Response to reviews of ACPD submission:
Linking gas, particulate, and toxic endpoints to air emissions in the Community Regional Atmospheric Chemistry Multiphase Mechanism (CRACMM) version 1.0

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Reviewer comments are reproduced in black with responses in blue. Modifications to manuscript are underlined ~~or indicated with strikethrough if deleted~~.

Reviewer #1

This manuscript developed a new chemical mechanism for 3D chemical transport modeling, CRACMM, which represents a major advance compared with the chemical mechanisms used in previous versions of CMAQ. I believe that the new mechanism will benefit the air quality modeling community, especially the researchers working on O3 and SOA simulations. The manuscript is clearly written. I think it can be accepted for publication after the authors address the following minor comments and suggestions.

We thank the reviewer for their supportive comments.

(1) CRACMM builds on the implementation of RACM2 chemistry coupled with aerosol chemistry of AERO6. As we know, AERO6 treats both organic and inorganic aerosol chemistry, but this manuscript describes only organic chemistry. Did you treat inorganic aerosol chemistry within or outside of CRACMM?

The specification of CRACMM (rather than a gas-phase mechanism + aerosol module as in RACM2_AE6 in CMAQ) is meant to emphasize that the gas-phase configuration is tied to the aerosol-phase configuration because the representation of aerosol, SOA in particular, involves specific gas-phase chemical intermediates. CRACMM includes a species list (coded as a namelist in CMAQ) that includes inorganic aerosol species like calcium, nitrate, etc. and the gas-phase mechanism includes SO_2 reaction that forms sulfuric acid (that will condense as sulfate). Thus, the term CRACMM refers to a specific inorganic aerosol configuration in CMAQ. CRACMM's CMAQ implementation continues to employ operator splitting. For example, ISORROPIA II is used to calculate thermodynamic equilibrium and driving forces for nitrate/nitric acid partitioning.

We add (Sect. 3):
"These 51 particulate species in CRACMM include inorganic aerosol species such as sulfate, nitrate, ammonium, calcium, and other trace metals as in previous versions of CMAQ. To fully describe the state

of atmospheric aerosol in CMAQ, CRACMM interacts with ISORROPIA II (Fountoukis and Nenes, 2007) and other algorithms describing nucleation and condensation."

(2) You simulated aging in addition to the initial oxidation of alkane-like species. Aging changes SOA yields. Are the simulated SOA yields of alkanes still consistent with chamber experiments after aging is considered?

We do find that our prompt alkane yields (Table 1) are similar to prompt laboratory yields (Figure S5) as mentioned in the paragraph on line 329 (ACPD version). Our aged SOA yields have been compared to the 2-D VBS (Figure 4) and show similar behavior. The evolution of alkane SOA yield with aging could be evaluated in future work using experimental data.

(3) Line 138: Do you mean gaseous L/S/IVOC emissions only, or both gaseous and particle-phase L/S/IVOC emissions?

The calculation of potentials (ozone formation potential, and OA formation potential) assumes that all species eventually undergo gas-phase reaction which gives an upper estimate on ozone and OA formation potential compared to assuming some portion of the mass is sequestered from reaction in the particle. For the property analysis of volatility, hydroxyl radical reactivity, and number of carbon (Fig. 1-3), the phase is not relevant. We do not provide POA formation potential which could be calculated when needed based on the species volatility and ambient concentration. See additional clarification added for the next item.

(4) Line 140: Are any SVOC emissions considered here?

Emission inventories sometimes exclude certain S/IVOC species from total ROC emissions due to gaps in gas vs particle-phase measurement techniques. No "missing" SVOCs due to measurement technique exclusion were added for biomass burning. SVOCs emitted as part of inventoried POA were considered.

We add the following to clarify how inventoried POA is treated in the emission analysis:
"L/S/IVOC emissions inventoried as part of primary PM$_{2.5}$ were estimated using published volatility profiles for vehicles (Lu et al., 2020) and wood burning (May et al., 2013; Woody et al., 2016). Other sources of POA were assumed to behave as a species with C_i^* of 10^{-2} µg m^{-3}."

(5) Line 154: What kinds of compounds are these? Can they be IVOC?

Unidentified, exempt compounds are measured in the gas phase and could be IVOCs (we don't know what they are).

(6) Line 180: You assumed equal RO2 reaction rates with HO2 and NO here, but what is the amount of RO2 reacted with HO2 vs NO? The latter determines whether this is a high-NOx or low-NOx condition and hence determines the SOA yields.

This line refers to the literature data assembled for creating the SOA yield simple SAR and specifically some SOA yields that were explicitly set rather than fit. Note, the SOA yield parameterization is meant for screening analysis and is not a rigorous representation of SOA yield which depends on ambient organic aerosol concentration, RO$_2$ fate, oxidant abundance, and other factors. For aromatics and monoterpenes, the RO$_2$ fate was not reported in the underlying data. We now use low NO$_X$ to refer to experiments with no added NO$_X$ and high NO$_X$ for the experiments with NO$_X$ added).

Text was updated to:
"Explicit yield assignments were made based on published data in the case of sesquiterpenes, monoterpenes, benzene, toluene, and xylene (Pye et al., 2010; Ng et al., 2007). Published single-ring aromatic yields were scaled up by the vapor wall loss factor (Zhang et al., 2014). An OA concentration of 10 µg m^{-3} and equal low-NO$_X$ vs high-NO$_X$ behavior ~~RO2 (organic peroxy) reaction rates with HO2 (hydroperoxyl radical) ("low-NOx") and NO (nitric oxide) ("high-NOx")~~, typical of northern hemisphere July conditions (Porter et al., 2021), were assumed for these explicit yield assignments."

In the case of other data used to build the SOA yield SAR, most data were for high-NO$_X$ (NO$_X$ added) conditions. The following was updated:

"In the case of OA potential, several sources, largely following ~~what is outlined~~ high-NO_X conditions as outlined in the work of Seltzer et al. (2021), were aggregated to estimate the SOA yield of individual species."

(7) Line 190: Which of the three methods did you actually use? Multiple linear regression, exponential/logarithmic equation, or averaging?

The fit depends on the system. The sentence has been revised:
"Within a given class, the MIR was fit ~~via multiple linear regression, an exponential/logarithmic equation, or through averaging~~ as a function of number of carbons per molecule, HO rate constant (from OPERA), number of oxygens, number of double bonds, number of ring structures, number of double bonded oxygen, and/or number of branches depending on the chemical class."

The fits are available in the supporting code (U.S. Environmental Protection Agency, 2022a) at: https://github.com/USEPA/CRACMM/blob/main/utilities/mir_estimates.py.

(8) Are these alkane-like L/S/IVOC species emitted into just the gas phase, or both gas and particle phases?

CRACMM provides the properties of the species (volatility, solubility, etc) to calculate gas-particle partitioning. Properties in Fig 1-3 do not depend on phase. Prohibiting L/SVOCs from contributing to HO reactivity or ozone formation potential (e.g., assuming they are sequestered in nonreactive particles) would not significantly affect conclusions about their role as it is already estimated to be small (Fig. 9). The OA potential of an individual L/SVOC could be overestimated by a maximum of 33% (yield of 150% instead of 100% partitioning) if it were sequestered from reaction due to emission and permanence in the particle (which is unlikely). Note that HO reactivity, O_3 formation potentials, and OA formation potentials do not consider competing losses such as deposition and thus do not replace the need for full model calculations.

(9) Line 372: "The decrease in $\log_{10}(C^*)$ per oxygen in the 2-D VBS box model was set at -2.3". This is likely the largest volatility decrease one oxygen addition might bring. This is a stronger volatility decrease than the default assumption in the 2D-VBS box model. The authors may want to note this in the manuscript.

Our original description was too simplistic and mischaracterized the impact of the parameters chosen for accounting for the decrease in C* with oxygen. We used Eq. 3 from Donahue et al. (2011), which includes an oxygen-oxygen interaction term ($b_O = 2.3$) and a carbon-oxygen nonideality ($b_{CO} = -0.3$) parameter. The equation for saturation concentration as a function of C and O is then:

$$log_{10}C_i^o = \left(n_C^0 - n_C^i\right)b_C - n_O^i b_O - 2\frac{n_C^i n_O^i}{n_C^i + n_O^i} b_{CO}$$

with $b_C = 0.475$, $b_O = 2.3$, $b_{CO} = -0.3$, and $n_C^0 = 25$. At low $n_O{:}n_C$, the equation for C^* reduction simplifies to:

$$\Delta log_{10}C_i^o = log_{10}C_i^o - \left(n_C^0 - n_C^i\right)b_C = -n_O^i\left(b_O - 2b_{CO}\right) = -1.7n_O^i$$

which is consistent with Chuang and Donahue (2016) and similar to the volatility reduction expected per oxygen for carboxylic acid addition. This increases to -1.925 at $n_O{:}n_C = 0.6$.

We have updated the manuscript text:
"The chemistry of secondary oxygenated L/S/IVOCs is parameterized using the 2-D VBS framework (Donahue et al., 2012) with some modifications. The decrease in $log_{10}(C_i^*)$ per oxygen in the 2-D VBS box model was calculated using the parameterization from Donahue et al. (2011) with the oxygen-oxygen interaction term set to 2.3, the carbon-oxygen interaction parameter set to -0.3 to correct for the behavior of diacids, and ~~set at -2.3, roughly equivalent to the magnitude expected for an alcohol (Pankow and Asher, 2008). T~~the ~~decrease in $log_{10}(C_i^*)$ per carbon was~~carbon-carbon interaction term set to 0.475 ~~with a carbon-oxygen interaction parameter (-0.3) to correct for the behavior of diacids (Donahue et al., 2011)~~. As identified in Donahue et al. (2011), the resulting decrease in $log_{10}C^*$ per oxygen is 1.7~~at low $n_O{:}n_C$ is 1.7n_O,~~ as -$n_O{:}n_C$ approaches zero and and is 1.~~,~~93 at $n_O{:}n_C$ approaches= 0.6. These values are consistent with the effect ~~per oxygen~~ of adding carboxylic acids to an alkane-like molecule (Pankow and Asher, 2008)..."

(10) Line 387-390: How did you select these species?

Based on detailed simulations with the 2D-VBS box model, we noted two major pathways for the evolution of oxidation products, broadly consistent with previous literature documenting the 2D-VBS. One pathway is the direct functionalization of reactants at low O:C, shown in Fig. R1 by the three arrows moving up and to the left in $\log_{10}C^*$-O:C space. To resolve this path, we ensured that every $\log_{10}C^*$ bin included at least one OXY species at mean O:C ≥ 0.1 and that lower volatility classes had 3 species, spanning a range of O:C, and bounding the higher O:C products observed in offline testing.

The second pathway, a feature of the increased role of fragmentation at higher O:C, yields products in the blue and pink regions of Fig. R1. Species OP2 was already present in the RACM2 mechanism. We added species OP3 to capture semivolatile compounds with moderately high O:C. These compounds are likely to fragment when oxidized with HO (f_{frag} = 76-82%) according to the parameterization for fragmentation branching ratio employed ($f_{frag} = (O:C)^{0.4}$).

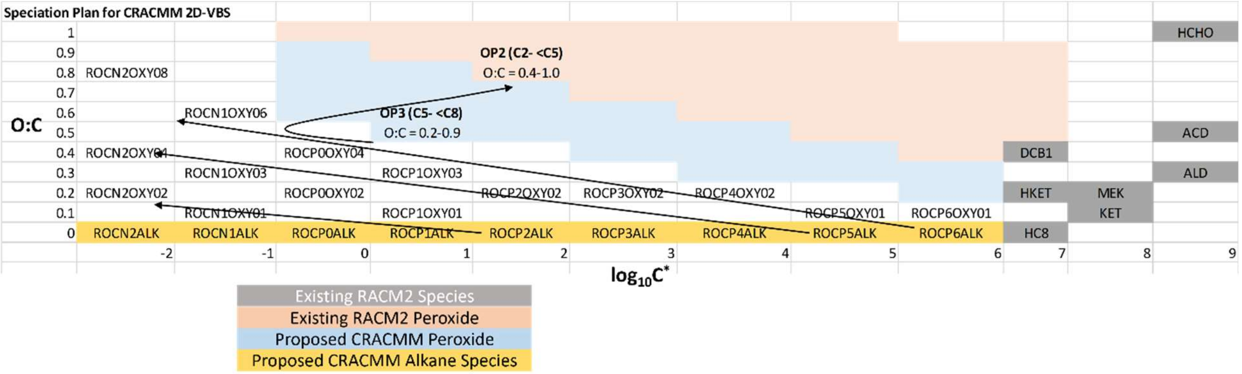


Figure R1. Schematic of ROCALK and ROCOXY species in C^* and O:C space.

(11) Line 401-403: Some products are mapped to aldehydes and some are mapped to ketones. Any science behind this assumption?

The mapping decisions for fragmentation products were made based primarily on carbon number. We limited the product species to those stable species already available in the RACM2 species list. If future evaluation of CRACMM indicates there are products of particular functionalities and carbon number that are underrepresented with this configuration, it will be updated at that time.

We have added the following to the text:
"The choice of functionality of the product species (e.g., aldehydes versus ketones) is entirely determined by the RACM2 species that were already available at each carbon number. Future measurements of the low molecular weight species produced by the oxidation of larger compounds would help constrain this choice and motivate the addition of new CRACMM species."

(12) Line 459-461: Among the products of furan, the model assumes that only furanone leads to SOA production. Is this true?

Yes, we assume all furan SOA forms from later oxidation of furanone. The ring-retaining channel (furanone) has a high potential of retaining the carbon backbone and functionalizing while the ring-opening channel (DCB1 - Unsaturated dicarbonyls) has a higher chance of fragmenting leading to more volatile products. This assumption is consistent with Jiang et al. (2019), who detected many ring-retaining products in the particle-phase. If more experimental studies are performed in the future to better determine the mechanistic formation of SOA from furans or other VOCs, we can update CRACMM accordingly.

We have added the Jiang et al. reference:
CRACMM assigns SOA from FURAN to further reactions in the ring-retaining product channel, FURANONE, consistent with products detected by Jiang et al. (2019) ~~which reacts to form ketones, glyoxal, and SOA~~.

(13) Line 516: From R9-R13, it is not clear how furanone was produced from aromatics oxidation.

Furanone is produced from aromatic RO_2+NO alkoxy radical products. We add a reference to reaction 477 in Appendix B as an example:

"Further oxidation of furanone produced from aromatic oxidation (e.g., Reaction 477, Appendix B) also results in small amounts of SOA (Sect. 3.4)."

(14) Line 522-523: Will setting the yields to match high-NOx experimental results lead to an underestimation of SOA yields under low-NOx conditions?

The *phenol* and *cresol* SOA yields (a fixed value for each species) were set to reproduce high- vs low-NOx *benzene* and *toluene/xylene* SOA yields. The yield of phenol and cresol from single-ring aromatic hydrocarbons is independent of NO level, in good agreement with experimental data for conditions below a few hundred ppb NO (Bates et al., 2021). Currently, phenol and cresol SOA yields are also NOx-independent. Adding further detail to the phenol and cresol systems was beyond the scope of this initial implementation. From line 524:

"The molar SOA yield using this method is estimated as 15% by mole for phenols and 20% by mole for cresols (Table 1), within the range of 24-52% by mass for phenols and 27-49% by mass for cresols as summarized by Bruns et al. (2016). Future work should expand upon this phenolic SOA treatment…"

(15) Line 651: It is not clear from the text if the organic peroxide products (OPB) lead to any SOA in the model.

Line 651 indicates: "The OPB peroxides and TRPN nitrates are assumed to remain in the gas phase (see representative structures in Appendix A)."

Line 657 indicates: "Further reaction or photolysis of OPB is assumed to produce products like existing organic peroxide reactions in RACM2 with products fed back to the lumped aldehydes (ALD), ketones (KET), and a saturated C10 RO2 (HC10P)."

Table 1 shows OPB+HO SOA yields of <1% through the HC10P path.

OPB is not a major SOA source.

Note the entry in Table 1 for OPB showed HO twice and that was corrected in the revision.

(16) Line 712-714: Does this mean the SOA yields from API oxidation will be much higher under high-NOx conditions than under low-NOx conditions?

The SOA yield from API can increase with increasing NO as that promotes formation of terpene nitrates which can react to make SOA. Previous work has indicated a complex role for NO_X in modulating monoterpene SOA. Pye et al. (2019), predicted and observed monoterpene SOA to be enhanced in the presence of NO due to increased oxidant abundance in the downwind Atlanta plume. For summer 2013 conditions in Centreville, AL, a 25% reduction in NO_X emissions was predicted to lead to a 14% reduction of monoterpene SOA from HO and ozonolysis pathways and 25% reduction in monoterpene SOA from organic nitrate pathways (Pye et al., 2015). A fully explicit, multigenerational α-pinene SOA mechanism is not yet available (but is an active area of research for multiple groups), and the representation of α-pinene chemistry is likely to continue to evolve in CRACMM.

(17) Line 772-784: Does this section have anything to do with SOA formation?

No. Acrolein and 1,3-butadiene do not make SOA in CRACMM and thus do not have entries in Table 1. They were explicitly added due to their importance for health (classification as Hazardous Air Pollutants with high potential for health risk, Fig. 6).

(18) Line 1141-1142: I don't think it is appropriate to define this metric as saturation ratio. I think saturation ratio typically means the ratio of vapor concentration to saturation vapor concentration.

That is correct. The actual saturation ratio as the reviewer has defined it was used for a previous iteration of the figures before the current metric was employed. Unfortunately, the label was not appropriately updated. We have removed the use of saturation ratio as the reviewer suggests, revisited the usefulness of this x axis metric, and made some minor adjustments to improve clarity.

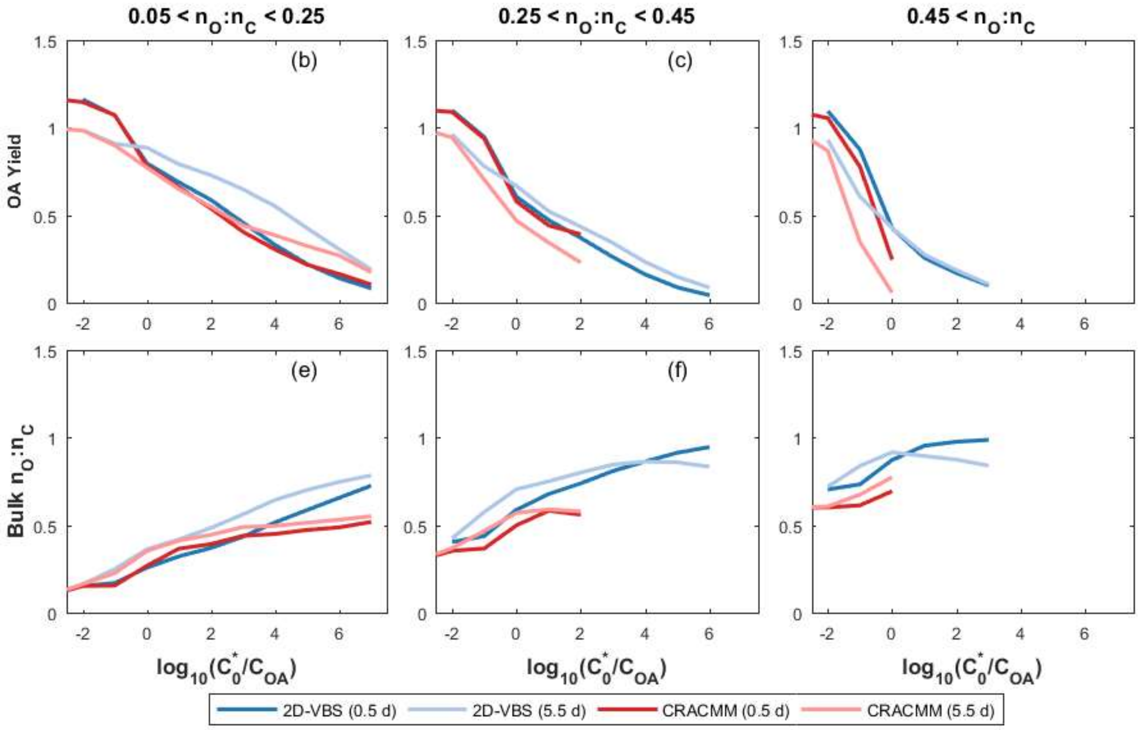


Figure 4: Organic aerosol yield and bulk $n_O: n_C$ predicted by the CRACMM oxygenated ROC aging mechanism (Sect. 3.2) and the 2D-VBS configuration reported by Zhao et al. (2016). The ~~saturation ratio~~x axis is defined as $log_{10}\left(\cancel{\frac{C_{OA}}{C_i^*}} C_0^*/C_{OA}\right)$ where C_{OA} is the background OA concentration and $C_{0\cancel{i}}^*$ is the saturation concentration of the precursor. The aging of each species is simulated at a constant HO concentration of 10^6 molec cm^{-3} for 12 hours (darker colors) and 2.5 days (lighter colors) at four different C_{OA} conditions (0.1, 1, 10, and 100 µg m^{-3}). In cases where multiple predictions are present for the same saturation ratio, values are averaged.

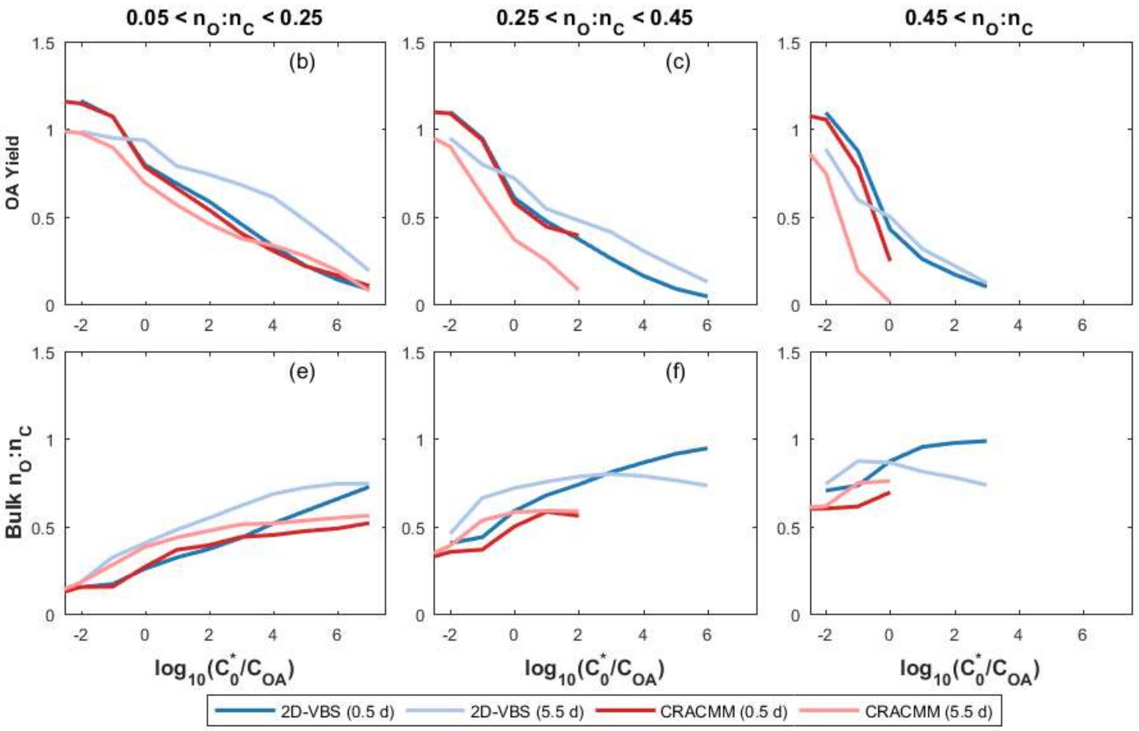


Figure S7: Organic aerosol yield and bulk O:C predicted for oxygenated ROC. Predictions are from the CRACMM oxygenated ROC aging mechanism and the 2D-VBS configuration reported by Zhao et al. (2015). The ~~saturation ratio~~x axis is defined as $\boldsymbol{log_{10}}\left(\frac{\cancel{C_{OA}}}{\cancel{C_i^*}}C_0^*/C_{OA}\right)$ where C_{OA} is the background OA concentration and $C_{0\cancel{i}}^*$ is the saturation concentration of the precursor. The aging of each species is simulated at a constant HO concentration of 10^6 molec cm^{-3} for 12 hours (black/blue) and 5.5 days (grey/cyan) at four different COA conditions (0.1, 1, 10, and 100 µg m^{-3}). In cases where multiple predictions are present for the same saturation ratio, values are averaged. This figure is the same as main text Figure 4 except the longer aging timescale is 5.5 days.

Reviewer #2

This paper aims to describe a new tropospheric organic-chemistry mechanism for use by the US EPA for understanding the concentrations of air pollutants such as O3, particulate matter and compounds hazardous to health.

It builds on a number of previous mechanisms but extends them to be more comprehensively relevant for SOA and compounds hazardous to health. It also provides a formal description of the chemistry and the choices made to develop this chemistry. This activity has been a substantial and thoughtful piece of work. It is very useful to the community to have the thoughts used in the development of the mechanism catalogued in a single location.

I don't have any real comments about the description of the mechanism. It is based on a number of well-regarded other mechanisms and brings them together. There will be by very necessity some inconsistencies between these mechanisms but I'm not to concerned about that.

We thank the reviewer for their supportive comments.

I have one comment for the editor about whether this paper is best published in ACP rather than GMD. The paper is a description of a model mechanism and the choices that went into developing it. It doesn't evaluate the mechanism against previously used mechanisms or against other mechanisms. Thus there isn't much in the way of "new science" here more documentation of a new mechanism. It would seem that this is ideally suited for a GMD paper whereas it's less clear that this is an ACP paper. What have we learnt from this paper? This is however an editorial decision rather than one for a reviewer so I would leave it at that.

We considered submitting to GMD but ultimately opted for ACP due to the information which seeks to understand the ambient atmosphere by examining properties of ROC emissions (Fig. 1-3), implications of ROC for health (Fig. 6), ambient trends in oxidation state (Fig. 7), particulate ROC in van Krevelen space (Fig. 8), and implications of ROC emissions for ozone and OA (Fig. 9). Section 5 is specifically focused on implications of the chemical evolution of ROC as described by the chemistry.

This paper also illustrates reasons why SOA should be coupled with ozone chemistry and that it is possible to do so. Specifically, several new SOA precursor systems (phenol, cresol, furanone) have been added to the mechanism for the first time, and Figure 5 illustrates why that must be done in a way that is coupled with commonly represented precursors such as benzene, toluene, and xylene.

Several things we learned from the analysis that are highlighted in the abstract:

- Inclusion of intermediate and lower volatility organic compounds were estimated to increase the coverage of anthropogenic and biomass burning ROC emissions by 40% compared to current operational mechanisms
- Integrating the radical and SOA chemistry enabled the implementation of previously unconsidered SOA pathways from phenolic and furanone compounds
- CRACMM organic aerosol species were found to span the atmospherically relevant range of carbon number, number of oxygens per carbon, and oxidation state with a slight high bias in number of hydrogens per carbon

At 88 pages, the current manuscript represents a significant amount of work; however, we anticipate fully describing CRACMM and documenting the impacts of the chemical representation will require multiple additional papers. This paper sets forth the argument that gas and particle phase chemistry should be coupled, shows how to do it, and provides documentation for the approach going forward. In the Discussion, we now cite a companion manuscript applying CRACMMv1 within CMAQ to predictions of ozone in the Northeast U.S. in summer:

"CRACMMv1.0 targeted SOA systems for development, but CRACMM updates impact O_3 as will be demonstrated for the Northeast U.S. in future companion work (Place et al., in prep.)."

To emphasize that this work sets out a framework, we removed version 1.0 from the title:
"Linking gas, particulate, and toxic endpoints to air emissions in the Community Regional Atmospheric Chemistry Multiphase Mechanism (CRACMM) ~~version 1.0~~"

My major comments can be split into a number of categories.

1. The introduction is written from a particularly EPA perspective. This is quite US-centric at times. The discussion around O3 bias in models (line 63) seems to suggest all models suffer from the same problems and only discusses the US. I appreciate that the development work here has been done with the US in mind. Still, I think the introduction should be looked at to make it clear that the comments are nearly always relevant only to the US and mainly comes from the perspective of people running EPA models.

The mechanism is not intended to be unique to the U.S. as more than half the CMAQ user base is outside the U.S. (https://www.epa.gov/cmaq/cmaq-user-community#CMAQ-Impact). The introduction (line 115) states "While the mechanism is presented here in the context of U.S. conditions, it is informed by conditions outside the U.S. (e.g., the work of Zhao et al. (2016) for China) and is meant to be generally relevant for tropospheric chemistry."

The secondary oxygenated ROC aging scheme (Section 3.2) is based on parameters developed for China (Zhao et al., 2016).

We have added additional references to the introduction highlighting conditions outside the U.S. and moved a few sentences:

“Atmospheric chemical mechanisms connect ROC emissions to endpoints like SOA, O_3, and secondary HAPs and are used to inform air quality management strategies to mitigate the impacts of air pollution. Chemical mechanisms were traditionally designed for estimating ambient O_3 although not necessarily the lower levels of O_3 observed today ~~in the U.S.~~ (Kaduwela et al., 2015) or sources of growing importance ~~like volatile chemical products (VCPs) (Coggon et al., 2021)~~around the globe such as volatile chemical products (VCPs, also referred to as solvents) (Coggon et al., 2021; Karl et al., 2018; McDonald et al., 2018; Zheng et al., 2018) and biomass burning (Jaffe and Wigder, 2012)~~. Controls on combustion~~ that are changing the composition of emissions~~ are shifting cities in the U.S.~~ towards increasingly oxygenated ROC (Venecek et al., 2018)~~ compared to the alkane-dominated conditions of the 1990s (Middleton et al., 1990). While mechanisms may predict O_3 reasonably well on broad spatial and temporal scales (fractional biases in O_3 are typically much less than 20% for models examined by Simon et al. (2012)), model-predicted O_3 can be biased low by 5 to 10 ppb (>20%) in wintertime western U.S. conditions and biased high by more than 5 ppb across the U.S. south in summer compared to observations (Appel et al., 2021). In addition, mechanisms~~. While mechanisms may predict O_3 reasonably well on broad spatial and temporal scales (Simon et al., 2012; Xing et al., 2015; Young et al., 2018), regional biases in predicted O_3 can exceed 10 ppb (Young et al., 2018; Solazzo et al., 2017) or 20% (Appel et al., 2012; Appel et al., 2021). Global model estimates of chemical production and loss of ozone also vary by a factor of ~2 (Young et al., 2018), and emerging chemical pathways missing from standard models, such as particulate nitrate photolysis, can increase free tropospheric ozone by 5 ppb (Shah et al., 2023) indicating a continued need for model development for ozone prediction. Furthermore, even when mechanisms are relatively similar in their O_3 predictions, they can differ substantially in terms of predicted intermediates like the hydroxyl radical (HO) and nitrate radical (NO_3) as well as products like formaldehyde and SOA ~~even if they are relatively similar in their O_3 predictions~~ (Knote et al., 2015)~~.~~. Model representations of organic aerosol are particularly diverse and span a factor of 10 in their estimates of global SOA source strength (Tsigaridis et al., 2014). Given parts of 22 different states are in marginal attainment to extreme nonattainment for the current U.S. 8-hour (2012) O_3 standard (as of August 2022) (U.S. Environmental Protection Agency, 2022b) as well as recent work demonstrating health effects below the current fine particle standards (Makar et al., 2017), increasingly accurate representations of emissions and how they connect to chemistry will be needed to inform air quality management strategies going forward. In addition, future implementation of global air quality guidelines, such as those from the World Health Organization, may need to account for the speciation of ambient aerosol since different species have different anthropogenic contributions (Pai et al., 2022).

…”

2. The paper focuses on the completeness of the mechanism developed but it should be clear that it ignores some important aspects. There is no representation of much of sulfur oxidation chemistry (DMS), halogen oxidation chemistry, or representation for HO2 uptake etc. These might be seen as boundary conditions for the primary objective here but there should be some explanation for why they have not been included, especially for things like chlorine chemistry which has been shown to have an impact on ozone in coastal locations etc. I think the introduction should spend some time putting some context around the developed mechanism. Why does it what it does but also what does it exclude things and why? Again in the section discussing future developments it would be worth including some description of the plans for including some of these other aspects of chemistry in future versions of the mechanisms.

This manuscript represents the initial version of CRACMM. In the introduction, we clarify the purpose: “~~The CRACMM effort includes development of rules for mapping emitted ROC to mechanism species and aims to improve representation of atmospheric chemistry by closely coupling the pathways to O_3 and SOA as well as representing several HAPs explicitly. While the mechanism is presented here~~The purpose of the CRACMM version 1.0 effort described here is to demonstrate a coupled representation of NO_X-ROC-O_3 chemistry including SOA and consideration of HAPs. In addition, this work includes development of rules for mapping emitted ROC to mechanism species and updates to rate constants leading to a publicly available mechanism upon which further developments can be built. CRACMM is expected to become the default option in CMAQ in the future (U.S. Environmental Protection Agency, 2021). While the mechanism is presented in the context of U.S. conditions, it is informed by conditions outside the U.S. (e.g., the work of Zhao et al. (2016) for China) and is meant to be generally relevant for tropospheric chemistry. ~~CRACMM will be available in the public release of CMAQv5.4 (expected in 2022) and is distributed as a stand-alone mechanism.~~CRACMM is available in the public release of CMAQv5.4 (U.S. EPA Office of Research and Development, 2022) and is distributed as a stand-alone mechanism (U.S. Environmental Protection Agency, 2022a). In.~~”~~

The first paragraph of Sect. 3 stated “ROC systems not previously represented in RACM2 (such as furans and L/S/IVOCs), precursors to SOA, and systems with new kinetic data (Sect. 3.10) were targeted for development in this initial CRACMM version.” In Sect. 3, we add:
“Future work will continue to expand this initial representation by extending it to new chemical systems and/or updating these parameterizations with new data.”

In the abstract, we highlight CRACMM is a demonstration of an approach:
This work builds on the Regional Atmospheric Chemistry Mechanism version 2 (RACM2) and develops the Community Regional Atmospheric Chemistry Multiphase Mechanism (CRACMM) version 1.0, which demonstrates a fully ~~couples~~ coupled representation of~~the~~ chemistry leading to ozone and secondary organic aerosol (SOA) with consideration of HAPs.

Developing a list of future updates is beyond the scope of this work. Note that while halogen chemistry is not explicitly included, the mechanism includes an empirical parameterized loss of ozone that was fit to reproduce halogen effects in a simulation with full halogen chemistry (Sarwar et al., 2015). This reaction is labeled HAL_Ozone and mentioned in footnote “e” of Appendix B and we add mention in the text (Sect. 3) as well:
“CRACMM specifically builds on the implementation of RACM2 chemistry coupled with aerosol chemistry of AERO6 (411 reactions) in the CMAQ v5.3.3 model which differs slightly from the original RACM2 implementation (Goliff et al., 2013) (363 reactions) due to SOA pathways, parameterized effects of halogens on ozone (Sarwar et al., 2015), and other minor updates (see the work of Sarwar et al. (2013) and Code Availability section for the CMAQ implementation of RACM2).”

3. Peroxy-radical self reactions. Historically these reactions have not been considered too important in these kinds of models as there is usually enough NO around for the fate of the peroxy radicals to be dominated by the reaction with NO rather than the reactions with other peroxy radicals. However, as NO emissions drop that assumption may be less convincing. However, including the reaction between each peroxy radical and each other add enormously to the number of reactions if implemented explicitly. It would be useful to have some comments here about the choices made for the fates of peroxy-radicals in the mechanisms and why the choices were made as they were.
In general, CRACMM includes a representation of RO$_2$ reaction with NO, HO$_2$, methl peroxy, and acetylperoxy radicals (standard set) for each organic peroxy radical. In some cases, the RO$_2$ can also undergo autoxidation (see Table 1 and Sect. 3) which may be implemented with a fixed yield (for fast reactions) or as a competitive fate. In some cases, specific RO$_2$ can react with other specific RO$_2$.

RO$_2$+RO$_2$ reactions are common in chemical mechanisms. Previous work found methyl peroxy radicals and acetylperoxy radicals were the most abundant RO$_2$ and thus are always represented in RACM2 (Stockwell et al., 1990) and now CRACMM (mentioned on line 505 and elsewhere). Previous work with the SAPRC mechanism (Pye et al., 2015) indicated that RO$_2$+RO$_2$ reactions can be 40% of the nocturnal RO$_2$ fate in the summer southeast U.S.

In addition, some RO$_2$+RO$_2$ reactions have unique products (dimers) of high interest for new particle formation. As a result, CRACMM includes RO$_2$+RO$_2$ reactions leading to C$_{20}$ dimers as well as monoterpene-RO$_2$ + isoprene-RO$_2$ cross reactions (Sect. 3.7). These are described in the paragraph on line 686 in the monoterpene section. Since we only included a limited number of cross RO$_2$ reactions (other than the standard set), we have a paragraph on line 743 noting that an application of CRACMM to new particle formation is likely needed to refine which RO_2+RO_2 reactions are most critical, and the specificity of product volatility needed.

Figure 1,2,3 Many of the figures are extremely data rich which is excellent however this can make understanding the detail of these rather hard. I find the use of the violin plots quite difficult to get my head around as its suggests compounds with non-integer values for the number of carbons. I think the overlain box plots show the 25th,50th and 75th percentiles of the carbon number not the emitted mass?

We provide the overlaid boxplots (with standard percentiles as indicated in the caption) since we recognize the violin plots are not as common and could be more difficult to interpret. The violin plots and boxplots both show emission weighted distributions. To create an emission weighted distribution, we take the emissions of all individual compounds within a CRACMM species. We truncate emission magnitudes to the nearest hundreds of Mg/yr (tenths of Gg/yr). Species with less 100 Mg/yr of emission are thus dropped (indicated in caption). This dropping of species with <100 Mg/yr of emissions for visualization avoids CRACMM species being depicted based on very trace species. Logistically, we then create a column of data in which the species appears once for every 100 Mg/yr of emission and use python to create the weighted boxplots and violin plots (similar to https://stackoverflow.com/questions/23412533/weighted-boxplot-in-pandas). Our exact code used to make Fig. 1-3 is provided in our public supporting data repository (linked as an asset and listed in the Code and Data Availability section as https://doi.org/10.23719/1527956) in a file labeled cracmm_visualize.py (function wtviolinplot_cracmm).

As an example of how data is visualized, consider the following distribution of individual compounds (cp1-cp5) within a lumped model species and their number of carbons per molecule:
100 Mg/yr of cp1 with nC=2
100 Mg/yr of cp2 with nC=3
100 Mg/yr of cp3 with nC=4
1000 Mg/yr of cp4 with nC=5
10000 Mg/yr of cp5 with nC=6

To prepare a set of data to represent this distribution for a weighted boxplot, violin plot, or histogram, cp1 would appear once, cp2 once, cp3 once, cp4 10 times, and cp5 100 times.

If these species were all counted equally and their emission magnitude ignored, then the median number of carbons (center of boxplot), nC, would be 4. However, less than 3% of the emissions are nC=4 or less. A more accurate median is 6 because there more nC=6 emissions than all the other emissions combined. Because our emission inputs had the potential to populate ~3000 different species (the size of the SPECIATE database), it was important to incorporate their abundance in visualization.

Are the colours representing the new species added to the system above the RACM2 mechanism, if so I think they should come below the grey bar in the key to signify they are additional to the base.

Figure 1's legend shows that grey bars are "RACM2 systems" and we prefer to leave it at the bottom of the legend so it doesn't appear like a heading. The figure caption has been updated to read that colors are "for families of species in Sect 3 that are either new or substantially updated compared to RACM2."

What is the difference between the RACM2 systems and the CMAQv5.3.3.

RACM2 does not include a description of PM2.5 formation which is handled by AERO6 in CMAQ. CMAQv5.3.3 refers to a specific version of RACM2.

From Sect 3 with modification:
"CRACMM specifically builds on the implementation of RACM2 chemistry coupled with aerosol chemistry of AERO6 (411 reactions) in the CMAQ v5.3.3 model which differs slightly from the original RACM2 implementation (Goliff et al., 2013) (363 reactions) due to SOA pathways, parameterized effects of halogens on ozone (Sarwar et al., 2015), and other minor updates (see the work of Sarwar et al. (2013) and Code Availability section for the CMAQ implementation of RACM2)."

I think these graphs are really informative but I have found it quite confusing to understand what exactly they were telling me. I'd suggest the authors go back and think about the figure and figure caption and from the perspective of a reader who is more detached from the figure than they might be.

> We appreciate that the information is dense which is why we included the boxplots as a more familiar visualization. This comment is available publicly and provides a detailed example with reference to the exact code used.

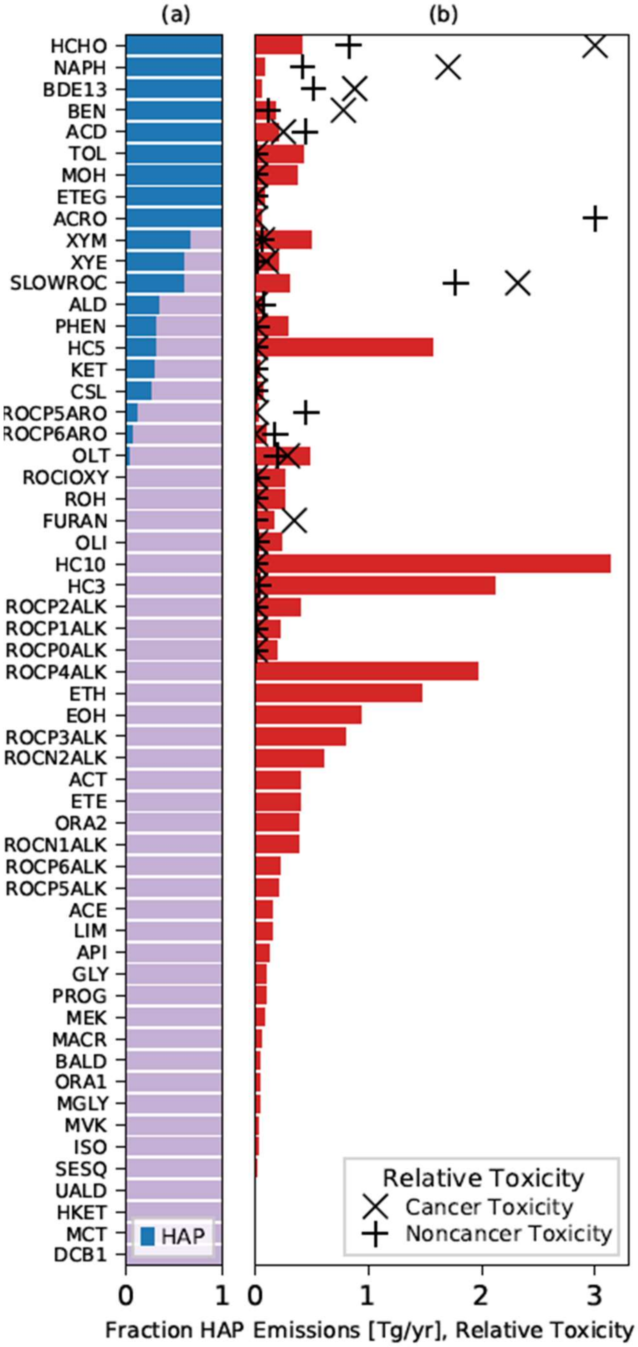
There are some minor typos in the brackets etc of some of the rate constants.

> We have corrected typos (see tracked changes) and modified the Appendix B caption to highlight a copy of the mechanism is available in the supporting data archive and CMAQv5.4.

Additional updates

1. CMAQv5.4, which is the first public release including CRACMM, was released in October 2022 and a reference to the code has been added.
2. We used a pre-release version of SPECIATEv5.2. SPECIATEv5.2 has now been released and the citation updated to include a URL.
3. RACM2 (and CRACMM) refers to the OH radical as HO. Both were used in the previous version. OH has now been replaced by HO (except in k_{OH}).
4. Some sentences in Section 5 were moved around (no changes in content, see tracked changes).
5. We added a reference in section 3.8:
   "Glyoxal SOA may include formation of salt-like structures in the aerosol phase (Paciga et al., 2014), but for simplicity, the oligomeric structure of Loeffler et al. (2006) is used as the representative structure of all glyoxal and methylglyoxal SOA."
6. In a few places, we refer to the cancer and non-cancer risk of emitted species. We have clarified the wording to be specific that we calculated emission weighted toxicity values (equivalently toxicity weighted emissions). Minor edits are in Section 4 (see tracked changes) and Figure 6 and its caption were updated:

Figure 6: Distribution of hazardous air pollutants (HAPs) across CRACMM emitted species. Panel (a) indicates the mass fraction of 2017 U.S. anthropogenic and biomass burning ROC emissions by CRACMM species that are HAPs (blue). Panel (b) indicates the magnitude of emissions in Tg yr^{-1} by CRACMM species (bars) and the emission-weighted toxicity for ~~relative potential for~~ cancer (x) or noncancer (+) ~~risks to~~ health effects. Cancer and noncancer ~~health risks~~toxicity are normalized for purposes of display such that the species with the maximum ~~relative risk~~value in each category is 3. Health risks are only shown for CRACMM species that contain non-zero ~~amounts~~ emissions of HAPs. This data is available in the supplementary archive as Table D3.


(a)
(b)
HCHO
NAPH
BDE13
BEN
ACD
TOL
MOH
ETEG
ACRO
XYM
XYE
SLOWROC
ALD
PHEN
HC5
KET
CSL
ROCP5ARO
ROCP6ARO
OLT
ROCIOXY
ROH
FURAN
OLI
HC10
HC3
ROCP2ALK
ROCP1ALK
ROCP0ALK
ROCP4ALK
ETH
EOH
ROCP3ALK
ROCN2ALK
ACT
ETE
ORA2
ROCN1ALK
ROCP6ALK
ROCP5ALK
ACE
LIM
API
GLY
PROG
MEK
MACR
BALD
ORA1
MGLY
MVK
ISO
SESQ
UALD
HKET
MCT
DCB1
HAP
Relative Toxicity
Cancer Toxicity
Noncancer Toxicity
0
1
0
1
2
3
Fraction HAP Emissions [Tg/yr], Relative Toxicity

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