Reply to reviewer's comments:

Reviewer 1 comments:

Reply to the reviewer in red.

1) The large amount of CTM simulations are very helpful to explore the influence of the perturbed parameters on the modelled OA fraction, and I congratulate the authors for such analysis. However, I believe that it is important to mention (i.e. in the introduction, methodology or conclusion sections) that the perturbed space explored here is embedded in the parent VBS scheme that has been adopted. Therefore, the results should be interpreted in the light of the original VBS blueprint used by the author (e.g. 1D VBS, among other details). For instance: what will happen to the whole perturbed parameter space, if the original VBS will account for processes such as Fragmentation/Formation of HOMs/Autoxidation or for different kernel distributions of the oxidation products (e.g. different number of oxygen additions during the first oxidation steps, and therefore different allocation of the oxidation products across the volatility bins)?

I am not asking for more runs, but I believe this is an important point that needs to be considered.

Indeed this is an important comment and it has been added in the methodology as follows: It is worth mentioning that the perturbed space explored here is embedded in the parent VBS scheme that has been adopted. There have been a large number of developments in, and variants of, the VBS aiming to address particular questions related to SOA formation at various levels of complexity (for example, the mechanistic measurement-constrained radical 2D-VBS examining the role of ELVOC and ULVOC in new particle formation; (Zhao et al., 2020; 2021). In the current study, our implementation has been developed from the VBS version available in the distribution version of WRF-Chem and our results should be interpreted in the context of the structural capabilities and limitations therein. More information about the VBS distributions and parameter space setup is in section S1 in the supplementary material.

Zhao, B., Shrivastava, M., Donahue, N. M., Gordon, H., Schervish, M., Shilling, J. E., Zaveri, R. A., Wang, J., Andreae, M. O., Zhao, C., Gaudet, B., Liu, Y., Fan, J., and Fast, J. D.: High concentration of ultrafine particles in the Amazon free troposphere produced by organic new particle formation, Proc Natl Acad Sci U S A, 117, 25344-25351, 10.1073/pnas.2006716117, 2020.
Zhao, B., Fast, J. D., Donahue, N. M., Shrivastava, M., Schervish, M., Shilling, J. E., Gordon, H., Wang, J., Gao, Y., Zaveri, R. A., Liu, Y., and Gaudet, B.: Impact of Urban Pollution on Organic-Mediated New-Particle Formation and Particle Number Concentration in the Amazon Rainforest, Environ Sci Technol, 55, 4357-4367, 10.1021/acs.est.0c07465, 2021.

2) The analysis of the diurnals is suggesting a strong influence of the meteorological parameters on the OA concentrations, likely of the boundary layer height diagnosed by the model. Even for the model runs with the best overall statistics and optimized scaling factors, there is a positive bias during the early morning hours. This is an important result. Uncertainties in the diurnal emissions factors are sound, but I think the work will benefit from more analysis on the meteorological side. Are there any radiosondes measurements available to better understand/evaluate the model vertical output (see my additional comments below for the Results section)? In addition, how are the synoptic conditions of the pre-monsoon season reproduced by the model?

We agree with the reviewer. There are no radiosondes measurements available to extend the analysis at the location and time period of our study. We have, in response to a question from reviewer #2, included comparisons of wind direction, speed, temperature and relative humidity between the model

and measurements for the study site. Hopefully these will help readers to evaluate the local model performance, even if we cannot provide information on the larger scale performance of the model.

# Minor comments:

# Abstract

Abstract, line 28: Suggestion: I would replace "POA production and aging" with: "emissions of POA and its chemical evolution". The authors replaced the phrase as suggested.

Abstract, line 29: Suggestion: "The main disadvantage is its complexity". I would not consider the complexity of the VBS a disadvantage per se, and I would suggest removing that sentence (I think it is already clear stated that the high numbers of parameters are difficult to probe). The line has been edited as follows: However, the evaluation of model uncertainty and the optimal model parameterisation maybe expensive to probe using only WRF-Chem simulations.

Abstract, line 40: Suggestion: "in two periods: the full period (1st -29th May) and the period 14:00-16:00 hrs local time, 1st-29th May". One period and one sub-period? Yes, that is correct. The line has been edited: The full period and one sub-period.

# Introduction

Introduction, Line 50: Suggestion: I would move the sentence beginning at this line in the following paragraph. It is a bit detached from its current paragraph. The change has been done.

Introduction, Line 58: Please consider including Ghosh et al., 2021 who has recently applied the online version of the WRF-CHIMERE model over the same domain, i.e. the Indo-Gangetic Plain. The reference has been added

Introduction, Line 64: Please consider replacing "POA processes" with "OA processes". The change has been done.

Introduction, Line 70: Suggest using a more updated and theoretically oriented reference for the definition of the VBS domain, e.g. Bianchi et al., 2019. The reference has been added

Introduction, Line 73: Please consider rephrasing "The resulting low volatility oxidized organic vapors can condense to produce oxidized primary organic aerosols (oPOA) (Shrivastava et al., 2008)" with ". The resulting low volatility oxidized organic vapors can condense to produce secondary organic aerosol (SOA) (Shrivastava et al., 2008)". I think the model assigns those products to SOA (?). (oPOA is considered secondary, and there is no heterogeneous chemistry accounted for, or?). The change has been done.

Introduction, Line 92: "the partitioning of matter between gaseous and particulate phases, and the chemical aging of POA". Suggest rephrasing the sentence as: "The partitioning of organic matter and the evolution of POA". The change has been done.

# Methodology

Methodology: How long was the spin-up period? In addition, was WRF nudged towards some reanalysis datasets (ERA5 or GFS)? A spin-up period of 11 days was used, from 19<sup>th</sup> April to 1<sup>st</sup> May. The meteorological driving fields were taken from ERA5 reanalysis data. Spectral nudging of the uv wind parameters, temperature and geopotential height variables to these, above model level 18 and for wavelengths greater than 950km, was used. We have added this information to the paper.

Methodology: What is the vertical resolution of the model and the first model layer height?

The model domain had 38, variable height and terrain following, model levels, up to a pressure of 50 hPa. The first model layer has a mean height of 59m over Delhi (and a mean height of 56m over the whole model domain). This information has been added to the paper.

Methodology: Which scheme was used for the PBLH in the model? Please consider adding this information (see also my additional comment below). The YSU PBL scheme was used, we added a note on this in the paper.

Methodology, Line 121: "SOA components, each covering 4 volatility bins". At which C\* (and T)? Please add such information. Do they cover the SVOC range, or also the LVOC/ELVOC range? In addition, it would be nice the report the IVOC yields with their oxidation pathways in the Methodology section. The 4 SOA volatility bins have C\* values (at 298 K) of 1, 10, 100, and 1000  $\mu$ g/m<sup>3</sup>, only covering the SVOC range. The yields are given in Table 2 of Tsimpidi et al (2010); we have added the details to the paper and we have also added a table containing the relevant yields to our supplementary material too. The IVOC compounds are included in the standard VBS scheme.

Methodology, Line 130: "To investigate the impact of these assumptions on the model predictions, we have modified the model code so that the VBS emissions, the oxygenation rates and VBS reaction rates, can be directly controlled via namelist options". This is very nice. Thank you, we appreciate your comment.

Methodology, Line 134: "The volatility distribution of biomass burning". Suggest rephrasing with "The volatility distribution of open biomass burning" so to differentiate from the residential biomass burning sector contained in EDGAR-HTAP, which has different patterns and estimation methods (this is already specified in Table 1, and I suggest correcting it also in Table 2 accordingly). The change has been done.

Methodology, Line 137: "The volatility distribution for anthropogenic emissions is also multiplied by a scaling factor of 3". For anthropogenic emissions, I suggest adding also Denier van der Gon et al., 2015 which proposed a revisited emission inventory for the residential wood combustion sector based on a bottom-up approach. The revisited emission inventory, which try to account for the semi volatile components of the primary organic matter fraction, was a factor 2-3 higher compare to previously used emission inventories. The reference has been added to the manuscript.

Methodology, Line 140 "Anthropogenic emissions are derived from the EDGAR-HTAP". Is there any spatial interpolation performed by the model on top of the original EDGAR-HTAP emission datasets? Please add. What was the projection used in the model? The EDGAR-HTAP emission dataset is interpolated onto the model grid, using the anthro\_emiss tool provided by NCAR. No further spatial processing of the EDGAR-HTAP emission dataset was carried out. Within Delhi the SAFAR-India emission dataset is used. The model domain projection is Lambert Conformal.

Methodology, Line 163 ''PBLH data were sourced from ECMWF ERA5 with 0.25 deg". Why not directly from WRF at 15km? After further reading, I have realized that the aim is to compare WRF and ERA5. Are there any direct radiosondes measurements available? Unfortunately we do not have direct radiosonde measurements of the boundary layer height in this location.

Methodology, Figure 1: Is this an average over the whole period? Please add. I would actually consider adding the model topography instead of the PM1 concentrations. Figure 1 has been updated with the topography data.

Methodology, Line 202: I am not sure I have understood this passage: The IVOCs are included by multiply by a factor of 1.5 the non-volatile OA mass (0.2+0.5+0.8). Several models have implemented such scaling factor (Koo et al., 2014; Tsimpidi et al., 2010) on top of the already increased emission inventory (i.e. by a factor of 3 to include SVOC based on partitioning theory calculation), so that the additional organic mass added to the model runs is 7.5 (i.e. 3 + 3\*1.5). Is this

the case also for this application? In addition, are the IVOCs factors, when increased or decreased during the sensitivity tests, applied to all the EDGAR sectors (i.e. one factor for all the sectors)? Please specify. The IVOCs are calculated from the final emission dataset, which includes all EDGAR sectors. In our study the base scaling factor for IVOCs (0.2+0.5+0.8=1.5) that gives us our initial emission amount, is scaled by another factor, between 0-3, to probe the sensitivity of the model to the abundance of IVOCs.

These details have been added to the paper.

Methodology, Line 209: Are those factors, i.e. the parameter number 5 (i.e. 0.1 - 4), applied on top of the emissions inventory as described in the paragraph at line 137? I.e. a value of one would correspond to the original emissions inventory (i.e. increased by a factor of 3). This is correct.

Methodology, Line 230, Table2. What are the units of the anthropogenic VBS ageing rate and of the open biomass burning VBS ageing rate? Please add. The VBS ageing rates are given in cm<sup>3</sup> molec<sup>-1</sup> s<sup>-1</sup>. We have added these units to Table 2.

### **Results and discussions**

Results and discussions, Line 255: "the VBS setup will affect OA concentrations and PM, with no implications to inorganic aerosols or gaseous species". Would not the different oxidation schemes, and amount of organic precursors available such as SVOCs, IVOCs and VOC, alter the overall radical budget (e.g. HOx concentrations), and therefore also the inorganic and gas-phase chemistry? The gas-phase reactions which generate the SOA components, and age the VBS components, in this study do not consume OH, and so do not alter the overall radical budget. We chose to do this so that we could probe the model OA concentration and PM response only. It is worth mentioning that we did not want to introduce a coupling between the scheme and oxidants so as to introduce a second order feedback that we could not constrain.

We carried out some preliminary studies on the impact of including OH consumption (or not) on the atmospheric chemistry while setting up this study. We include below plots from these preliminary studies ('High' aging indicates a VBS reaction rate of  $1 \times 10^{-11}$  cm<sup>3</sup> molec<sup>-1</sup> s<sup>-1</sup>; 'low' aging indicates a VBS reaction rate of  $1 \times 10^{-12}$  cm<sup>3</sup> molec<sup>-1</sup> s<sup>-1</sup>), to give some guidance on how this impacts gas-phase chemistry. We found that including OH consumption could lead to 10-25% reduction in OH concentrations, depending on reaction rate. The impact on local ozone concentrations was not large (and not linear).



Results and discussions, Line 267: "Total\_OM". I guess it refers only to the modelled particle phase? Yes, this has been clarified in the manuscript.

Results and discussions, Line 289: "we identified two potential structural errors in the WRF-Chem outputs, the early morning peak and the late evening low concentrations". This is an interesting part of the model evaluation section:

- I would suggest plotting also the diurnal of CO, which is probably a better candidate then OA for identifying the performance of the PBLH in the model. I think a diurnal of CO is already included in Figure S5 but I am not sure I have understood it properly (I would suggest improving the readability of Figure S5, mainly the choice of the colors. I think Figure S5 is also not referenced in the manuscript). It seems model data are reported every three-hours (?) and have a more flat profile compared to the observations (?). How this relate to the diurnal of OA? Along this line, why do the diurnals of O:C decrease in the model during daytime considering that daytime O3 concentrations are over predicted ? Yes, the modelled CO concentrations are flatter than the CO observations. This is opposite to what is observed with OA, where, in general, OA observations are higher than the modelled OA. Figure S5 has now been referenced in the manuscript and a description has been added. We have also adjusted the colours in Figure S5 to make it more readable.

- For the analyzed period, it would be nice to know which one of the emissions sector in the EDGAR-HTAP dataset is the dominant one in that area, and what is the relative contribution of each of the sources.

- Is there any factor analysis on the AMS data to compare with? How are the modelled and the observational-based OA components (i.e. HOA and OOA) reproduced? Is it an overestimation related to the primary or to the secondary fraction? In addition, how will the model resolution (i.e. 15km) affect the transport and therefore the equilibration time between the particle and gas-phase organic material? Yes, there is a PMF analysis that we performed from the same dataset along with other datasets (Reyes-Villegas et al., 2021). In the following figure we highlight with a red square the PMF results that correspond to the data of the current study. We can see a morning peak around 9-10 hrs, which is higher in the secondary organic aerosol (more oxidised oxygenated OA – MO-OOA). This suggests that the large morning peak of the modelled organic mass that we observed in our diurnal analysis corresponds to the potential structural errors in the model, as mentioned in the manuscript.



Reyes-Villegas, E., Panda, U., Darbyshire, E., Cash, J. M., Joshi, R., Langford, B., Di Marco, C. F., Mullinger, N. J., Alam, M. S., Crilley, L. R., Rooney, D. J., Acton, W. J. F., Drysdale, W., Nemitz, E., Flynn, M., Voliotis, A., McFiggans, G., Coe, H., Lee, J., Hewitt, C. N., Heal, M. R., Gunthe, S. S.,

Mandal, T. K., Gurjar, B. R., Shivani, Gadi, R., Singh, S., Soni, V., and Allan, J. D.: PM1 composition and source apportionment at two sites in Delhi, India, across multiple seasons, Atmos. Chem. Phys., 21, 11655-11667, 10.5194/acp-21-11655-2021, 2021.

- I am not sure how much the comparison between ERA5-PBLH and WRF-PBLH could help here. Following one of my previous comment, I think it would be beneficial to clarify which parameterization has been used in WRF for the PBLH. ERA5 is based on the Richardson number method. How about those WRF runs? In addition, why the wind direction is not reported in Figure S4?

In WRF-Chem the PBLH is calculated using the critical bulk Richardson number. The wind direction analysis has been added to the supplement in figure S4. The model and observation wind directions (WD) are in reasonable agreement. The model and observation wind speed (WS) are of similar magnitude, though the model does show some systematic underprediction of WS, without the observed daytime increase in WS.



Figure S4. Wind roses and diurnal cycles of temperature, RH, ws and PBLH. May – 2018. Circles highlight the mean.

Model evaluation, line 340: "these model runs have low SVOC volatility distribution". Does this mean low factors (e.g. towards 0.05) and therefore a more volatile distribution (e.g. first panel in Figure S2)? Please add more details in the manuscript.

Yes – lower values on the SVOC volatility distribution scale does mean that the emitted VBS compounds will be more volatile. We have added clarification on this to the paper: "low SVOC volatility distribution (emitted VBS compounds are more volatile) and SVOC scaling". This has been added to the manuscript.

Model evaluation, Figure 5: This is a very nice plot, and clearly elucidates the various performance of the model runs. If possible, I would consider adding short acronyms for each of the 5 parameters at the pentagon's vertices (at least for one pentagon). It would highly facilitate the comprehension of the panel. In addition, even though provided on the github link, I would suggest to include a supplementary table were the 5 model's parameters are reported for the TRAIN 127, 121 126, 036, 117, 104, 115, 119 and 058. This will facilitate the use of such values by the modelling community. Finally, at least for TRAIN127 and TRAIN120, I would suggest producing in the manuscript a plot similar to Figure S2 where all the parameters are reported as insets on top of the volatility distribution plot associated to the final scaling factors. Again, this will highly facilitate the use of such parameters. The acronyms have been added to the figures for a better visualization. Table S2 in the supplement has been edited adding the 10 parameter values for the 111 model runs.

## Conclusions

Conclusion, line 493: "low SVOC scaling". A more volatile distribution? Yes, this has been edited in the conclusion.

Reviewer 2 comments:

# Reply to the reviewer in red.

#### Major comments

(1) 10 perturbed parameters: I don't think those are a scientifically meaningful set of parameters to improve OA modeling in Delhi. The authors only include semi-volatile POA only, especially for emission and formation processes, and SOA parameters are not considered in this study. I think biogenic SOA would be minor in May in Delhi, but at least aromatic SOA can not be ignored in the urban atmosphere. Furthermore, loss processes such as wet and dry depositions and photolytic losses (e.g., Zawadowicz et al. 2020) are not considered. If the photolytic loss was not included in the base model, I understand that adding a new reaction would be out of scope in this study. However, the authors could simply set up a set of parameters to change dry and wet deposition processes in the model, by changing the single parameter - effective Henry's law constant. As there are orders of magnitude variabilities in this constant and it is important for SVOCs (Hodzic et al. 2014), this parameter can give a sufficient range for use in this emulator approach.

We thank the reviewer for their comments and we agree that photolytic losses are not included and are out of scope. We recognise that our study relates to perturbation of a limited set of possible parameters but disagree with the contention that these are not scientifically meaningful. We now explicitly state that SOA parameters are not perturbed from the base case and are exploring the sensitivity to, and plausible perturbations of, the emissions and ageing of primary OA that would be necessary to explain the observations. We also appreciate that there will be sensitivity to the deposition rate of OA components. We have focused our study on the sensitivity of the OA production processes at a constant deposition rate within WRF-Chem allowing reasonable conclusions about the plausible range of the other parameters to be drawn notwithstanding this limitation. The treatment of dry deposition of gaseous species within WRF-Chem has no explicit dependence on solubility. Moreover, without consideration of the functionality of the partitioning components, there is no straightforward mapping of the solubility to the VBS descriptors without an implementation of a similar representation to Hodzic et al. (2014). As with photolytic losses, which are not considered in the base model, effective Henry's Law as a function of structure of condensing oxidation products is not considered in the base model and is considered out of scope of the current study. As such, we recognise that our study is only concerned with sensitivities to the production of OA from primary emissions. In the same way that Hodzic et al. (2014) did not consider perturbations to the factors relating to OA production (POA or SOA), nor to any POA processes, we now make it clear that we are not considering SOA processes, nor sensitivities to loss processes, though do attempt to examine the sensitivity to POA processes that would plausibly explain pre-monsoon Delhi OA variability.

(2) Aging with 7.5~45% mass added per reaction: I think those extra masses upon aging reaction can be exaggerated. The studies cited here had been published before the vapor wall loss was reported (Zhange et al. 2014). I guess those additional masses are introduced to compensate for the vapor wall loss (and missing sources like VCPs; McDonald et al. 2018), especially for SVOCs that are sticky and so wall losses are even higher than others. I agree that chemical aging happens in the atmosphere, but this approach only considers functionalization processes that always add mass and shift the volatility towards the lowest saturation vapor pressure bin, and neglects fragmentation processes. Field measurements also showed that there was no mass change with aging (Hodshire et al. 2019). This study is for the collection of field studies from biomass burning plumes, so may not be relevant to the anthropogenic OAs. There are some field studies from urban areas that showed a mass increase with photochemical age, but still I am not sure if the mass increase must be included especially in this study, because of the observed OA profiles shown in Figure 4. Looking at the observed diurnal cycles, O:C ratio increases during the daytime, meaning that there is aging in the atmosphere. However, OA mass decreases during the daytime. PBL evolution couldn't explain this, as observed PBL heights are similar between 12~16 pm. So I guess there were two compensating aging processes (functionalization and fragmentation) at this location. I would suggest that the authors should add the cases with no additional mass (Parameter  $3 \rightarrow 0$ ) or even no aging process (Parameter  $1 \rightarrow 0$ ) for meaningful suggestions to the OA modeling community.

We agree with the reviewer that showing the modelling community the lower limits for these processes will be useful. We believe, however, that we have already done that in this study. Regarding the adjustment of photochemical aging so that no extra oxygen is added to OM (which is the main point of including parameter 3), we do not feel that this would be informative – as it would simply give a flat O:C ratio which, as you note above, would not be realistic. As such, our lowest values for the parameter 3 are in scenario TRAIN048, which has a value of 0.08, and TRAIN048, which has a value of 0.086. For the aging process (parameter 1) our preliminary studies indicated that once the reaction rate is down to  $1 \times 10^{-13}$ , or less, the reaction rate is low enough that other processes dominate (especially with regards to local processes). Our lowest parameter 1 values are in scenario TRAIN014, at  $2.48 \times 10^{-13}$ , and TRAIN109, at  $2.98 \times 10^{-13}$ .

If we were to extend the range of our parameter space down to include zero for either of these processes, it would have skewed the parameter space such that we would have gained less information from the runs that were made – over coverage of the parameter space in regions which are more atmospherically relevant would have been sparser.

(3) Inclusion of 5 biomass burning parameters. I think those 5 parameters related to biomass burning are not necessary, compared to the loss processes and no aging cases suggested above. Those could be excluded for computational efficiency. This can be justified by comparing the magnitude of emissions in this area. For example, if biomass burning emission is 10 while anthropogenic emission is 100, no one expects that the variance of biomass burning scaling will be higher than that of anthropogenic scaling. Unless applying a lot higher maximum scaling factor to biomass burning than anthropogenic, the emulator should give the results that changing anthropogenic parameters is the effective way. When we started doing the simulations we did not know if the biomass burning parameters

would have a considerable contribution to OA concentrations. It was when we performed several runs when we noticed this behaviour. However, we retained the biomass burning parameter for consistency in the analysis. Our original intention was to continue the simulations for periods where there was a greater influence of biomass burning, but this was precluded by unavailability of measurement data for constraint.

(4) Figure 3: Looking at the top panel of Figure 3, the model runs with low O:C ratios show very high OM concentrations (yellowish). Since all model runs are considering aging reactions with added oxygen rates from 7.5 to 45%, I am not able to understand the results. Since the aging in this model adds oxygen mass only while the carbon mass is fixed, I think O:C ratio should be higher for the higher OM concentrations. Please explain why the model shows this tendency. We explored a range of emission multipliers (both IVOC and SVOC scaling), using upper limits for these which had been suggested as being reasonable by studies using this VBS scheme, but in other regions with other emission datasets. These upper limits turned out to be too high for our emission dataset, and these are the model runs which produced the very high OM mass loadings (rather than these being predominately caused by high oxidation rates). When the OM mass loading is high, more of the higher volatility (and, here, less aged) compounds condense into the condensed-phase. The VBS scheme we have used has only gas-phase reactions, and so once in the condensed-phase these compounds do not age further. This process leads to the lower mean O:C ratios that are observed here.

#### Minor comments

1) Line 50 - 51: The authors are talking about the air quality in India in this paragraph, then the last sentence is saying the large fraction of organic aerosols "globally". It would be better to cite other works that measured organic aerosols in India, or the authors can change the paragraph to present the global air pollution problem instead. But since the next paragraph is presenting OA modeling works in India, the former could be better. The word "globally" has been changed by "in urban environments".

# 2) Line 59: Spell out "VOC" The change has been made.

3) Line 63-64: Please provide the references to support this statement. I think there are also a lot of uncertainties about SOA mechanism as well, for example, large variabilities of enthalpy of vaporizations, pH-dependency (e.g. IEPOX), aging reaction parameters, photolytic loss, etc. The lines "mainly attributed to the lack of understanding of the emission sources and the POA processes." Has been changed by "mainly attributed to the lack of understanding of the emission sources, the POA processes and SOA mechanisms."

In this paper we have shown the capability of the method to constrain what we have studied. We cannot say whether unperturbed SOA parameters or unperturbed deposition rates or photolysis rates would give better agreement as this has not been the focus of the paper.

The focus of this paper is to perturb the anthropogenic and biomass burning POA, this has been highlighted in the methodology editing as follows: In this study, a WRF-Chem parameterisation is proposed to simulate organic mass concentrations and organic to carbon (O:C) ratios over the region of New Delhi, India, that includes primary and ageing parameters in the VBS scheme. In this parameterisation we focused on the perturbation to the chosen anthropogenic POA and biomass burning POA parameters that would be needed to give the best fit to the observed OA , without perturbing the SOA parameters from the base case. The model performance is evaluated over a multi-dimensional parameter uncertainty space that explores parameter uncertainty in these schemes.

4) Line 122-123: As far as I know, WACCM doesn't simulate ARO1 and ARO2, instead, benzene, toluene, and lumped xylenes. How did the authors match WACCM aromatics to the aromatics in WRF-Chem? Since the WRF-Chem domain used in this study is small, I think boundary conditions can be also important. A table with species mapping information, especially for OA and OA precursors, would be helpful to understand. Our apologies – we should have noted here that the SOA scheme taken from Tsimpidi et al (2010) has been mapped onto the CRI-v2-r5 scheme, and that, instead of the 'anthropogenic' compounds (from that scheme) we have mapped the SOA production reactions onto TOLUENE plus BENZENE, for ARO1, and OXYL (xylene and other aromatics), for ARO2. For the 'biogenic' compounds we have used isoprene, and APINENE to represent monoterpenes. We have added more details on the choice of SOA precursors to the pape. We have also added to the supplementary material the mapping for WACCM to CRI-r2-v5 that was used. Also, we mistakenly gave the size of the subset of the domain used (55x55) for the analysis presented in this manuscript, not the full model domain (127x127) simulated in WRF-Chem, this has been updated in the manuscript.

5) Table S1: Please add the corresponding saturation vapor pressure value for each VBS bin. And the unit of the aging rate is missing. Notes have been added to Table S1 giving these details.

6) Line 140: Emission inventories usually provide OA emissions as carbon mass, not matter mass (e.g., HTAP). How did the authors convert carbon mass to matter mass when those are emitted in the model? Is it implicitly considered when the authors apply the scaling factor of 3? Please clarify that. We assumed a ratio of matter mass to carbon mass of 1.4, dividing the emission inventory matter mass by this to get the carbon mass. Within the model each VBS compound is stored as two variables, the oxygen part and the 'non-oxygen' part. When adding emissions we multiple the carbon mass by 1.17 to get the 'non-oxygen' mass (carbon, plus other atoms), and by 0.08 to get the oxygen mass. These scaling factors were taken from Shrivastava et al, ACP, 2011. We then apply the SVOC scaling factor, and volatility distribution, to give the final SVOC emission profile. The IVOC scaling factor is applied to the same base emissions to get the IVOC emission profile.

7) Line 144: Please specify the version of MEGAN that this study is using. Depending on the version, the authors may need to change the reference to Guenther et al. (2012). WRF-Chem v 3.8.1, which we use for this study, includes MEGAN v2.04 (Guenther et al., 2006). We have added the MEGAN version number to Table 1.

8) Line 189: Can the authors elaborate on the meaning of this scalar variable? Can the authors convert/relate it to some variable with a physical meaning, such as photochemical age?

The scalar variable is the result of our attempt to create a sensible range of possible emitted volatility distributions. We needed a method by which we could represent the variation of possible volatility distributions within the process emulator. The direct approach would be to include a scaling factor for each volatility bin as parameters. This would not have worked because it would have exploded our parameter space (which is already too large), and these parameters would not be independent of each other, leading to a lot of wasted space and inefficiency in the final emulator.

We decided that we needed to use a single variable to represent the variation in the emitted volatility distribution. We also wanted to have volatility distributions which looked fairly sensible – rather than randomly scattering mass through the volatility range. Something which spanned the range of 'all gaseous', through an even spread of volatilities (and so more evenly partitioned between gas and condensed phases), down to 'all condensed'. We considered using a gaussian distribution, using a mean volatility, width, magnitude, and skew to describe the distribution. But decided against this because it would still involve adding too many variables, which would all be quite dependent on each other, into the process emulator. We could have distilled a sensible range of gaussian distributions

into a single scalar variable, on a very similar scale to that used here – and this is certainly an option which anyone else looking to use process emulators to further explore could use. However, we felt that using a simple reaction model, where each step a fraction of each volatility bin would be 'aged' and moved to the next volatility bin, would be a simpler and more appropriate method for creating the volatility distribution. This approach also allowed us to include some simple partitioning, with aging process stopped for any condensed matter; replicating the behaviour of the model these distributions will later be injected into.

# Given that we used a simple, fractional, aging process, we don't feel that it would be appropriate for us to try to relate it to a physical variable. We have included Figure S2 instead, which gives example volatility distributions through the range of this scalar value used in for our study.

9) Table 2: Please clarify that "anthropogenic" here includes traditional VOCs (e.g., benzene, toluene) or only for semi-volatile POA. The 'anthropogenic' and 'biomass burning' descriptions in Section 2.3 (and also Table 2) only refer to the two semi-volatile POA schemes that we are perturbing for this experiment. We have amended the text in Section 2.3, and caption for Table 2 to make this clear.

10) Line 251: Section S2 -> S3. The modification has been made in the manuscript.

11) Line 255-256: Although the VBS only affects OA and PM, doesn't it affect inorganic aerosols through microphysical processes and gases through changing clouds and aerosol-radiation interactions? This is a fair point – we have been too simplistic in our description of the OA and inorganic aerosol interactions here. We have amended the sentence to read: "As mentioned in the methods section, the VBS setup will directly affect OA concentrations and PM. The oxidative budget for inorganic chemistry is not directly affected, however, by changing the aerosol size distribution there are some indirect effects on inorganic aerosol and gaseous species through changes in aerosol water content, cloud fields, and aerosol-radiation interactions".

12) Line 258-259: The model also substantially underestimates CO. I understand that this is not easy to fix, but are there any previous studies that reported the same problem in this area? This can be also applied to PM2.5 and NO2, but I think those are not necessary unless there is a previous modeling study at the same location, because those are short-lived species.

We are not aware of previous any independent studies for this region which have investigated CO model predictions. Most air-quality studies for India have focused on particulate pollution. One exception was Jena et al (2015), who compared OMI satellite measurements of NOx with WRF-Chem (MOZART-4 gas-phase chemistry) predictions using a number of different emission datasets. They found that model predicted NO<sub>2</sub> tropospheric columns, using bottom-up emission datasets, were generally higher than the OMI measured NO<sub>2</sub> columns over the IGP region. They suggest that this indicates a general overestimation in NOx emissions in these datasets, which would tie in with what we have observed in this study too.

Recently Jat et al (2021) used WRF-Chem (though with CBM-Z gas-phase chemistry, and with the basic MOSAIC aerosol scheme), with EDGAR-HTAPv2 emissions processed using our emission tools, and also found significant underprediction of CO at measurement sites across India during the 2015-16 winter months. We believe it is likely this is due to an underestimation of CO emissions for India in the EDGAR-HTAPv2 inventory.

Jena, C., Ghude, S.D., Beig, G., Chate, D.M., Kumar, R., Pfister, G.G., Lal, D.M., Surendran, D.E., Fadnavis, S., van der A, R.J., 2015. Inter-comparison of different NOX emission inventories and associated variation in simulated surface ozone in Indian region. Atmos. Environ. 117, 61–73. https://doi.org/10.1016/j.atmosenv.2015.06.057. 13) General: I understand that making a table with all the parameters used for all 111 model runs would be overkill, but it would have been helpful if the authors could provide a table with a set of parameters for some runs, such as model runs used in Figure 4. It is difficult to read parameters from Figures 5 and S7. Table S2 (now Table S4) in supplementary material has been updated. The 111 runs have been added with the values of the 10 parameters.

14) Line 294-295: Even with the same PBL height, the concentrations can be significantly changed due to the PBL mixing scheme. For example, the early morning peak of OA shown in the model does not exist in the observation, although there are good agreements between the model and the observation before noon. Here or in the methodology section, the authors could discuss the PBL mixing scheme and potential errors from it. We talk about it in lines 294-298 and mention that due to this lack of agreement in the diurnal cycle, we decided to select a period with no structural errors (14-16 hrs).

15) Figure 9 and S13: Axis labels are impossible to read. We are sorry for this, they are clearer in our original pdf figures. We will discuss with the ACP production team how to best improve the readability of the figures in the final paper.

16) Line 432: These plots -> Figure 10 The change has been made in the manuscript.