In this study, the authors document the development effort for GOCART-2G model and evaluate the benchmark simulation against multiple observations. Overall, the manuscript is in good shape. I have a few comments for the authors to consider.

General Comments:

1. I would suggest that the authors show the changes or improvements of aerosol related fields, such as AOD, aerosol mass budgets, and aerosol mass concentrations, between GOCART-2G and previous GOCART version. Although the authors provide the documentation of changes in parameterizations and code refactoring, there is no information regarding how those changes impact actual fields (AOD, aerosol mass budgets, aerosol mass concentrations, etc). This is especially important for readers outside of the GEOS-GOCART community.

2. I think more effort is needed for the analysis of aerosol mass budget (i.e., section 4.1). It would be more helpful to show global mean total column mass, or burden in other words, and aerosol lifetime. I would suggest the authors compare them with previous GOCART and results from AeroCom III (e.g., Gliß et al., 2021) and other recent studies (e.g., dust from Kok et al., 2021). I think tables or bar plots showing the global mean statistics (e.g., burden, emission, deposition, chemistry production) are more helpful than the time series plots (Figure 2) and just showing the annual mean spatial distributions of emission and deposition from GOCART-2G (Figures 1 and 3).

3. It seems that the authors have focused on comparisons of AOD with MODIS and AERONET. However, I think some key metrics are either not shown or spread in many individual plots. I would suggest the authors improve the ways they present the results and add more discussions. First, tables showing statistics of global annual mean land, ocean, total AOD from GOCART-2G, previous version of GOCART, MODIS (Aqua and Tera), and MISR would be helpful. Figures 8-11 give limited information, most of which is similar to Figure 6. I would suggest the authors to combine them and show the key results. For example, just show panels (a) and (c) with statistics. The discussion of low AOD biases over Europe may need more effort. The comparison of surface $PM_{2.5}$ shows high biases for GOCART-2G. There is a low bias of attenuated backscatter between 1 km to 5 km over CONUS region. I would suggest the authors look at other seasons and annual mean as well.

Specific comments:

Lines 14-15, should be “the sources, sinks, and chemistry within”.

Line 18, it is not clear to me. Do you mean “so that multiple instances of an aerosol …” or “such that …”

Line 138, could you give the full name for QFED when it is first referred to?

Line 151, should be “biogenic VOCs”. Isoprene, monoterpene, and other terpenes are gas species.

Lines 158-159, I’m a bit confused here. Is OCS a prescribed species, a species with fixed concentration, or an active tracer in the model?

Lines 165-167, why the mechanism will not be used for typical model simulations.

Could you give any statistics to show the computational performance of GOCART-2G compared with previous version?

Do the authors perform any data sampling related to orbital space and time for comparisons with MODIS and CALIOP?

Line 275, is emissions for BC, OA, and sulfate from shipping included? I did not see carbonaceous emissions over ocean. If it is not included, what is the reason?

Figure 12 gives limited information and can be either put in supplement or combined with other plots.

Figure 14, I would suggest showing aerosol extinction. Why only show JJA results? What about annual mean and other seasons?

Reference:

Gliß, J., Mortier, A., Schulz, M., Andrews, E., Balkanski, Y., Bauer, S. E., Benedictow, A. M. K., Bian, H., Checa-Garcia, R., Chin, M., Ginoux, P., Griesfeller, J. J., Heckel, A., Kipling, Z., Kirkevåg, A., Kokkola, H., Laj, P., Le Sager, P., Lund, M. T., Lund Myhre, C., Matsui, H., Myhre, G., Neubauer, D., van Noije, T., North, P., Olivié, D. J. L., Rémy, S., Sogacheva, L., Takemura, T., Tsigaridis, K., and Tsyro, S. G.: AeroCom phase III multi-model evaluation of the aerosol life cycle and optical properties using ground- and space-based remote sensing as well as surface in situ observations, Atmos. Chem. Phys., 21, 87–128, https://doi.org/10.5194/acp-21-87-2021, 2021.

Kok, J. F., Adebiyi, A. A., Albani, S., Balkanski, Y., Checa-Garcia, R., Chin, M., Colarco, P. R., Hamilton, D. S., Huang, Y., Ito, A., Klose, M., Leung, D. M., Li, L., Mahowald, N. M., Miller, R. L., Obiso, V., Pérez García-Pando, C., Rocha-Lima, A., Wan, J. S., and Whicker, C. A.: Improved representation of the global dust cycle using observational constraints on dust properties and abundance, Atmos. Chem. Phys., 21, 8127–8167,

https://doi.org/10.5194/acp-21-8127-2021, 2021.