The authors presented a comprehensive evaluation of an updated and expanded EMAC model. The implementation of explicit mechanisms and the gas-particle condensation module to the chemistry GCM can be very useful for studying organic vapors and their chemistry in the atmosphere. The model evaluation was made on OH, CO, some key VOCs, AOD and the chemical components of PM2.5. Significant biases present in OVOC simulations, whilst the model performance for aerosol composition is similar to that of previous model configurations. The reasons that lead to the model biases were explored but I feel there are still somewhat insufficient which may be improved with more discussions. Overall I think this is a relevant, important paper and recommend it to be accepted after addressing the following comments.

Specific comments:

(1) Line 81: Are those emissions reasonable compared with other global model inputs or estimates? It is better to add a few sentences to discuss it. It looks like the work did not apply regional emission inventories (e.g., NEI for US, EMEP for Europe, MIX for East Asia or MEIC for China). Would this affect the simulations in anthropogenic-influenced regions? In particular, for aerosol composition, I think the regional inventories may matter significantly.

(2) Section 3.2.5: I think the authors only used the EANET data for 2010. Please specify.

(3) Line 238-239: What kind of biogenic CO precursors?

(4) Line 280-281: How high? I am surprised that temperature difference can be significant between the model parameters and the observations. If the overestimation is due to excess of biogenic VOCs, it is better to compare the surface concentrations of isoprene etc. Some measurements are available in those tropical areas and the data are published (e.g., from AMAZE-08).

(5) Consistent model underestimation present in mid- and high latitudes of Northern Hemisphere for CH3OH, CH3COCH3, HCOOH, CH3COOH, and PAN. The authors made
some explanations. For example, Biogenic or biomass burning VOCs emissions might be
too low during summertime in those regions. In-cloud chemistry may happen. In what
regions, such in-cloud chemistry can be a significant source (e.g., HCOOH)? This model
bias is too consistent, which made me wonder how this is related to underrepresented
emission inventories of VOCs in EDGAR (e.g., contributed by outflow of polluted areas). If
biomass burning is not well represented, what does the model-satellite comparison look
like for different seasons? Or if it is biogenic, would the simulation become better in
winter? I think the current discussion about the potential bias is a bit too general.

(6) Line 389-390: What are those concentrations? The magnitudes seem not matching
with annual mean surface concentrations of OH.

(7) Section 4.3: What about the model performance of surface OH in different regions?
This would affect VOC chemistry and SOA formation significantly. Surface OH
concentrations can be quite high in polluted areas (e.g., in China). A recent paper pointed
the model underestimation of OH because of the lack of HONO source [Miao et al., 2021].
Would that be a problem in the updated model presented here?

(8) Section 4.5: Is the model performance lower in winter than in summer? Residential
combustion in China seems to be a big source of PM2.5 associated with large uncertainty
in the inventory. Would that affect the comparison in Asia?

(9) Line 470-473: What do the model values in higher vertical layers compare with the
observations?

(10) Line 487-488: Nitrate overestimation seems a general problem in chemical transport
models. A main reason could be the inappropriate deposition parameters [Miao et al.,
2020; Zhai et al., 2021]. But more pronounced bias in winter than in summer is
inconsistent with the other studies.

(11) Section 4.6.5: It would be good to make some connections for model biases on
species. For example, how would the significant biases for the oxygenated organics
discussed in the paper affect the OA simulations? So does OH as I mentioned in #7.

(12) Line 553- 562: The observations in Asia used here for aerosol composition are quite
limited. Studies have suggested the emission inventories for semivolatile and intermediate
volatility organic compounds are insufficient and lead to the majority of the model biases
in simulating OA [Miao et al., 2021; Zhao et al., 2016] in regions that are influenced
significantly by anthropogenic emissions. In Europe, residential wood burning also
contributes a lot to the primary OA in winter. Would that be a possible bias? For the rural
areas, fragmentation of OA is not included in the model, which may contribute to the
overestimation over North America. OA includes both primary OA and SOA. It might be
more clear if the two cases are separated discussed.

Technical remarks:

Line 15: Update “Volatile Organic Compounds” to be “Volatile organic compounds”.

Line 29: Please provide the species name for “HNCO”.

Line 217: C3H6 instead of C4H6?

Definition of abbreviations such as MM, OM, MSTD and OSTD etc. are repeated for
multiple times in the paper (e.g., page 25-26).
Please be consistent when using the term of “column densities”. In many places, “column” is used to stand for column densities or column concentrations (Line 238, 274, and so on).

Line 254: Remove “tends to”.

Line 266-268: Revise what are in the parentheses.

Line 389: "hydroxyl" means "OH"? Also in Line 396, to be consistent, OH should be used instead of “hydroxyl radical”.

Line 466-467: This sentence needs to be revised. Are model values low or observations low?

Figure 16: Colors (light vs dark) are not clear.

Line 543: Do you mean “Table 5” instead of “Fig. 5”?

References:


