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Supplement of

Analysis of secondary organic aerosol simulation bias in the Community Earth System Model (CESM2.1)

Yaman Liu et al.

Correspondence to: Xinyi Dong (dongxy@nju.edu.cn)

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Table S1. 15 reactions relative to SOAG production.

| Reactants      | Products                                                                 | Rate Constant                |
|----------------|--------------------------------------------------------------------------|------------------------------|
| 1 ISOP + OH    | ISOP + OH + 0.0031*SOAG0 + 0.0035*SOAG1 + 0.0003*SOAG2 + 0.0271*SOAG3 + 0.0474*SOAG4 | 2.54e-11 exp(410/T)         |
| 2 ISOP + O3    | ISOP + O3 + 0.0033*SOAG3                                                | 1.05e-13 exp(-2000/T)       |
| 3 ISOP + NO3   | ISOP + NO3 + 0.059024*SOAG3 + 0.025024*SOAG4                            | 3.03e-12 exp(-446/T)        |
| 4 MTERP + NO3  | MTERP + NO3 + 0.17493*SOAG3 + 0.59019*SOAG4                             | 1.20e-12 exp(490/T)        |
| 5 MTERP + O3   | MTERP + O3 + 0.0508*SOAG0 + 0.1149*SOAG1 + 0.0348*SOAG2 + 0.0554*SOAG3 + 0.1278*SOAG4 | 6.30e-16 exp(-580/T)       |
| 6 MTERP + OH   | MTERP + OH + 0.0508*SOAG0 + 0.1149*SOAG1 + 0.0348*SOAG2 + 0.0554*SOAG3 + 0.1278*SOAG4 | 1.20e-11 exp(440/T)        |
| 7 BCARY + NO3  | BCARY + NO3 + 0.17493*SOAG3 + 0.59019*SOAG4                             | 1.900e-11                   |
| 8 BCARY + O3   | BCARY + O3 + 0.2202*SOAG0 + 0.2067*SOAG1 + 0.0653*SOAG2 + 0.1284*SOAG3 + 0.114*SOAG4 | 1.20e-14                   |
| 9 BCARY + OH   | BCARY + OH + 0.2202*SOAG0 + 0.2067*SOAG1 + 0.0653*SOAG2 + 0.1284*SOAG3 + 0.114*SOAG4 | 2.00e-10                   |
| 10 BENZENE + OH| BENZENE + OH + 0.0023*SOAG0 + 0.0008*SOAG1 + 0.0843*SOAG2 + 0.0443*SOAG3 + 0.1621*SOAG4 | 2.30e-12 exp(-193/T)       |
| 11 TOLUENE + OH| TOLUENE + OH + 0.1364*SOAG0 + 0.0101*SOAG1 + 0.0763*SOAG2 + 0.2157*SOAG3 + 0.0232*SOAG4 | 1.70e-12 exp(352/T)        |
| 12 XYLENES + OH| XYLENES + OH + 0.1677*SOAG0 + 0.0174*SOAG1 + 0.086*SOAG2 + 0.0512*SOAG3 + 0.1598*SOAG4 | 1.700e-11                  |
| 13 IVOC + OH   | OH + 0.2381*SOAG0 + 0.1308*SOAG1 + 0.0348*SOAG2 + 0.0076*SOAG3 + 0.0113*SOAG4 | 1.340e-11                  |
| 14 SVOC + OH   | OH + 0.5931*SOAG0 + 0.1534*SOAG1 + 0.0459*SOAG2 + 0.0085*SOAG3 + 0.0128*SOAG4 | 1.340e-11                  |
| 15 GLYOXAL + aer| SOAG0                                                                   | f(SAD), γ=0.0002b          |

a Emmons et al. (2020)

b Function of aerosol surface area density (SAD), see Emmons et al. (2020) for details
Table S2. Chemical formula and description of species in Table S1 (Emmons et al., 2020).

| Species | Chemical Formula | Description |
|---------|------------------|-------------|
| ISOP    | C5H8             | isoprene    |
| MTERP   | C10H16           | lumped monoterpenes |
| BCARY   | C15H24           | beta-caryophyllene and other sesquiterpenes |
| BENZENE | C6H6             | benzene     |
| TOLUENE | C7H8             | toluene     |
| XYLENES | C8H10            | lumped xylenes |
| IVOC    | C13H28           | intermediate volatility organic precursor of VBS SOA |
| SVOC    | C22H46           | semi-volatile organic precursor of VBS SOA |
| GLYOXAL | C2H2O2           | glyoxal     |
| OH      | OH               | hydroxyl radical |
| O3      | O3               | ozone       |
| NO3     | NO3              | nitrate radical |
| SOAG0   | C15H38O2         | SOA gas-phase precursor VBS bin 0 (mol.wt. = 250 g/mol) (Shrivastava et al., 2015) |
| SOAG1   | C15H38O2         | SOA gas-phase precursor VBS bin 1 |
| SOAG2   | C15H38O2         | SOA gas-phase precursor VBS bin 2 |
| SOAG3   | C15H38O2         | SOA gas-phase precursor VBS bin 3 |
| SOAG4   | C15H38O2         | SOA gas-phase precursor VBS bin 4 |
Table S3. The average OA of four ATom campaigns (Wofsy et al., 2018) and CAM-Chem-SD; the mean bias (MB), normalized mean bias (NMB), normalized mean error (NME), root mean square error (RMSE) and correlation coefficient (CC) between campaigns and CAM-Chem-SD.

| Aircraft | Mean Obs. | Mean Sim. | MB   | NMB(%) | NME(%) | RMSE | CC  |
|----------|-----------|-----------|------|--------|--------|------|-----|
| ATom1    | 0.38      | 0.26      | -0.12| -29.6  | 64.1   | 0.70 | 0.72|
| ATom2    | 0.16      | 0.10      | -0.06| -39.6  | 76.8   | 0.26 | 0.58|
| ATom3    | 0.28      | 0.13      | -0.15| -54.7  | 76.5   | 0.61 | 0.40|
| ATom4    | 0.38      | 0.11      | -0.27| -70.8  | 81.2   | 0.90 | 0.33|
Figure. S1. Seasonal cycle of 1988–2019 average surface Primary Organic Aerosol (POA) anthropogenic emission used for CAM-Chem-SD (red upper triangles) over CONUS (a), EUS (b) and WUS (c).
Figure. S2. Validation against ATom flight. (a) trajectories of four ATom flights; subdomains divisions are shown in grey boxes. Vertical profile of organics concentration of CAM-Chem-SD (red lines) and flight measurements (black marker lines) during ATom1 (b), ATom2 (c), ATom3 (d) and ATom4 (e) campaigns.
Figure. S3. Vertical profile of SOA formed by 15 reactions over US in July, 2010. The vertical average relative contribution of each reaction is shown as the number in the legend.
Figure S4. The relationship between surface OA bias and MTERP (panel a, d, g), ISOP (panel b, e, h), TOLUENE (panel c, f, i) flux in the summer of 1988 to 2019 over CONUS (panel a ~ c), EUS (panel d ~ f) and WUS (panel g ~ i). Surface VOCs flux are split into biogenic emission flux from MEGAN (light red dots) and other flux referred as anthropogenic flux (dark red dots). The numbers shown in each panel are the correlation coefficient between OA bias and biogenic flux, followed by the correlation coefficient between OA bias and anthropogenic flux.
Figure S5. 2013.03–2014.02 CONUS surface OA concentration of IMPROVE (blue dots), CAM-Chem-SD with MOZART-TSI chemistry (red dots) and CAM-Chem-SD with MOZART-TS2 chemistry (green dots). Every blue box denotes the 10th, the 25th, the median, the 75th and the 90th percentiles of the observations for all selected sites in each month.
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