This manuscript describes the GCAP 2.0 model framework, in which the GEOS-Chem 3D CTM is driven by version E2.1 of the NASA GISS model to simulate past, present, and future atmospheric compositions. The work is built on previous versions of offline coupling between the GISS model and the GEOS-Chem model. This manuscript first explains how the meteorological fields were generated from the GISS E2.1 model, the boundary conditions and emissions taken from latest CMIP6-relevant datasets, and how these data drive the GEOS-Chem model for different time periods. The manuscript then extensively evaluate the E2.1 meteorological dataset against a reanalysis product (MERRA-2), the E2.1 product submitted to CMIP6, and an intermediate product in which E2.1 winds were nudged to MERRA-2 winds. The resulting differences in GCAP 2.0-simulated atmospheric composition and chemical diagnostics are also evaluated against observations.

In my view, this paper is extremely well written. It is particularly commendable in its extensive evaluation of the meteorological data sets and chemical diagnostics. I only have a few minor comments. In all, I think this is a great guide for researchers interested in using GCAP 2.0, as well as a great reference for those who are interested in understanding the results from global CTMs. I recommend publication of this manuscript after minor revisions.

Line 18: "... for the recent past...": This reads awkward after the last sentence of the previous paragraph, which is (largely) not an issue of the recent past.

Lines 131-132: This sentence is confusing for readers unfamiliar with GEOS or MERRA-2. Is MERRA-2 a product of GEOS-DAS?
Section 5.1.1, lines 259-264: I am confused by this paragraph. So, does GCAP 2.0 use only CEDS, or does it also use a hybrid emission inventory with CEDS superseded by regional inventories?

Line 438: "There is a slight overestimate within the boundary layer and underestimate above ...": overestimate and underestimate of what? Are the authors referring to 222Rn concentration or its vertical gradient? Please revise to improve clarity.

Figure 18: What is the time period examined in this figure?