Response to Referee #2

We are grateful to the reviewer for their helpful comments and guidance that have led to important improvements of the original manuscript. Our point-by-point responses are listed below. Reviewer's comments are in black font, and authors' responses are in dark blue. Page and line numbers refer to discussion paper *Atmos. Chem. Phys. Discuss., 15, 23231-23277, 2015*.

This paper describes a modeling study of SOA formation and aging using the NCAR CAM4-chem model implementing a new 4-product VBS scheme with different aging parameterizing and comparing to the standard 2-product model, turning on and off aging or high/low-NOx chemistry and reducing NOx. The differences in total OA, type of OA (POA/HOA, SOA/OOA), spatial and vertical distributions, changes in relative contributions from the different VOC-type + low/high NOx pathways are compared within the model and to IMPROVE filter and online AMS aircraft measurements. Small differences (and slight improvement with comparisons) are shown for using the 4-product model and large increases in SOA are shown for the aging scheme. 50 percent NOx reductions result in insignificant changes for global, SE US, and Amazonia SOA. Discussions of compensating effects on oxidants and SOA yields are discussed to explain the lack of overall sensitivity to NOx reductions.

The manuscript is generally well written and describes a clear and straightforward modeling study exploring basic chemical parameterizations and comparisons to observations. This manuscript warrants publication in ACP after relatively minor revisions.

General comments followed by a detailed list of comments are below.

General/Main Comments:
The title and abstract should include more about the results from the different VBS schemes, aging, and comparison to measurements. The NO_x sensitivity study is really only one part of the study. as presented in the paper. The summary reflects a better balance of the overall study investigations and results.

Response: In the abstract, we include the main results of updated SOA model and comparison with observations. To be more sufficient, we add: "We have updated the SOA scheme in the global NCAR Community Atmospheric Model version 4 with chemistry (CAM4-chem) by implementing a 4-product Volatility Basis Set (VBS) scheme, including NO_x-dependent SOA yields and aging parameterizations. **Small differences are found for the no-aging VBS and 2-product schemes; large increases in SOA production and SOA-to-OA ratio are found for the aging scheme.** The predicted organic aerosol amounts capture both the magnitude and distribution of US surface annual mean measurements from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network by 50%, and the simulated vertical profiles are within a factor of two compared to Aerosol Mass Spectrometer (AMS) measurements from 13 aircraft-based field campaigns across different region and seasons."

We do not change the title because the effect of NO_x reduction is the main application result in this study, and this conclusion holds for every SOA scheme, no matter whether or not using VBS framework, including NO_x dependency or adding aging effects.

In a number of instances describing the model setup, certain important choices were not explained. E.g. Why was a high-NO_x isoprene pathway not included? Why use such a high, unjustified value of OM/OC for POA? Why is oxygen not added during aging? Such omissions make the reader wonder if the agreement and differences are more of an artifact of such choices rather than a reflection of the actual SOA chemistry in the atmosphere.

Response: The VBS scheme we implemented in CAM4-chem is based on Pye et al. (2011), which does not include the high-NO_x isoprene pathway. To account for this omission, we performed an additional simulation that includes the high-NO_x isoprene pathway using the VBS parameters from Lane et al. (2008), and discuss the results in the Supplement.

We originally chose the POA-to-POC ratio of 2.1 based on Turpin and Lim (2001), which suggests a factor of 2.1 ± 0.2 for nonurban organic aerosols. Now, we have changed the POA-to-POC ratio to a traditionally used value of 1.4, because POA has a lower OA-to-OC ratio than nonurban aerosols (dominated by SOA). Fig. 4 to Fig. 7 and related statistics in the main text have been modified accordingly.

We do not account for the added oxygen when aging due to the large uncertainties in the aging processes and the complexity of various SOA species. The change of O/C ratio is not the focus of this study because we do not have sufficient observational evidence to validate the simulated O/C ratio in the global model. We choose a high aging parameter $k_{OH} = 4\times10^{-11}$ cm^3 $molec^{-1}$ s^{-1} to provide an upper limit of SOA estimation, which would partly offset the omission of added oxygen mass during aging.

Modifications include:
At P23238 L26, we delete "and assumes a POA-to-OC ratio of 2.1…", and add: "Later in Section 4.2, we assume a POA-to-POC ratio of 1.4 (Aiken et al., 2008; White and Roberts, 1977) to calculate modeling POA and OA to compare with observations."

At P23239 L21, after "SOA formed from OH-initiated photooxidation of isoprene still only has one set of yields following the low-NO_x parameterizations", we add "We do not change this isoprene-SOA parameterization to remain consistent with the VBS framework from Pye et al. (2010). Additional simulations that include the high-NO_x pathway of isoprene chemistry are discussed in the Supplement."

At P23240 L19, after "The oxygen-to-carbon ratio (O:C) is assumed to be constant for each surrogate SOA product thus increase in SOA mass due to the addition of oxygen is not considered in the aging process", we add: "Considering the complexity of various

SOA species and the large uncertainties in aging process, the assumption of fixed O:C ratio for each SOA product surrogate is acceptable for global model parameterizations."

Can the authors add a short discussion of the significance of omitting high-NO pathway for isoprene oxidation and how that may be effecting the results?

Response: We performed additional simulations that include the high NO pathway for isoprene oxidation and add a discussion in the Supplement as follows.

"The VBS schemes (VBS, VBS_agHigh, etc.) that we implement into CAM4-chem as described in the main text are based on the published VBS parameterizations from Pye et al. (2010), which does not consider the high-NO_x pathway for isoprene oxidation. Here we performed an additional simulation "VBS_agHigh_isop" to evaluate the effect of this omitted pathway.

"In the VBS_agHigh_isop scheme, we add the high-NO_x branch for isoprene-oxidized SOA formation ($ISOPO_2$ + NO) using the parameters from Lane et al. (2008). The SOA mass yields are 0, 0.00026, 0.0195, 0.013 for C* of 0.1, 1, 10, 100 μg m^{-3}, respectively, fit using a density of 1.3 g cm^{-3} to be consistent with Pye et al. (2010). Adding this pathway, the VBS_agHigh_isop scheme gives an annual mean SOA burden of 0.86 ± 0.05 Tg[C] and a net SOA production of 46.8 ± 1.9 Tg[C] per year, both of which are about 20% smaller than the VBS_agHigh scheme, and larger than the two no-aging schemes (2-product and VBS). The VBS_agHigh_isop scheme shows no significant advantage over the VBS_agHigh scheme in model-observation comparisons.

"We then performed a sensitivity run using the VBS_agHigh_isop scheme, in which anthropogenic NO emissions are reduced by 50%. Results in the southeast US and the Amazon are shown in Fig. S5. SOAM and anthropogenic SOAs through each branch behave similar to the VBS_agHigh. For isoprene oxidation in the southeast US, the HO_2 pathway increases and NO pathway decreases. While the effect of shifting to high-yield HO_2 pathway is masked due to the reduced OH concentration (about -15%, Fig. S2). The total SOA concentration decreases from 3.4 ± 0.5 to 3.0 ± 0.4 μg m^{-3}. This 0.4 μg m^{-3}, 10% reduction is smaller than the change in VBS_agHigh scheme (0.5 μg m^{-3}, 12%), and is smaller than one standard deviation of multi-year variation, suggesting the SOA response to the NO perturbation is not significant. In the Amazon, all branches show a decrease with reduced NO. The total SOA reduction (0.1 μg m^{-3}, 2%) is non-significant. These conclusions are consistent with the VBS_agHigh results."

There are a handful of instances where the authors speculate on the reasoning for specific model output results.

For example in Section 4.2.3, it stated: "Consistent with the comparison with the IMPROVE network in Sect. 4.2.1, the models overestimate POA in most regions especially in North America, which will likely increase SOA production due to the larger aerosol surface area available for condensation."

Or in Section 4.3 it is stated: "The dependence of SOA on oxidant concentration indicates a maximum at medium oxidant level of approximately 0.8e12 molecules cm-3. The low SOA concentration at high oxidant level might be explained by, again, the lower yields of high-NOx pathway, which are associated with high NOx and oxidant levels."

It would seem that rather than speculate on such causes/effects, these hypotheses can be tested with sensitivity studies using the model and speculation should be reserved for instances when there is no further information available. The authors have done a nice job of presenting, deconstructing and discussing much of the results however cases like these examples (and a few others) seem to stop short of using the information the model provides to the full extent possible. I.e. the answer to these speculations is most certainly available from the model – whereas, whether what is happening in the model truly represents certain processes in the atmosphere could be speculated upon.

Response: We appreciate the reviewer's comments about the speculations. We make several modifications in the manuscript.

In Section 4.2.3, the major influence is more OA mass to promote condensation of semi-volatile species. At P23246 L24-25, we delete "… which will increase SOA production due to the larger aerosol surface area available for condensation", and add "which will promote condensation of semivolatiles onto pre-existing organic matter thus forming more SOA".

In Section 4.3 and Fig. 8, we have re-examined the model outputs and find that the high SOA concentrations associated with relative low branching ratios and medium oxidant levels are mostly from tropical rainforests, where there are large emissions of precursor VOCs. Therefore we modify the paragraph as follows:

At P23248 L4, we delete "which may reflect the fact that low-NO_x pathway has higher SOA yields", and replace by "which mostly locate in tropical rain forests with large BVOC emissions and high SOA production efficiency through the low-NO_x pathway".

At P23248 L9, we delete "might be explained by, again, the lower yields of high-NO_x pathway, which is associated with high NO_x and oxidants levels", and replace by "mostly occur in polluted regions where SOA production is overwhelmingly dominated by the high-NO_x (low-yields) pathway".

Detailed Comments:
P23233, L8: remove extra period

Response: corrected.

P23234, L18,19: missing article - add "the" before "low-NOx" and "high NOx" or make "pathway" plural.

Response: corrected.

P23234, L17-19: Is generally true? According to the Pankow SIMPOL model (see Table 1 in Kroll and Seinfeld, Atmos Environ 2008) nitrate functional groups lower vapor pressure more than hydroperoxy groups.

Response: The organic nitrates can be low in volatility. The high-NO_x pathway has lower yields because organic nitrates ($RONO_2$) are not the dominant product of the RO_2 + NO reaction channel. The major channel is alkoxy formation (RO_2+NO->RO+NO_2) which does not directly form SOA. Our previous statement was not precise.

At P23234 L17, we delete "… their oxidation products ROOH formed from the low-NO_x pathway have a lower volatility than the oxidation products $RONO_2$ under the high-NO_x pathway, thus are more likely to condense to form SOA." Instead we add: "… both the ROOH groups and the $RONO_2$ groups can be low in volatility thus facilitating SOA formation, but $RONO_2$ is not the dominant product of the $RO_2 + NO$ branch, therefore the high-NO_x pathway usually has lower yields of SOA."

P23234, L20: Delete "the"

Response: corrected.

P23234, L20: "unique chemical signature" seems a bit strong/misleading. As pointed out later in that sentence, $RONO_2$ are formed through a different (and often comparable pathway).

Response: We replace the word "unique" by "distinctive".

P23234, L25: make "contributes" past tense. This is not necessarily generally true for that location (i.e. study was done in spring/summer. Certainly may not be the case in winter).

Response: corrected.

P23234, L25: delete "to" before "approximately". Otherwise it reads that it comprises part of a third.

Response: corrected.

P23234, L26-L2 next page: This logic is a bit non-sequitur in that it notes the effect of NO_x on O_3 in low-NO_x conditions followed a statement that seems to rely on low-NO_x conditions dominating (i.e. increase NO_x, increase OH, O_3). Such a statement should not be made without presenting evidence to support such a connection.

Response: At the beginning of the sentence, we clarify that the positive relationship between NO_x and oxidants (i.e. increase NO_x, increase OH, O_3) happens "in the NO_x-limited regime". This is part of well-understood NO_x gas-phase chemistry. Later in the manuscript (e.g. Section 4.3 and Fig. S2), we have shown the positive relationship between NO_x and oxidants at global scale and in the two target regions, the southeast US and the Amazon, confirming that both the two regions are in the NO_x-limited regime in terms of ozone formation.

At P23234 L26, We rephrase the sentence here and add a reference book: "In the NO_x-limited regime **(in terms of O_3 formation)**, the OH-initiated oxidation of CO, methane (CH_4) and other VOCs in the presence of NO_x produces O_3. Thus **in such conditions**, increasing NO_x by human activities should, in principle, lead to the increase in atmospheric oxidation capacity (OH and O_3) **(Seinfeld and Pandis, 2006)**, and result in higher SOA yields."

At P23247 L21, we modify: "…both of which are mostly in the NO_x-limited regime **in terms of ozone formation** due to their large BVOC emissions (Lane et al., 2008), **i.e. the concentration of O_3 and OH are positively related to concentration of NO_x."**

P23236, L1: "total organic matter" could be confused by some as including gas-phase. Clarify that is meant to apply only to the particle phase.

Response: we add "total **particle phase organic matter**".

P23236, L2: "usually"? It's this always the case?

Response: Yes the term OC always refers to organic carbon in atmospheric chemistry. We delete the word "usually".

P23236, L11-12: Can you provide a reference for the IMPROVE-OC filter analysis? I thought that was an offline technique where the filters were brought back to the lab for analysis. This is not a minor detail as leaving filters around for extended periods, transporting, and analyzing later may impose substantial biases due to evaporation of semi-volatiles or unwanted chemistry. Also, say what the method actually is so the reader doesn't have to go dig up the documentation to get a general sense.

Response: At P23236 L11, we delete "IMPROVE-OC is measured by semi-online filter analyzer", and add "IMPROVE OC is collected using quartz fiber filters for 24 hours every third day, analyzed offline by thermal optical reflectance (TOR) (Chow et al., 1993), and corrected for an approximate positive artifact (Dillner et al., 2009). Assumptions made in this correction may not always be appropriate (Watson et al., 2009), and the potential negative artifacts due to the volatilization of particulate organics are not accounted."

P23236, L20: Specify STP. This varies.

Response: We add: "… in standard temperature and pressure conditions **(STP: 298K, 1atm)**".

P23237, L5-7: Provide references for PMF factors.

Response: The 42 AMS surface measurements are summarized in Spracklen et al. (2011) and Zhang et al. (2007), and use a multiple component analysis (MCA). This MCA method uses a different algorithm to solve the same mathematical problem as the positive matric factorization (PMF) model. There results are in general agreement (Canagaratna et al., 2007; Lanz et al., 2007).

At P23237, we add: "The HOA and OOA are determined by a multiple component analysis (MCA, Zhang et al. 2007)."

P23237, L12: As a climatological AVERAGE? Climatology by itself is the study of climate.

Response: The term "climatology" refers to normal values or multi-year average in the context of climate science, so we do not change this word usage. In chemistry-climate modeling, one commonly refers to "ozone climatology" and "aerosol climatology".

P23238, L25: should probably cite Donahue 2006 also since the C* formulation is being used rather than the Pankow one.

Response: Reference added.

P23238, L26: POA-to-POC of 2.1? Primary OA has a much lower ratio (near 1 for HOA and 1.5-1.7 for BBOA, see Aiken et al. 2008 EST and the many papers that followed). You mean overall OM/OC OA, mostly dominated by OOA (SOA)?

Response: We originally chose this factor of 2.1 based on Turpin and Lim (2001), which suggests a factor of 2.1 ± 0.2 for nonurban organic aerosols. Now we have changed the POA-to-POC ratio to a traditionally used value of 1.4, because as the reviewer suggested, POA has a lower OA-to-OC ratio than nonurban aerosols (dominated by SOA).

At P23238 L26, we delete "and assumes a POA-to-OC ratio of 2.1…", and add: "Later in Section 4.2, we assume a POA-to-POC ratio of 1.4 (Aiken et al., 2008; White and Roberts, 1977) to calculate modeling POA and OA to compare with observations."

At P23246 L3, we delete "The model underprediction of total OA is not as large as in Heald et al. (2011) probably due to the high POA-to-POC ratio of 2.1…"

Fig. 4, 5, 6 and 7 have been updated using the POA-to-POC ratio of 1.4, as well as corresponding statistics reported in the main text.

P23239, L20: Please clarify in the text why a high-NO_x isoprene parameterization is not included. This seems like it could be a major omission, especially considering that this study primarily is an investigation of the NO_x dependence of global SOA.

Response: At P23239 L21, after "SOA formed from OH-initiated photooxidation of isoprene still only has one set of yields following the low-NO_x parameterizations", we add "We do not change this isoprene-SOA parameterization to remain consistent with the VBS framework from Pye et al. (2010). Additional simulations adding the high-NO_x pathway of isoprene are discussed in the Supplement."

P23240, L17-19: Why is oxygen not added when aging? Presumably the underlying mechanism for lowering the volatility is primarily by the addition of oxygenated functional groups. This would seem to underestimate the mass and the O/C of the SOA formed.

Response: We do not account for the added oxygen when aging due to the large uncertainties in the aging processes and the complexity of various SOA species. The change of O/C ratio is not the focus of this study because we do not have sufficient observational evidence to validate the simulated O/C ratio in the global model. We choose a high aging parameter $k_{OH} = 4\times10^{-11}$ cm^3 $molec^{-1}$ s^{-1} to provide an upper limit of SOA estimation, which would partly offset the omission of added oxygen mass during aging.

At P23240 L19, we add: "Considering the complexity of various SOA species and large uncertainties in aging process, the assumption of fixed O:C ratio for each SOA product surrogate is acceptable for global model parameterizations."

P23246, L24-25: Is surface area really the more important affect or rather OA mass? Obviously higher surface area can help outcompete deposition to the surface of the earth which is part of the model. However, OA mass will promote condensation of higher volatility species. Please clarify. If not obvious from the data, it seems like something that could be probed with the model.

Response: The major influence is more OA mass to promote condensation of semi-volatile species.

At P23246 L24-25, we delete "… which will increase SOA production due to the larger aerosol surface area available for condensation", and add "which will promote condensation of semivolatiles onto pre-existing organic matter thus forming more SOA".

P23249, L22-23: If a high-NO isoprene chemistry were included in the model and produced less SOA there couldn't there be a compensating effect of increasing SOA by shift from the NO to the HO_2 pathway. Can the authors discuss this possibility? The isoprene low-NO pathway is clearly an important contribution to the total SOA production in all regions and models.

Response: We performed additional simulations and discuss the effect of including the high-NO isoprene chemistry in the Supplement. Please see the response earlier in this document, which include: "… For isoprene oxidation in the southeast US, the HO_2

pathway increases and NO pathway decrease. While the effect of shifting to high-yield HO_2 pathway is masked due to the reduced OH concentration (about -15%, Fig. S2)…".

P23250, L2: make "consider" plural

Response: corrected.

P23250, L5: change "interference" to "influence"?

Response: corrected.

P23250, Section 4.3.3: Can the authors comment on why NO_3+monoterpene SOA decreases so little with the 50% reduction?

Response: This is because the reduction in NO_3 is not as large as 50% due to the NO_x gas-phase chemistry. In the southeast US, the 50% reduction in anthropogenic NO emission is equivalent to 45% in total NO emission, and leads to a 49% decrease in atmospheric NO_x (=$NO+NO_2$) and only a 24% decrease in NO_3 concentration.

At P23250 L7, we add: "This reduction in NO_3-branch compared to its normal value is relatively small because the decrease in NO_3 concentration is only 24%."

Figure 1: Some white contour line labels are missing.

Response: corrected.

Reference:

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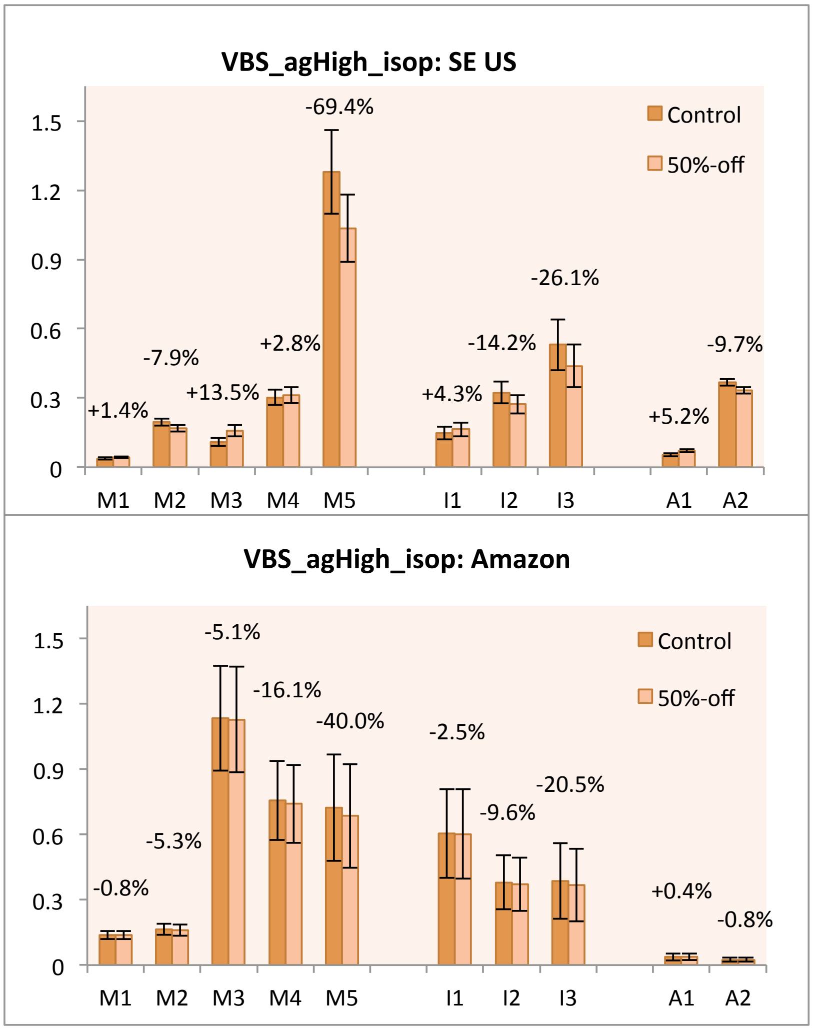

M1: MTP+OH(HO_2)
M2: MTP+OH(NO)
M3: MTP+O_3(HO_2)
M4: MTP+O_3(NO)
M5: MTP+(NO_3)

I1: ISOP+OH(HO_2)
I2: ISOP+OH(NO)
I3: ISOP+NO_3

A1: AVOC+OH(HO_2)
A2: AVOC+OH(NO)

Figure S5. Annual mean surface SOA concentration (µg m^{-3}) in the control run and the sensitivity run (with 50% anthropogenic NO emission off) from different pathways using the VBS_agHigh_isop scheme, averaged over the southeast U.S. [32°-40°N, 95°-77°W] and the Amazon [17°S-5°N, 77°-55°W]. The numbers above each bar denote the relative contributions (%) of each SOA formation pathway to the total SOA concentration change. The sum of all numbers equals -100% due to the overall decrease in total SOA.