

Interactive comment on “Understanding sources of organic aerosol during CalNex-2010 using the CMAQ-VBS” by M. C. Woody et al.

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We appreciate the reviewer's time and effort to provide thoughtful and thorough comments. We have incorporated the reviewer's comment into the manuscript in an effort to clarify the text. Regarding the reviewer's comment of VBS SOA performance compared to other studies, the reasons for varying performance are numerous (e.g. differences in models, scales, VBS implementations, emission inputs, volatility split of emissions, chemical mechanisms, etc.). Specific to the studies that provide metrics for SOA performance (Hodzic et al., 2012; Shrivasta et al, 2011; and Fountoukis et al., 2015), two (Hodzic et al., 2012; Shrivasta et al, 2011) used considerably higher SVOC and IVOC emissions (7.5x POA inventory) to achieve good SOA performance compared to measurements.

Below are responses to the reviewer's specific comments, with the reviewer's original comment shown followed by our response.

Abstract 1. Please explain briefly the general characteristics of CMAQ-VBS against the CMAQAE6 simulation runs so as the reader can understand why there is an underestimation. In line 9 and line 12 the authors report this in parenthesis but it is insufficient. Please delete the parenthesis and explain a little bit more in a separate sentence.

We provided a greater level of detail of how the OA treatment in CMAQ-AE6 differs from CMAQ-VBS. The text now reads: "Traditionally, CMAQ treats primary organic aerosol (POA) as non-volatile and uses a 2-product approach to represent secondary organic aerosol (SOA) formation. CMAQ-VBS instead treats POA as semivolatile and lumps OA using volatility bins spaced an order of magnitude apart."

2. Line 10. Please replace NMdnB with NMB throughout the whole manuscript

NMdnB is commonly used to abbreviate normalized median bias to avoid potential confusion with normalized mean bias (NMB) (e.g. Appel et al., 2008; Foley et al., 2010; Bash et al., 2014). We have not made the reviewer's recommended change to also avoid confusion and to continue the precedence established by previous work.

3. Line 23. Please explain what the term "intrinsic SOA" means before starting using it.

The use of "intrinsic", or inherent, is used to describe the model's SOA formation efficiency ("intrinsic SOA formation efficiency"). We did not intend for it to be interpreted as a new term ("intrinsic SOA").

4. Line 20-24. This sentence is too big and too problematic. "based on species ratios", which species and what ratios? "SOA parameterization from the observation", you mean SOA parameterization based on observations? In that case what kind of observations? The parenthesis used here are also confusing.

In an effort to provide clarification and answer questions raised by the reviewer, we

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have revised this sentence to read: “We use two new methods, one based on species ratios (SOA/ Δ CO and SOA/Ox) and another on a simplified SOA parameterization based on AMS observations, to apportion the SOA underprediction for CMAQ-VBS to too slow photochemical oxidation (estimated as 1.5× lower than observed at Pasadena using $-\log(\text{NO}_x : \text{NO}_y)$), low intrinsic SOA formation efficiency (low by 1.6 to 2× for Pasadena), and too low emissions or excessive dispersion for the Pasadena site (estimated to be 1.6 to 2.3× too low/excessive).”

5. Line 27. “too low by about 7x”. In comparison to what?

The 7x refers to comparisons against observations. We have revised the text to read: “too low by about 7x compared to observations.”

6. Page 26747, line 3-8. Too big sentence. Please make shorter. Replace “compared to” with “followed by”

We have broken the sentence into two to make it shorter as requested by the reviewer. However, we did not replace “compared to” with “followed by” since we are not intending to rank the sources (note: “other” sources, which comprise 13%, is listed last). The revised text now reads: “From source-apportioned model results, we found most of the CMAQ-VBS modeled POA at the Pasadena CalNex site was attributable to meat cooking emissions (48%, and consistent with a substantial fraction of cooking OA in the observations). This is compared to 18% from gasoline vehicle emissions, 13% from biomass burning (in the form of residential wood combustion), and 8% from diesel vehicle emissions. All “other” inventoried emission sources (e.g. industrial/point sources) comprised the final 13%.”

7. Page 26747. How you estimated the 1.7 factor? This is based on SVOCs measurements?

The 1.7 factor was estimated by comparing AMS measured to modeled POA concentrations. We have updated the text to include the factor was estimated using AMS

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measurements. "Using AMS measurements, we estimated. . ."

Methodology The structure of this section is inadequate. In several paragraphs the authors repeat same or similar information. The flow of the text would be greatly improved if the authors separate this section in subsections (i.e. 2.1 Model Description and Application, 2.2. Meteorology, 2.3 Emission Inventory, 2.4 Sensitivity Simulations, 2.5 Measurements etc.). In addition this will help the reader to understand the main features of the model which unfortunately it is very difficult to be identified with the current format of this section.

As recommended, we have reorganized this section by adding the following subsections: Model Application; CMAQ-VBS OA Treatment; Emissions; Sensitivity Simulations; Meteorology, Boundary and Initial Conditions; and Measurements.

8. Page 26750, line 8. Please report the actual value of the C^* for the non-volatility bin used in the model. If it is set to 0 then you can mention that under typical atmospheric conditions at Pasadena this bin can represent all the compounds with $C^* \leq 10^{-1} \mu\text{g m}^{-3}$.

For the nonvolatile bin, $C^* = 0$. We have updated the text to include this and that this would represent compounds with a $C^* \leq 10^{-1} \mu\text{g m}^{-3}$ at typical ambient conditions in Pasadena.

9. Page 26750, line 10. Please rephrase. The sentence is too big and too confusing. From my understanding the authors assumed that IVOCs are represented as a naphthalene-like surrogate specie and therefore they used the aerosol yield of ARO2 (which includes naphthalene).

To clarify, we have rephrased this sentence to the following two sentences: SOA yields from IVOC precursors are based on the Murphy and Pandis (2009) yields for the SAPRC ARO2 model species (Koo et al., 2014). ARO2 was used because it represents naphthalene (among other compounds), where naphthalene has previously been

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used as a surrogate to represent IVOCs (Pye and Seinfeld, 2010).

10. Page 26752, line 8. What is the 10% here? (a) The 10% of the oxidation product is allowed to move to SOA and the rest remains as POA? Or (b) all the oxidation product is moved to SOA which is calculated to be approximately 10% of the reacted POA? If the (a) is correct then explain more the rationale behind this assumption. If (b) is correct please report this in your results and not here.

The 10% is meant to represent (a). To clarify, we have revised the text as follows: "...and a portion (~10%) of the OA mass shifted from the primary SVOC (POA) to the secondary SVOC (SOA) set (Koo et al., 2014). The transfer of oxidized primary SVOCs (i.e. POA) to secondary SVOCs (i.e. SOA) is used as a modeling technique to maintain accurate O:C ratios, a feature of the 1.5-D VBS used in CMAQ (Koo et al., 2014), using existing POA and SOA basis sets and avoid additional computational burden of added model species (e.g. oxidized POA basis set)."

11. Page 26752, line 12-14. So a compound after 4 oxidation steps is still considered as POA? This is not correct. It should be compared against OOA and not HOA.

We agree that oxidized POA should be compared against OOA rather than POA but the modeling framework does not allow such a comparison. We found that oxidized POA represented < 10% of OA mass and therefore has only a small impact on our results. We have also added the following text to acknowledge the added uncertainty in using this approach: "With this treatment, the majority (~90%) of slightly aged POA (after a single aging reaction) resides as POA-like while very aged POA (after four aging reactions) would reside as two-thirds POA and one-third SOA. We acknowledge this approach, which prioritizes O:C ratios, adds uncertainty when model results are compared against AMS measurements and is an area where future research is needed to better understand that uncertainty. At Pasadena, our model predictions indicated ~8% of modeled OA was comprised of oxidized POA and suggests this approach has only a small impact in this application."

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12. Page 26752, line 11-14. How much of the total OA is coming from the boundaries in your domain? You have to make a simulation without emissions to verify the assumption that is reported here.

Based on a CMAQ-AE6 simulation, approximately 1% of total OA at Pasadena is attributable to boundary conditions. We have updated the text to the following: “Neither larger scale simulation included CMAQ-VBS OA species, though the impact is likely small as a CMAQ-AE6 sensitivity simulation indicated most (99%) of the OA at Pasadena originates from local or regional sources located in our modeling domain.”

13. Page 26752, the paragraph that begins in line 15. Here is an example of how confusing is this section. 4 paragraphs before this, the authors give some information of how SVOCs are treated in their model, then there is a paragraph with the model domain (which actually will fit better in the first paragraph of this section), then a paragraph with emissions used, then a paragraph with the meteorology, and then here they have an additional paragraph which again has information on emissions and how SVOCs are treated by their model. Please follow a more detailed structure and try not to repeat similar information.

As recommended, we have reorganized the entire methodology section.

14. Page 26752, line 26-28. Does the underlying chemical mechanism include species that are considered SVOCs or IVOCs ($C^* \leq 106$)? If yes please give a couple of examples. If it includes only VOCs then there is no double counting.

The technique to recycle OH in reactions with SOA precursors is commonly used in CMAQ (e.g. benzene and toluene) as these species are not explicitly represented in the gas-phase mechanism. Instead, generic model species (e.g. paraffins, olefins, nonreactive, etc.) are used to represent the gas-phase ozone chemistry and explicit species are used to represent the SOA chemistry. To clarify, we have revised the text as follows: “OH is artificially recycled (i.e. not depleted) in oxidation reactions of IVOCs and SVOCs (primary and secondary) to prevent double counting and impacts

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to the gas-phase chemistry of the underlying chemical mechanism as these species are likely already represented in the model (e.g. paraffins, olefins, nonreactive, etc.). This technique is identical to that used by a number of existing CMAQ SOA precursors (e.g. benzene and sesquiterpene).”

Results and Discussion 15. Page 26755, line 2. Delete “modeled OA”.

We have made the recommended deletion.

16. Page 26755, line 15. Replace “or improve” with “resulting in degraded”

We have made the recommended deletion.

17. Page 26756, line 6-8. If CMAQ-POA includes OA from biomass burning emissions you have to compare the CMAQ-POA against the sum of AMS HOA and BBOA. Furthermore, you have to add LOA in the comparison. Is there any indication (or interpretation) on what is LOA (oxidized or not)? If it is considered aged material it has to be added to the comparison against CMAQ-SOA while if it is considered fresh it has to be compared against POA. Overall, you cannot compare all CMAQ-OA components against part of the observed OA except if you assume that you are missing a specific OA source that is attributed to a specific AMS OA type.

The AMS measurements did not detect BBOA at Pasadena and therefore we chose to keep the CMAQ-VBS BBOA from residential wood combustion emissions separate from CMAQ-VBS POA. The AMS measured LOA was correlated with PM_{1.15} Ti suggesting it may be related to paint or surface coatings (Hayes et al., 2013), a source which does not emit POA in CMAQ. Both types of OA do not contribute significantly to total OA (< 10%) and including/excluding them would have little to no impact on our results or conclusions.

18. Page 26755, line 9. The authors switch randomly from OA to OC concentration or SOA to SOC through the whole text which I found it rather confusing and hard to follow. I strongly suggest the use of “OA” instead of “OC” and “SOA” instead of “SOC”

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throughout the text. Either way, the authors should report the factor used to convert modeled OA to OC (or measured OC to OA).

Our use of OA vs. OC is defined by the measurement type. Comparisons with filter-based measurements (CSN, IMPROVE, and CalNex) use OC. Comparisons with AMS data use OA. In instances where both measurement types are used (e.g. Fig. 2a), OC is used. We have updated the text to indicate how CMAQ-VBS OA and AMS OA was converted to OC: “When compared against the filter-based and routine monitoring network (CSN and IMPROVE) measurements of OC, CMAQ-VBS OA is converted to OC using OA/OC ratios reported in Koo et al. (2014). . . When comparisons using both AMS and the filter-based OC measurements are made, AMS OA is converted to OC using OA/OC ratios reported in Hayes et al. (2013).”

19. Page 26756, line 24-27. Can you make a small comment on how this scenario affected CMAQ performance regarding SOA?

This estimate was made as a post-processing step outside of the model and therefore we unfortunately do not have corresponding SOA performance.

20. Page 26758, line 22-25. This sentence is confusing. Please rephrase.

We have rephrased the sentence to read: “We calculated that when modeled OA concentrations were increased to match measured OA, partitioning of SVOCs increased POA-o concentrations by 20%. The 20% increase in modeled POA-o corresponded to a 2.3x underprediction of afternoon POA-o, with little change to morning and evening performance.”

21. Page 26758, line 26. Please delete the whole sentence in the parenthesis

We have made the recommended deletion.

22. Page 26759, line 26. Add “that” after “suggest”

We have made the recommended insertion.

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23. Page 26759, line 27. Add “therefore” after “2x and”

We have made the recommended insertion.

24. Page 26760, line 2. Add “which” after parenthesis

We have made the recommended insertion.

25. Page 26760, line 3. Add “that” after “assume”

We have made the recommended insertion.

26. Page 26760, line 4. Add “the” before “missing”

We omitted using the article “the” prior to “missing emissions” to imply some ambiguity in the term and would therefore prefer to continue to omit the article.

27. Page 26760, line 4. Has less impact on what?

We have revised the text to indicate the impact is on POA predictions: “However, we assume that this has less impact on POA predictions. . .”

28. Page 26760, line 5. Replace “or 25% excluding CIOA” with “(or 25% of POA excluding CIOA)”

We have made the recommended insertion.

29. Page 26761, line 16-19. Please rephrase. It is not clear how the numbers reported in this sentence suggest a factor of 2 error in the SOA precursor to CO emission ratio used by CMAQ

In an effort to clarify, we have added the following text” “The relative good agreement of SOA precursor concentrations, along with the factor of two underprediction of CO, suggests the SOA precursor to CO emission ratio was incorrect by a factor of two.”

30. Page 26763, line 1. Add “that can” after “indicated”

We have made the recommended insertion.

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31. Page 26763, line 13-20. Very confusing sentence. I can't see any connection between the work of Jathar et al. (2014) and the sensitivities reported in this sentence. Therefore no conclusions can be made about the effect of Jathar et al. (2014) suggestions on CMAQ performance. Furthermore, the sensitivities reported here have not been presented earlier in the text. The authors should add a paragraph with all the sensitivities in the methodology section with their explanation and the rationale behind them. For instance, there is no point to use 7.5x POA emissions in this application. This has been chosen in Mexico City where the emissions were based on ambient measurements (which is not the case here).

We have included the SVOC sensitivity simulations performed in the methodology section and rational for each. For example, the 7.5x POA, which we note is based on Robinson et al. (2007) when a POA emission inventory estimate is made before partitioning (and not specific to Mexico City), represents an upper bound of values used in the literature and simulations with it and a lower bound (1x POA emissions) provides constraints on CMAQ predicted SOA from IVOC emissions. We have also revised this paragraph to better connect the work by Jathar et al. (2014) with our sensitivity simulations. The revised text now reads: "Current CMAQ-VBS IVOC emissions are scaled to primary SVOC emissions (1.5x) based on the results of a diesel generator (Robinson et al., 2007) and could potentially be updated to utilize more recent results, such as those reported by Jathar et al., 2014 who indicated unspciated organics (S/IVOCs) dominated SOA mass formed from combustion emissions. Future work is needed to explore if better constraining IVOC emissions and yields in CMAQ would help improve model performance, but it would likely not account for the entire missing SOA mass based on sensitivity simulations using upper bound S/IVOC emissions. In these simulations, S/IVOC emissions were increased. . ."

32. Page 26766, line 7-8. Replace "the break down of POA was as follows:" with "POA comprised of"

We have made the recommended change.

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33. Page 26766, line 24-25. Another example of bad writing. What kind of biogenics? More SOA at Pasadena than where? Or When? You can rewrite the sentence as: “The aging of biogenic SOA produced approximately $0.5 \mu\text{g m}^{-3}$ on average additional SOA at Pasadena throughout the day.”

We have revised this sentence to read: “In the simulation that aged secondary biogenic SVOCs, SOA concentrations were $\sim 0.5 \mu\text{g m}^{-3}$ higher throughout the day at Pasadena compared to simulations that did not age secondary biogenic SVOCs. . .”

34. Page 26767, line 11-12. Again, what kind of biogenic? Replace the sentence with “On the other hand, biogenic VOCs emitted in the Central Valley and surrounding mountains are thought to be the major source of biogenic SOA observed in the LA basin”

We have made the recommended change.

35. Page 26767, line 13-14. “underprediction of monoterpenes”: Do you mean under estimation of monoterpene emissions?

Yes, we were referring to monoterpene emissions and have added the word “emissions” for clarification. That portion of the sentence now reads: “...if the underprediction of monoterpene emissions applies to other areas. . .”

36. Page 26767, line 17. Replace “contribution for” with “contribution of”. Also, contribution to what? To predicted SOA concentrations?

We have made the recommended change and added “to predicted SOA concentrations” for clarification. The sentence now reads: “Figure 8 also provides the contribution of the three standard SOA formation pathways in CMAQ-VBS (VOCs, IVOCs, and aging) to predicted SOA concentrations at Pasadena.”

37. Page 26768, line 4-5. Delete “has been presented (Hodzic and Jimenez, 2011; Hayes et al., 2015) and” and “here”

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We have made the recommended change.

38. Page 26768, line 11. Delete “when”

We have made the recommended change.

39. Page 26768, line 15. Delete “and”

We have made the recommended change.

40. Page 26768, line 16-18. Delete the whole sentence. Unnecessary information for the purpose of this study.

We have made the recommended change.

41. Page 26768, line 19-23. There is no need for this to be in a separate paragraph. Also, you can reverse the order of these two sentences as follows: “In our implementation in CMAQ-VBS we use an emission rate of 0.069 g VOC* g⁻¹ CO and a KOH=1.25x 10⁻¹¹ cm³ molec⁻¹ cm⁻¹. Hayes et al. (2015) found that using these optimal values, the SIMPLE parameterization...”

We have made the recommended change, combining paragraphs and changing the order of the sentences.

42. Page 26768, line 25. Replace “with the right diurnal cycle” with “following similar diurnal cycle” and place “(Fig. 9)” at the end of the sentence.

We have made the recommended change.

Conclusions 43. Page 26770, line 12. Add “from” after “50%” and “emissions” after “non-fossil”

We have made the recommended change.

44. Page 26770, line 15. Add “that” after “estimated” and add “the observed” after the “50% of”

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We have made the recommended change.

45. Page 26770, line 16. Add “of SVOCs” after “66%”

We have made the recommended change.

46. Page 26770, line 20-27. I do not agree that is a matter of modelling needs and goals. During the last decade, the traditional treatment of OA has been proved to predict accurately the total OA in some cases (i.e., urban centers) but for the wrong reasons (overpredicts POA and underpredicts SOA). Therefore, the traditional treatment should not be considered appropriate anymore. The only advantage of the traditional approach was its computational efficiency compared to VBS but this doesn't seem to be an issue anymore for VBS approach since during the last 5 years it has been applied even in global models with great success (Pye and Seinfeld, 2010; Farina et al., 2010; Jathar et al., 2011; Jo et al., 2013; Tsimpidi et al., 2014). Please re-write the whole paragraph in order to highlight why the user has to switch to the CMAQ-VBS version by mentioning the great advantages of VBS (more accurate prediction of POA/SOA split without a significant computational cost).

We agree that the VBS approach in treating POA as semi-volatile is more appropriate than treating POA as non-volatile in the current AE6 formulation. However, the AE6 approach that explicitly relates parent VOC with SOA provides source specificity and chemical process information that is often useful for scientific and regulatory applications which is not available in the traditional VBS approach. Given that AE6 could be updated to treat POA and semi-volatile similarly to VBS and incorporate IVOC emissions for SOA formation the future may lead to a hybrid type of approach meaning an endorsement of the traditional VBS or AE6 would not be appropriate for this manuscript. Additionally, with respect to the VBS approach, there are number of uncertainties within VBS give us pause, including how best to apply existing POA emission inventories in a VBS framework or a number of empirical OA representations (aging only for anthropogenic SVOCs, lack of fragmentation, etc.). Given these

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uncertainties, it is reasonable to ask if VBS performs better for “the wrong reasons” by overemphasizing contributions from aging and/or IVOCS, excluding fragmentation, etc. The intent of this manuscript is to utilize routine and special study measurements to evaluate multiple approaches for estimating OA in a photochemical grid model and articulate the where improvements may be needed. We appreciate the reviewer’s point of view here but we do not feel a technical manuscript is a venue for an endorsement of a particular approach. We have revised this paragraph to read: “Regarding which OA treatment is more appropriate, CMAQ-VBS or CMAQ-AE6, depends on the user’s modeling needs and goals. The traditional CMAQ-AE6 treatment, while it has known limitations (generally overpredicting POA and underpredicting SOA), more accurately predicts total OA measured at routine monitoring networks. Conversely, CMAQ-VBS treats primarily emitted OA as semivolatile and easily incorporates an estimate of IVOC emissions missing from the inventory to provide improved predictions on the total SOA mass and the POA/SOA split at Pasadena. The AE6 approach provides some utility in that parent VOCs and reaction processes are more clearly linked to SOA which is sometimes useful for scientific and regulatory model applications. Due to the difference in SOA/POA splits, the two CMAQ configurations may respond differently to VOC and/or NO_x emission reductions, which should be examined in future work. Another area for future work is updating the POA emission inventory, originally developed for a non-volatile POA treatment, to account for semi-volatile POA and likely improving CMAQ-VBS total OA predictions.”

Tables-Figures 47. Page 26781, line 10. Similar comment to the one above: the nonvolatile bin should represent all the compounds with $C^* \leq 10^{-1}$ in Pasadena

Similar to the previous comment, we have indicated on the table that for the non-volatile bin $C^* = 0$, which at typical ambient conditions at Pasadena represents compounds with $C^* \leq 10^{-1}$.

48. Page 26786, Figure 2a. Did you convert AMS-OA to AMS-OC? If yes, which factor did you use? Furthermore, why there is no data for AMS during the period of 18/5-30/5

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in figure 2a? According to Figure 2b it seems that there is available AMS data during this period.

AMS-OA was converted to OC using AMS-measured OM to OC ratios (Hayes et al., 2013). Only days with at least 16 hours of AMS data are included in Figure 2a, as indicated in the caption. We have updated the caption to indicate how AMS-OA was converted to AMS-OC (and per an earlier comment included this in the text) and the days < 16 hours of AMS measurements were available (18/5, 20/5-26/5, 28/5, and 29/5). "AMS measurements in (a) were converted to OC using OM to OC ratios reported in Hayes et al. (2013) and include only days with > 16 hourly measurements (i.e. 18/5, 20/5-26/5, 28/5, and 29/5 are excluded due to missing measurements).

References Appel, K. W., Bhave, P. V., Gilliland, A. B., Sarwar, G., and Roselle, S. J., 2008. Evaluation of the community multiscale air quality (CMAQ) model version 4.5: sensitivities impacting model performance; part II—particulate matter. *Atmospheric Environment*, 42(24), 6057-6066. Bash, J. O., Carlton, A. G., Hutzell, W. T., & Bullock Jr, O. R. (2013). Regional Air Quality Model Application of the Aqueous-Phase Photo Reduction of Atmospheric Oxidized Mercury by Dicarboxylic Acids. *Atmosphere*, 5(1), 1-15. Foley, K. M., Roselle, S. J., Appel, K. W., Bhave, P. V., Pleim, J. E., Otte, T. L., Mathur, R., Sarwar, G., Young, J. O., Gilliam, R. C., Nolte, C. G., Kelly, J. T., Gilliland, A. B., and Bash, J. O. (2010). Incremental testing of the Community Multiscale Air Quality (CMAQ) modeling system version 4.7. *Geoscientific Model Development*, 3(1), 205-226.

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