Supplement of

Modeling the smoky troposphere of the southeast Atlantic: a comparison to ORACLES airborne observations from September of 2016

Yohei Shinozuka et al.

Correspondence to: Yohei Shinozuka (yohei.shinozuka@nasa.gov) and Paquita Zuidema (pzuidema@miami.edu)

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Table S1. Comparison of flight-day values to the monthly-mean climatology formulated from the same model. Shown are the mean bias (MB), and root-mean-square deviation (RMSD), as well as their ratio (%) to the monthly mean.

|                          | WRF-CAM5 | GEOS-5 |
|--------------------------|----------|--------|
|                          | MB       | RMSD   | MB   | RMSD   |

*Smoke Top Height (m) simulated as observed by HSRL-2 on ER2*

|                          | 500      | 505    | 117  | 122    |
|--------------------------|----------|--------|------|--------|
| MB                       | +125     | (+3%)  | +369 | (+11%) |
| RMSD                     | (11%)    | (15%)  |      |        |

*Smoke Base Height (m) simulated as observed by HSRL-2 on ER2*

|                          | 426      | 292    | 424  | 319    |
|--------------------------|----------|--------|------|--------|
| MB                       | +371     | (+29%) | +103 | (+8%)  |
| RMSD                     | (33%)    | (23%)  |      |        |

*Black Carbon Mass (ng m⁻³)*

|                          | 182.5    | 198.2  | 182.5| 198.2  |
|--------------------------|----------|--------|------|--------|
| 3-6 km                   | -4.5     | (-1%)  | -1.8 | (-0%)  |
|                          | (27%)    | (30%)  |      |        |
| FT≤ 3km                  | +161.1   | (50%)  | -98.8| (-9%)  |
|                          | (25%)    | (38%)  |      |        |
| MBL                      | +40.1    | (63%)  | +82.1| (+21%) |
|                          | (+31%)   | (93%)  |      |        |

*Organic Aerosol Mass (µg m⁻³)*

|                          | 1.5      | 2.9    | 1.5  | 2.9    |
|--------------------------|----------|--------|------|--------|
| 3-6 km                   | -0.0     | (-0%)  | +0.0 | (+0%)  |
|                          | (27%)    | (32%)  |      |        |
| FT≤ 3km                  | +1.3     | (50%)  | -1.6 | (-10%) |
|                          | (25%)    | (40%)  |      |        |
### Sulfate Aerosol Mass (ug m⁻³)

|                | MBL       | 3-6 km | FT ≤ 3 km | MBL       |
|----------------|-----------|--------|-----------|-----------|
|                | +0.3      | 0.2    | --        | +0.1      |
|                | (+36%)    | (+19%) | --        | (+13%)    |
|                | 0.6       | (+70%) | --        | 0.3       |
|                | (+19%)    | (+44%) | --        | (+44%)    |

### Volumetric Mean Diameter (nm)

|                | 3-6 km    | FT ≤ 3 km | MBL       |
|----------------|-----------|-----------|-----------|
|                | -7        | 10        | --        |
|                | (-3%)     | (+4%)     | --        |
|                | 10        | (+4%)     | --        |
|                | (+4%)     | (+4%)     | --        |

### Aerosol Optical Depth simulated as observed by HSRL-2 on ER2

|                | Above clouds | FT ≤ 3 km | MBL       |
|----------------|--------------|-----------|-----------|
|                | +0.018       | +0.019    | +0.001    |
|                | (+7%)        | (+1%)     | (+1%)     |
|                | 0.055        | 0.057     | 0.036     |
|                | (21%)        | (26%)     | (16%)     |

### Aerosol Optical Depth simulated as observed by 4STAR on P3

|                | Above clouds | FT ≤ 3 km | MBL       |
|----------------|--------------|-----------|-----------|
|                | +0.031       | -0.019    | -0.019    |
|                | (+12%)       | (+9%)     | (+9%)     |
|                | 0.048        | 0.057     | 0.057     |
|                | (18%)        | (26%)     | (26%)     |

### Extinction Coefficient (Mm⁻¹) simulated as observed by HSRL-2 on ER2

|                | 3-6 km      |
|----------------|-------------|
|                | +3          | +2         | +2         |
|                | (+7%)       | (+5%)      | (+5%)      |
|                | 12          | 12         | 12         |
|                | (23%)       | (30%)      | (30%)      |
**Extinction Coefficient (Mm⁻¹) simulated as observed by neph+PSAP on P3**

|          | 3-6 km | FT ≤ 3 km | MBL  |
|----------|--------|-----------|------|
| Extinction Coefficient | -1.0  | 13.0      | -1.0 |
|             | (-3%)  | (25%)     | (-3%)|
|             |        | (-3%)     | (33%)|
| FT ≤ 3 km  | +11.0  | 22.0      | -8.0 |
|             | (+22%) | (42%)     | (-13%)|
|             | (22%)  | (33%)     | (39%)|
| MBL        | -2.0   | 10.0      | +7.0 |
|             | (-6%)  | (32%)     | (+6%)|
|             |        | (2%)      | (62%)|

**Scattering Ångström Exponent**

|          | 3-6 km | FT ≤ 3 km | MBL  |
|----------|--------|-----------|------|
|          | +0.1   | 0.1       | +0.0 |
|          | (+5%)  | (6%)      | (0%) |
|          | (+6%)  | (2%)      | (2%) |
| FT ≤ 3 km| +0.0   | 0.1       | +0.0 |
|          | (+4%)  | (12%)     | (+2%)|
|          | (+4%)  | (12%)     | (6%) |
| MBL      | +0.1   | 0.2       | +0.1 |
|          | (+29%) | (44%)     | (+10%)|
|          | (+29%) | (44%)     | (30%)|

**Absorption Ångström Exponent**

|          | 3-6 km | FT ≤ 3 km | MBL  |
|----------|--------|-----------|------|
|          | +0.0   | 0.0       | +0.0 |
|          | (+1%)  | (1%)      | (0%) |
|          | (+1%)  | (1%)      | (0%) |
| FT ≤ 3 km| +0.0   | 0.0       | -0.0 |
|          | (+0%)  | (2%)      | (-1%)|
|          | (+0%)  | (2%)      | (-1%)|
| MBL      | +0.0   | 0.1       | -0.0 |
|          | (+1%)  | (5%)      | (-2%)|
|          | (+1%)  | (5%)      | (2%) |

**Single Scattering Albedo**

|          | 3-6 km | FT ≤ 3 km | MBL  |
|----------|--------|-----------|------|
|          | -0.00  | 0.01      | -0.00|
|          | (-0%)  | (1%)      | (-0%)|
|          | (-0%)  | (1%)      | (1%) |
| FT ≤ 3 km| -0.01  | 0.01      | -0.00|
|          | (-1%)  | (2%)      | (-0%)|
|          | (-1%)  | (2%)      | (1%) |
| MBL    | -0.02 | 0.03 | -0.01 | 0.01 |
|--------|-------|------|-------|------|
|        | (-2%) | (3%) | (-1%) | (1%) |

*Carbon Monoxide (ppbv)*

|          | 3-6 km | FT≤ 3 km | MBL      |
|----------|--------|----------|----------|
|          | +0     | 23       | +0       |
|          | (+0%)  | (15%)    | (+0%)    |
|          |        | (10%)    | (13%)    |
|          | +12    | 29       | -2       |
|          | (+10%) | (23%)    | (-2%)    |
|          |        | (23%)    | (16%)    |
|          | +3     | 5        | +1       |
|          | (+5%)  | (7%)     | (+2%)    |
|          |        |          | (15%)    |

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2 The optical properties are at 500-550 nm. The values are for the P3 flights unless otherwise noted, in the diagonally and horizontally aligned boxes.
Table S2. The differences of box-average model values from the observations. Shown are the mean bias (MB), and root-mean-square deviation (RMSD), as well as their ratio (%) to the observed mean.

|                      | WRF-CAM5 | GEOS-5  | GEOS-Chem | EAM-E3SM | UM       | ALADIN-Climate |
|----------------------|----------|---------|-----------|----------|----------|----------------|
|                      | MB       | RMSD    | MB        | RMSD     | MB       | RMSD           | MB     | RMSD    | MB     | RMSD    |
| Smoke Top Height (m) |          |         |           |          |          |                |        |         |        |         |
| compared to HSRL-2 on ER2 |         |         |           |          |          |                |        |         |        |         |
| FT                   | -167     | 415     | -456      | 596      | -473     | 763            | -114   | 460     | +6     | 440     | -176   | 830   |
| ≤3km                 | (-3%)    | (9%)    | (-9%)     | (12%)    | (-10%)   | (16%)          | (-2%)  | (10%)   | (+0%)  | (9%)    | (-4%)  | (17%) |
| MBL                  | -422     | 553     | -1401     | 1424     | -877     | 938            | -688   | 784     | -616   | 709     | -299   | 566   |
|                      | (-21%)   | (27%)   | (-69%)    | (70%)    | (-43%)   | (46%)          | (-34%) | (38%)   | (-31%) | (35%)   | (-15%) | (28%) |
| Black Carbon Mass (ng m⁻³) |          |         |           |          |          |                |        |         |        |         |        |       |
| 3-6 km               | +62.2    | 172.6   | +49.2     | 206.8    | +9.7     | 282.5          | -254.8 | 285.9   | -232.8 | 277.6   | --     | --    |
|                      | (+10%)   | (28%)   | (+8%)     | (34%)    | (+2%)    | (47%)          | (-42%) | (47%)   | (-38%) | (46%)   | --     | --    |
| FT                   | -11.7    | 459.3   | +171.0    | 524.6    | +7.3     | 399.6          | -515.9 | 647.3   | -131.3 | 304.6   | --     | --    |
| ≤3km                 | (-1%)    | (57%)   | (+20%)    | (62%)    | (+1%)    | (47%)          | (-81%) | (76%)   | (-16%) | (36%)   | --     | --    |
| MBL                  | -5.1     | 119.9   | +291.5    | 553.7    | +82.6    | 238.0          | +4.7   | 98.6    | -45.5  | 92.6    | --     | --    |
|                      | (-3%)    | (69%)   | (+188%)   | (319%)   | (+48%)   | (137%)         | (+2%)  | (53%)   | (-26%) | (53%)   | --     | --    |
| Organic Aerosol Mass (ug m⁻³) |          |         |           |          |          |                |        |         |        |         |
| 3-6 km               | +0.0     | 2.4     | +3.5      | 5.0      | +1.8     | 4.0            | +5.3   | 5.7     | -1.9   | 2.9     | --     | --    |
|                      | (+0%)    | (42%)   | (+62%)    | (89%)    | (+32%)   | (71%)          | (+94%) | (102%)  | (-34%) | (52%)   | --     | --    |
| FT                   | +0.7     | 3.1     | +7.7      | 9.2      | +3.7     | 5.4            | +2.9   | 4.1     | +0.6   | 2.9     | --     | --    |
| ≤3km                 | (+12%)   | (53%)   | (+119%)   | (141%)   | (+57%)   | (84%)          | (+44%) | (63%)   | (+9%)  | (45%)   | --     | --    |
| MBL                  | +0.3     | 0.8     | +5.4      | 8.9      | +2.1     | 3.9            | +3.7   | 5.1     | +0.3   | 0.9     | --     | --    |
|                      | (+26%)   | (83%)   | (+545%)   | (900%)   | (+210%)  | (392%)         | (+352%)| (493%)  | (+27%) | (96%)   | --     | --    |
| Sulfate Aerosol Mass (ug m⁻³) |          |         |           |          |          |                |        |         |        |         |
| 3-6 km               | +0.5     | 0.6     | --        | --       | --       | --             | +0.2   | 0.3     | -0.4   | 0.6     | --     | --    |
|                      | (+66%)   | (79%)   | --        | --       | --       | --             | (+21%) | (43%)   | (-56%) | (74%)   | --     | --    |
| FT                   | +0.4     | 0.7     | --        | --       | --       | --             | +0.1   | 0.5     | -0.7   | 0.9     | --     | --    |
| ≤3km                 | (+37%)   | (55%)   | --        | --       | --       | --             | (+6%)  | (39%)   | (-56%) | (72%)   | --     | --    |
### Volumetric Mean Diameter (nm)

|                | MBL | 3-6 km | FT | ≤3 km | MBL |
|----------------|-----|--------|----|-------|-----|
|                | -0.5 | +43    | -80 | +86   | +86 |
| (-38%)         | (60%)| (+21%) | (22%)| (+41%)| (+41%)|
|                | 0.7  | 44     | 83  | 95    | 95  |
| (+12%)         | (+42%)| (57%)  | (+21%)| (28%)| (+17%)|
|                | --   | --     | --  | --    | --  |
|                | --   | --     | --  | --    | --  |
|                | --   | --     | --  | --    | --  |
|                | +1.5 | +28    | +34 | +28   | +28 |
| (121%)         | (+14%)| (14%)  | (26%)| (18%)| (14%)|
|                | 1.5  | 28     | 35  | 27    | 27  |
| (121%)         | (14%)| (18%)  | (32%)| (13%)| (13%)|

### Aerosol Optical Depth compared to HSRL-2 on ER2

#### Above clouds

|                | MBL | RMSD | MB | RMSD | MB | RMSD | MB | RMSD | MB | RMSD | MB | RMSD |
|----------------|-----|------|----|------|----|------|----|------|----|------|----|------|
| Aerosol Optical Depth | -0.042 | 0.077 | -0.101 | 0.123 | +0.138 | 0.189 | +0.069 | 0.093 | 0.053 | 0.087 | -0.108 | 0.125 |
| Above clouds | (-12%) | (23%) | (-30%) | (37%) | (+42%) | (57%) | (+21%) | (28%) | (-16%) | (26%) | (-32%) | (37%) |
| Aerosol Optical Depth | -0.068 | 0.098 | -0.126 | 0.183 | +0.016 | 0.103 | +0.063 | 0.096 | -0.148 | 0.181 | -0.099 | 0.137 |
| Above clouds | (-19%) | (28%) | (-38%) | (56%) | (+5%) | (31%) | (+19%) | (29%) | (-45%) | (55%) | (-30%) | (42%) |

### Extinction Coefficient (Mm\(^{-1}\)) compared to HSRL-2 on ER2

#### 3-6 km

|                | MBL | RMSD | MB | RMSD | MB | RMSD | MB | RMSD | MB | RMSD | MB | RMSD |
|----------------|-----|------|----|------|----|------|----|------|----|------|----|------|
| Extinction Coefficient | -16 | 23 | -28 | 32 | +24 | 33 | +1 | 17 | -43/-19 | 49/23 | -- | -- |
| 3-6 km | (-23%) | (32%) | (-38%) | (44%) | (+33%) | (45%) | (+1%) | (23%) | (-59/-26%) | (66/31%) | -- | -- |

### Extinction Coefficient (Mm\(^{-1}\)) compared to neph+PSAP on P3

#### 3-6 km

|                | MBL | RMSD | MB | RMSD | MB | RMSD | MB | RMSD | MB | RMSD | MB | RMSD |
|----------------|-----|------|----|------|----|------|----|------|----|------|----|------|
| Extinction Coefficient | -10 | 18 | -20 | 23 | +19 | 40 | +6 | 17 | -42/-33 | 46/36 | -- | -- |
| 3-6 km | (-17%) | (31%) | (-33%) | (39%) | (+32%) | (67%) | (+11%) | (28%) | (-71/-56%) | (77/61%) | -- | -- |
| FT | +3 | 38 | -12 | 38 | +24 | 39 | +16 | 28 | -30/-16 | 44/32 | -- | -- |
| ≤3km | (+4%) | (62%) | (-18%) | (58%) | (+36%) | (59%) | (+24%) | (43%) | (-46/-25%) | (67/49%) | -- | -- |

### MBL

|                | MBL | RMSD | MB | RMSD | MB | RMSD | MB | RMSD | MB | RMSD | MB | RMSD |
|----------------|-----|------|----|------|----|------|----|------|----|------|----|------|
| Extinction Coefficient | +2 | 8 | +88 | 125 | +68 | 83 | +115 | 122 | -3/+70 | 12/73 | -- | -- |
| (+6%) | (31%) | (+327%) | (463%) | (+255%) | (310%) | (+406%) | (433%) | (12/+260%) | (44/272%) | -- | -- |

### Scattering Ångström Exponent
|        | 3-6 km | FT ≤3km | MBL     |
|--------|--------|---------|---------|
|        |        |         |         |
| Absorption Ångström Exponent |        |         |         |
| 3-6 km | -0.6   | -0.8    | -0.5    |
|        | 0.6    | 0.8     | 0.5     |
|        | -0.1   | -0.0    | -0.4    |
|        | 0.1    | 0.1     | 0.6     |
|        | -0.0   | +0.1    | -0.2    |
|        | 0.1    | 0.1     | 0.4     |
|        | --     | --      | --      |
|        | +0.0/- | +0.0/-  | -0.1/-0.3 |
|        | 0.0    | 0.0     | 0.4/0.4 |
|        |         |         |         |
| Single Scattering Albedo |        |         |         |
| 3-6 km | -0.03  | -0.00   | -0.02   |
|        | 0.04   | 0.01    | 0.04    |
|        | -0.01  | -0.01   | -0.3    |
|        | 0.02   | 0.02    | 0.6     |
|        | +0.06  | +0.07   | +0.04   |
|        | 0.06   | 0.07    | 0.06    |
|        | +0.02  | +0.06   | +0.03   |
|        | 0.02   | 0.07    | 0.03    |
|        | -0.07/-0.02 | 0.04/+0.05 | 0.03/0.05 |
|        | 0.08/0.0 | 0.05/0.0 | 0.03/0.05 |
|        |         |         |         |
| Carbon Monoxide (ppbv) |        |         |         |
| 3-6 km | -37    | -24     | -21     |
|        | 44     | 43      | 24      |
|        | -19    | -13     | -11     |
|        | 30     | 32      | 21      |
|        | -38    | -8      | -4      |
|        | 45     | 34      | 19      |
|        | --     | --      | --      |
|        | --     | --      | --      |
|        | --     | --      | --      |
|        | --     | --      | --      |
|        | --     | --      | --      |
|        | --     | --      | --      |
|        | --     | --      | --      |
|        | --     | --      | --      |
|        | --     | --      | --      |
The optical properties are at 500-550 nm. The values are for the P3 flights unless otherwise noted, in the diagonally and horizontally aligned boxes. The hyphens indicate products unavailable. For UM the pair of values, where given, correspond to dry and ambient humidity conditions in this order.