

We sincerely thank the editor for the constructive and insightful comments, which have greatly helped us improve the clarity and quality of our manuscript. We have provided point-by-point responses to each comment. The editor's comments are shown in black, and our responses in blue. All revisions to the manuscript are in red. The corresponding changes have been incorporated into the updated manuscript.

Please incorporate comments 1-7 and 2-7 into your manuscript. After doing so, it will be ready for publication.

**Response:**

We thank the editor for the comment. We have incorporated comments 1-7 and 2-7 into the manuscript:

Lines 385-390: “It was also found that wind speeds were overestimated at GZ (Fig. S14), which could lead to excessive dilution and consequently reduce the simulated concentrations of locally emitted POA and freshly formed SOA (both of which typically exhibit lower O/C ratios than aged POA or SOA). This effect would be expected to increase the simulated O/C. However, the low biases in O/C at GZ indicate that meteorological biases are unlikely to be the dominant factor, and that limitations in the SOA representations may play a more important role.”

Lines 405-409: “The observations revealed significant differences in the O/C ratios between DY and GZ, reflecting distinct aging processes associated with site characteristics (i.e., regional transport at DY versus dominant local emissions at GZ), rather than differences in oxidant levels, as comparable O<sub>3</sub> concentrations were observed at both sites. As discussed above, current SOA parameterizations inadequately represent chemical aging processes and thereby fail to reproduce the observed spatial contrasts in oxidation state.”

Lines 553-567: “Therefore, we summarize several recommendations based on this study for future improvements in OA modeling:

- (1) Given the substantial contribution of L/S/IVOCs to SOA, emission inventories should be further refined not only in terms of total magnitudes but also volatility-resolved distributions, and constrained using ambient measurements. In addition, primary emissions from sources such as cooking, open biomass burning, and mobile sources require improved representation. Better treatment of nighttime SOA formation pathways, particularly NO<sub>3</sub> oxidation and aqueous-phase chemistry, is also needed to reduce SOA mass underestimation.

- (2) While updated SOA yields can partially improve model performance, explicitly accounting for autoxidation processes and the formation of HOMs (including both biogenic and anthropogenic origins) would provide a more physically based description of O/C evolution. Moreover, SOA aging schemes should be better constrained by chamber experiments, particularly with respect to the relationship between the degree of oxygenation and multigenerational aging. Constraining POA O/C ratios using source-specific measurements also represents a promising approach for improving the modeled elemental composition of OA.
- (3) The linkage between OA volatility and  $T_{g,org}$  (and viscosity) requires revisiting the volatility assignments of existing SOA surrogates (e.g., isoprene-derived SOA in CMAQ) and developing more accurate, dynamic parameterizations of  $\kappa_{org}$ .”