OC ratio does your model give at T2?

This section is no longer a part of the main text and has been moved to the supporting information. But our calculated OC are also derived from OM using measured OM:OC ratios of each component (see Aiken et al., ES&T, 2008) as discussed in the main text, and not a constant factor of 1.4 as was often done in older studies. In addition biogenic and traditional anthropogenic SOA in the model would need to be converted to OC assuming OM:OC ratios. So we disagree that this method would be less uncertain, it seems to us that the same uncertainties in OM/OC ratios would enter in the calculation either way. We added the following lines in the supporting information explaining this:

"Another way would be to derive OC from WRF-Chem predictions for direct comparison to measured OC. However, both methods have uncertainties associated with OM/OC ratio, as OC emissions in the inventory are derived assuming fixed OM/OC of 1.25 for fossil and 1.57 for biomass emissions. In addition, traditional biogenic and anthropogenic SOA in WRF-Chem need to be converted to OC. In this work, we choose the former method of comparing OA instead of OC, acknowledging the inherent uncertainties with both approaches"

R1.29) Figure 7c. The curve showing measurements looks surprisingly spiky, for being a 24-day average of AMS-measurements. How did you calculate the averages? Is it 24-day averages of hourly mean concentrations? (I guess that the AMS-data have a very high time-resolution and that you, in principle, could calculate a more continuous curve?) The model curves look much more continuous and seem to have higher resolution than 1h? The same question is also valid for Figure 6. Several of the AMS-curves look a bit "unsmooth"; are these curves also 24-day averages and why are they not calculated with higher time resolution than 1h?

Figure 7 has been removed from the main text into the supplementary material. We have revised our calculations for Figure 6 for the PMF data. The way we calculated averages is :

1. Use high resolution AMS PMF data
2. Average them using 1 hour interval values. For example all values between 0030 and 0130 hours on all days are averaged and assigned to 0100 hour.

The higher time resolution AMS data is only one reason contributing to the curves that are not totally smooth. The other reason is that there are more day-to-day variations at certain hours of the day than represented by the model. Day-to-day variations in actual emissions could be a factor contributing to the apparent spikiness in the AMS data since the anthropogenic model emissions are the same every day, except for differences between weekday and weekend periods. Also a main cause of the spikiness in the early morning hours is the arrival of discrete very large biomass burning plumes on specific days (see e.g. Aiken et al., ACP 2010), the effect of which is not removed by the 3-week averaging used here. This is an example of noise in an average due to limited sampling of a skewed distribution. If there were more days sampled, computing a diurnal variation in PMF components may look somewhat smoother.

R1.30) Chapter 3.5 The AMS-data used in the comparisons are 10s-data. What time resolution is the model calculated concentrations? What about the temporal variation of emissions in the model? Do you use hourly variations in emissions or something more sophisticated? I am not surprised that the model does not show the same scatter as a set of 10s AMS data. In itself, this does not highlight "significant uncertainties in representation" of temporal (or spatial) variation of emission sources. You could never expect a 3D-emission inventory to have a 10s-time resolution and I do not think that Figures 8a-d, and the long discussion about them in the manuscript, are very interesting (thus I think this (page 30236-30237) is one of the parts that can be removed from the manuscript; or moved to the supplementary if the authors prefer that). Did you preparesimilar scatterplots for 10s-AMS data for any of the surface measurements (and did that show similar differences in scatter)?

Yes we use hourly variation of emissions in the model. In this figure, model values at hourly intervals have been interpolated to the 10-s sampling periods. The reviewer is correct that the model cannot be expected to represent the 10-s variability seen in the data. However, we have averaged the data to 60-s. Given the aircraft speed of ~100 m/s, a 60-s sampling interval is made over ~6 km which is more comparable to our 3 km grid spacing. Even when the data is averaged to 60 s, there is scatter that is not represented by the model. The scatter plots in Figure 8 and their discussions are now moved to supplementary information as suggested.

R1.31) Figure 9 is much more interesting since it compares the different model versions to each other and to the measurements.
Agreed.

R1.32) Page 30240, lines 2-6: The sentence starting by "Because both total OA and SOA are overpredicted: : : " is rather confusing and I think that it should be rephrased (or it could also be removed since it just describes some ideas about how to try to tune the model to get better results).

Removed as suggested.

R1.33) Figures 10 and 11: These figures need much better figure captions. I think it should be possible to get some idea of what is shown from the figure caption without having to search the text for the section that deals with it.

We are not sure which specific details the reviewer is referring to. We have added some additional definitions for some of the acronyms uses in the figure caption.

R1.34) Figure 10 b and d: Case 1 and Case 3 give almost identical results, which I find very surprising since these two cases are so different. Please explain this (I guess that there may be an erroneous labelling in the Figure; in Figure 10c Case 2 and Case 3 are rather similar (as expected))

Figure 10 is now Figure 7 in the main text. Figure 7c is HOA which represents POA emissions. So it is more straightforward to see that Case 1 is lower than Case 2 and 3 as it has half the anthropogenic POA and related S/IVOC emissions. Figure 7b and 7d are consistent with each other as SOA is dominant component of OA at downwind locations. If we compare Case 1 and Case 3, we note that Case 1 has half anthropogenic POA and related S/IVOC emissions as compared to Case 3. But Case 3 has 7 times slower OH reaction rates as compared to Case 1. Also in Case 3, 50% oxygen mass is added while 15% oxygen mass is added in Case 1, after every generation of oxidation. There are significant differences in emissions, reaction rates, added oxygen mass and volatility distribution between Case 1 and Case 3, with one effect offsetting the other when it comes to SOA formation. The apparent similarity between Case 1 and Case 3 for the given flight is just a matter of chance. This is the reason why model-measurement evaluations need to be done at many locations and times throughout the modeling domain.

R1.35) Chapter 3.6, about Non-fossil carbon fraction, contains no interesting information and I think it should be excluded from the paper. The available observations give somewhat conflicting results and the model results presented in this paper are not (at this stage) useful for resolving the issue. You use some rough estimates of the fraction of nonfossil carbon in the "urban carbon" and states "15% of biogenic SOA is PBAP", which I do not understand at all; do you consider 15% of the biogenic secondary organic aerosol to be Primary Biological Aerosol Particles? What exactly do you mean by this? Also, the biomass burning emissions in this study are apparently too low and, according to the authors, the B-V-SOA is too low and to handle this, the fraction of non-fossil carbon is adjusted by increasing B-V-SOA by a factor of 5! Since the whole chapter 3.6 is unimportant for the main conclusions of this paper (and since it is does not give any additional conclusions/results and seems to be mostly rather rough "guess-work"). I think that it should not be included.

We have moved this section to the Supplementary Information section of the paper. We think that this section does have some value and prefer to retain it there, where it can also serve as reference for future studies.

R1.36) Figure 12. I think this should also be removed (partly because I think the whole chapter about non-fossil carbon should be excluded and partly because I do not understand why it is interesting to show diurnal variations of fNF anyway).

This figure has been moved to the Supp. Info. as discussed in response to the previous comment.

R1.37) This also means that I suggest that you remove the discussion-section, starting on Page 30246, line 25 and ending on Page 30247, line 5. This part also clearly shows that the whole chapter about non-fossil carbon can "the overall conclusion is unclear: : :" Chapter 3.7, which contains a comparison between the WRF-Chem and CHIMERE models, could also be removed from the paper with no loss of essential information. The differences between the two models are large (different emissions, different chemistry schemes, different VBS-schemes, different meteorological drivers) which makes comparisons of results rather difficult to interpret and, to be honest, I do not think the comparison presented in Chapter 3.7 and Figures 13-15 gives any useful/interesting information.

While the comparisons of WRF-Chem and CHIMERE are not central point of the paper, we have retained them in the supplementary material. We feel that new treatments (be it SOA or other processes) need to be compared against previous model results whenever possible. This provides a much better understanding of differences in performance than can be obtained by simply comparing results from separate papers.

We have added following lines of text in the main text with referring to the Supporting Information for the interested readers:
"Section S3.0 in the supporting online information compares predictions from WRF-Chem using 9-species VBS (Case 2) against CHIMERE model predictions described by Hodzic et al. (2010). The main differences include the following: (1) WRF-Chem uses online meteorology and CHIMERE uses offline meteorology, as discussed earlier, (2) the CHIMERE model includes the wet deposition of aerosols, whereas WRF-Chem does not, and (3) the emissions of POA are based on two different inventories. Differences in coupling of different processes between online and offline air quality modeling, as discussed by Grell et al. (2004), likely contribute to some of the differences in the chemical fields between WRF-Chem and CHIMERE. Additional differences in gas-phase chemistry, dry deposition, and implementation of VBS between the two models are discussed in section S3.0. Section S3.0 shows that predictions of WRF-Chem and CHIMERE are comparable at the T0 site, while WRF-Chem predicts on average 50% higher (and closer to observed) SOA than CHIMERE at the T1 site."

R1.38) Page 30246, lines 23-24: "Dry deposition of S/IVOC vapors downwind would reduce SOA formation bringing model predictions closer to AMS PMF analysis" But in chapter 2.3 you state that dry deposition is calculated for all gas-phase SOA precursor species? Did you NOT include dry deposition of S/IVOC in the gas phase?

Yes dry deposition of S/IVOC is included in gas phase. We have clarified what was meant and also added a reference:

"In addition, the effect of loss mechanisms such as dry deposition of S/IVOC vapors downwind needs to be quantified experimentally. Karl et al. (2010) recently showed that dry deposition of oxygenated VOCs is substantially larger than previously assumed for deciduous ecosystems. Models need to account for changing dry deposition as a function of photochemical aging of

organics in the atmosphere. Accurate representation of all physical and chemical processes affecting OA is necessary to get the right answers for the right reasons in climate models."

R1.39) Page 30248, lines 21-23: "The computational expense can be reduced further if sources of organic aerosols and O:C ratios are not required." This is true also for the 9-species VBS (not only the 2-species). Also, it would be interesting to know how much the expense would be reduced by these simplifications (though I can not see that you would be willing to neglect the O:C ratios in many applications).

Yes we agree it is true both for 9-species and 2-species VBS. We also agree that O:C ratios are required in many applications.

R1.40) To save space (and for ease of comparison) I suggest that Tables 3 and 5 are combined into a single table.

Table 3 and 5 are both moved to supporting info to save space. Tables 2 and 4 have been combined. We just have 3 Tables in the main text now.

R1.41) Technical corrections

All corrected.

Page 30219, line 20: "Table 2" should be: Table 3 Page 30219, line 25: "Table 1" should be: Table 2 Page 30220, line 13-15: the sentence about POA(a) and SI-SOA(a) is a bit strange; I thought that the authors nomenclature was that (a) meant aerosol-phase (that is particulate)? If so, it does not make sense to write that POA(a) and SI-SOA(a) would remainbe excluded without loss of contentin the aerosol phase under most atmospheric conditions: : : The POA(a) with C*=1E5 ug m-3 will not stay in the aerosol phase under most atmospheric conditions.

This is point is taken. Changed the main text as:
"Note that POA $(a)_{i=2,e,x}$ would almost entirely remain in the gas phase under most atmospheric conditions due to its high volatility."

Page 30231, line 6: across the depth of the boundary layer
Page 30231, line 19: of the boundary layer
Page 30238, line 4: of the 2-species VBS
Page 30238, line 6: lower than the other 2 modelling cases
Page 30239, lines 21-24: The sentence starting with "These include V-SOA" could be removed since the definitions are given (several times) earlier in the paper.
Page 30240, lines 26-27: as compared to the other two cases
Page 30240, line 28: suggests revising the SVOC
Page 30241, line 1: or the volatility distribution of the emissions
Page 30246, line 7: In addition, models need to capture
Page 30247, line 12: difficulty : : : the AMS collection efficiency
Page 30248, line 12: to a 9-species

Page 30265, last sentence: as discussed in the text. A general style comment: I think many sections in the text are too long. This makes the text unnecessarily difficult/tedious to read. In many cases it would be easy to just split the long sections into several parts (but I think the paper needs to be condensed/ rewritten anyway so perhaps the overly long sections will disappear then). One of many examples of very long sections is starting on Page 30247, line 6 and ends on

Since many sections and 7 figures have been moved to supporting information, the main paper is much shorter now. We hope the reviewer finds these revisions satisfactory.