In this paper, Gao et al. updated the MATRIX aerosol microphysical scheme to include the volatility basis set (VBS) framework for simulating the organic aerosol formation and growth. The manuscript is of interest to the GMD audience and the scientific methodology used sounds valid. Furthermore, the presented module has the potential to be a great addition to the NASA GISS ModelE ESM. However, I would recommend to describe the new scheme in more detail prior to publication.

Specific comments:

1. Page 3 lines 21-25: This paragraph needs to be moved to the model description

2. Section 2.1: I suggest to describe more in detail here the aerosol populations used by the MATRIX model.

3. Section 2.2: I suggest to improve the description of the VBS framework used. Important information that affect the presented results are missing here: For instance, the description of the ageing mechanism (i.e., the oxidation rate constants used, the reduction in volatility after each oxidation step, and the oxygen mass added per oxidation). Furthermore, how do you describe the formation of SOA from traditional VOCs in the model? Do you use a narrower volatility distribution (e.g., up to 10^3)? Do you use aerosol yields for the oxidation of VOCs (if so please report them)? Another issue is how do you perform the partition between the two phases. Do you assume instant equilibrium? Do you account for the temperature dependence of saturation concentrations? If so, what are the enthalpies of vaporization used for each of your organic species? Finally, since you use terms such as "high volatility range", "intermediate volatility range", and "low volatility range" in your results I would recommend to define these terms here in respect to the effective saturation concentration.

4. Page 5, lines 1-4: Which is the extra population compared to the "14 populations" configuration? Furthermore, according to Table 2, the aerosol populations ACC, BC1, BC2, and BCS do not contain organics. Please explain what do you mean that these populations "are set to contain organics as semi-volatile VBS species".

5. Page 6 lines 1-2: Why the overestimation of biomass burning emission factors is not an issue for your experiments? Your experiments include forested areas during summer (where you have high emissions from open biomass burning) and highly populated areas during winter (where you have high biomass burning emissions from residential heating).

6. Page 7 lines 1-12: Most of this discussion belongs to the model description.

7. Page 11 line 30: You did not only include the volatility of the organics, but also their reactivity (by ageing with OH).

8. Table 2: What is the size of the mode (Aitken, accumulation, or coarse) used for the aerosol populations OCC, BC1, BC2, BCS, BOC, OCS, and MXX? Also, does the OCC has sulfate? Because in that case it seems to be identical to the OCS.

9. Figure 2: "P2" and "OCAR" seem to have very similar color. Please change the color of "P2". Furthermore, it would be nice to add for comparison the "OCAR" dashed line in the "Aerosol phase" column as well.

10. Figure 4: The label is wrong. It writes that the second column is "July new" and the third for "January old" which is not the case.