

## Supplemental Information

| #1 Great Lakes                       |               |          | Annual |       | Spring |       | Summer |       | Autumn |       | Winter |       |
|--------------------------------------|---------------|----------|--------|-------|--------|-------|--------|-------|--------|-------|--------|-------|
|                                      |               |          | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend |
| TS1.1 surface<br>(sampled)           | 2005-<br>2020 | AQS      | 38.2   | -0.11 | 44.5   | -0.13 | 48.6   | -0.57 | 33.8   | -0.24 | 26.2   | 0.45  |
|                                      |               | a priori | 40.0   | -0.04 | 43.2   | -0.17 | 55.7   | -0.45 | 37.1   | 0.09  | 24.1   | 0.35  |
|                                      |               | KF-AQS   | 37.7   | -0.07 | 43.5   | -0.13 | 49.0   | -0.51 | 33.1   | -0.03 | 25.4   | 0.39  |
| TS1.2<br>surface                     | 2005-<br>2020 | a priori | 40.2   | -0.06 | 43.4   | -0.18 | 55.7   | -0.47 | 37.4   | 0.09  | 24.8   | 0.27  |
|                                      |               | KF-AQS   | 37.9   | -0.09 | 43.5   | -0.14 | 49.1   | -0.52 | 33.5   | -0.04 | 25.7   | 0.30  |
| TS1.3<br>trop. column<br>(convolved) | 2005-<br>2020 | OMI      | 39.3   | -0.09 | 40.6   | -0.16 | 46.9   | 0.12  | 36.7   | -0.17 | 32.8   | -0.15 |
|                                      |               | a priori | 36.9   | -0.11 | 37.5   | -0.16 | 45.9   | -0.13 | 35.0   | -0.09 | 29.1   | -0.03 |
|                                      |               | KF-OMI   | 38.8   | -0.21 | 40.4   | -0.31 | 47.7   | -0.03 | 36.3   | -0.23 | 30.4   | -0.24 |
| TS1.4<br>trop. Column                | 2005-<br>2020 | a priori | 37.4   | -0.03 | 38.1   | -0.12 | 45.9   | -0.13 | 35.8   | 0.03  | 30.1   | 0.12  |
|                                      |               | KF-AQS   | 36.8   | -0.03 | 37.9   | -0.10 | 44.3   | -0.13 | 35.0   | 0.01  | 30.1   | 0.12  |
|                                      |               | KF-OMI   | 40.2   | -0.29 | 42.5   | -0.44 | 48.3   | -0.09 | 37.8   | -0.23 | 32.2   | -0.39 |
|                                      | 2005-<br>2009 | KF-AQS   | 36.6   | -0.48 | 38.0   | -0.32 | 44.1   | -1.57 | 34.9   | -0.37 | 29.3   | -0.10 |
|                                      |               | KF-OMI   | 41.8   | -0.42 | 44.5   | -0.29 | 47.6   | -1.30 | 39.4   | -0.52 | 35.3   | -0.47 |
|                                      | 2010-<br>2014 | KF-AQS   | 37.1   | -0.63 | 38.4   | -1.15 | 45.5   | -1.95 | 34.7   | -0.11 | 30.1   | -0.06 |
|                                      |               | KF-OMI   | 40.2   | -0.97 | 43.1   | -1.45 | 49.6   | -1.77 | 37.0   | -0.41 | 30.7   | -0.49 |
|                                      | 2015-<br>2020 | KF-AQS   | 36.7   | 0.05  | 37.4   | -0.04 | 43.7   | 0.38  | 35.3   | -0.19 | 30.8   | 0.11  |
| KF-OMI                               |               | 38.9     | -0.15  | 40.2  | -0.41  | 47.7  | 0.16   | 37.1  | -0.35  | 30.9  | 0.02   |       |

**Table. S1.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> concentrations in 2005-2020 over Great Lakes (Region #1) from observations (AQS and OMI) and a priori and a posteriori (KF) simulations. Grids with low and medium anthropogenic NO<sub>x</sub> emissions are excluded in Great Lakes.

| #2 Northeast US                      |               |          | Annual |       | Spring |       | Summer |       | Autumn |       | Winter |       |
|--------------------------------------|---------------|----------|--------|-------|--------|-------|--------|-------|--------|-------|--------|-------|
|                                      |               |          | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend |
| TS2.1 surface<br>(sampled)           | 2005-<br>2020 | AQS      | 37.4   | -0.07 | 43.9   | -0.11 | 47.1   | -0.61 | 31.9   | -0.05 | 27.2   | 0.42  |
|                                      |               | a priori | 38.1   | 0.03  | 41.6   | -0.02 | 52.2   | -0.41 | 34.7   | 0.20  | 24.1   | 0.33  |
|                                      |               | KF-AQS   | 36.4   | -0.04 | 42.4   | -0.08 | 46.3   | -0.52 | 31.0   | 0.05  | 26.2   | 0.36  |
| TS2.2<br>surface                     | 2005-<br>2020 | a priori | 38.4   | 0.01  | 41.9   | -0.05 | 52.4   | -0.42 | 34.9   | 0.19  | 24.7   | 0.28  |
|                                      |               | KF-AQS   | 36.6   | -0.06 | 42.5   | -0.11 | 46.4   | -0.53 | 31.2   | 0.03  | 26.5   | 0.32  |
| TS2.3<br>trop. column<br>(convolved) | 2005-<br>2020 | OMI      | 39.3   | -0.12 | 40.8   | -0.21 | 47.3   | 0.02  | 37.2   | -0.21 | 31.9   | -0.08 |
|                                      |               | a priori | 37.0   | -0.11 | 37.1   | -0.18 | 45.8   | -0.13 | 35.3   | -0.11 | 29.6   | 0.00  |
|                                      |               | KF-OMI   | 38.8   | -0.22 | 40.1   | -0.36 | 48.0   | -0.06 | 36.7   | -0.27 | 30.5   | -0.17 |
| TS2.4<br>trop. Column                | 2005-<br>2020 | a priori | 37.4   | -0.02 | 38.0   | -0.07 | 45.3   | -0.17 | 35.9   | 0.04  | 30.5   | 0.14  |
|                                      |               | KF-AQS   | 36.8   | -0.02 | 37.9   | -0.07 | 43.7   | -0.18 | 35.1   | 0.02  | 30.6   | 0.14  |
|                                      |               | KF-OMI   | 40.3   | -0.31 | 42.9   | -0.48 | 47.9   | -0.12 | 37.8   | -0.21 | 32.7   | -0.39 |
|                                      | 2005-<br>2009 | KF-AQS   | 36.6   | -0.33 | 37.7   | 0.00  | 43.9   | -1.21 | 34.9   | -0.67 | 30.0   | -0.07 |
|                                      |               | KF-OMI   | 42.0   | -0.33 | 45.1   | -0.11 | 47.6   | -0.89 | 39.1   | -0.74 | 36.2   | -0.63 |
|                                      | 2010-<br>2014 | KF-AQS   | 37.1   | -0.51 | 38.6   | -0.91 | 44.7   | -1.96 | 35.0   | -0.15 | 30.2   | -0.03 |
|                                      |               | KF-OMI   | 40.4   | -0.86 | 43.9   | -1.16 | 48.9   | -1.92 | 37.3   | -0.42 | 30.6   | -0.46 |
|                                      | 2015-<br>2020 | KF-AQS   | 36.6   | 0.10  | 37.4   | -0.07 | 42.8   | 0.20  | 35.4   | -0.13 | 31.4   | 0.28  |
| KF-OMI                               |               | 38.9     | -0.08  | 40.2  | -0.46  | 47.2  | 0.03   | 37.2  | -0.23  | 31.5  | 0.20   |       |

**Table. S2.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> concentrations in 2005-2020 over Northeast US (Region #2) from observations (AQS and OMI) and a priori and a posteriori (KF) simulations. Grids with low and medium anthropogenic NO<sub>x</sub> emissions are excluded in Northeast US.

| #3 West Coast                        |               |          | Annual |       | Spring |       | Summer |       | Autumn |       | Winter |       |
|--------------------------------------|---------------|----------|--------|-------|--------|-------|--------|-------|--------|-------|--------|-------|
|                                      |               |          | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend |
| TS3.1 surface<br>(sampled)           | 2005-<br>2020 | AQS      | 41.0   | 0.15  | 44.5   | -0.07 | 50.8   | -0.15 | 41.0   | 0.39  | 28.1   | 0.37  |
|                                      |               | a priori | 41.6   | -0.01 | 43.8   | -0.18 | 49.2   | -0.09 | 40.8   | 0.12  | 32.9   | 0.08  |
|                                      |               | KF-AQS   | 41.2   | 0.11  | 44.5   | -0.10 | 50.6   | -0.13 | 41.4   | 0.34  | 28.8   | 0.29  |
| TS3.2<br>surface                     | 2005-<br>2020 | a priori | 41.6   | -0.01 | 43.8   | -0.18 | 49.2   | -0.09 | 40.8   | 0.12  | 32.9   | 0.08  |
|                                      |               | KF-AQS   | 41.2   | 0.11  | 44.5   | -0.10 | 50.6   | -0.13 | 41.4   | 0.34  | 28.8   | 0.29  |
| TS3.3<br>trop. column<br>(convolved) | 2005-<br>2020 | OMI      | 39.2   | 0.00  | 42.1   | 0.21  | 43.8   | 0.13  | 37.9   | -0.15 | 32.7   | -0.19 |
|                                      |               | a priori | 37.5   | -0.09 | 39.5   | -0.06 | 42.7   | -0.12 | 35.6   | -0.09 | 31.9   | -0.10 |
|                                      |               | KF-OMI   | 39.8   | -0.14 | 43.2   | -0.06 | 45.2   | 0.02  | 37.7   | -0.23 | 32.7   | -0.28 |
| TS3.4<br>trop. Column                | 2005-<br>2020 | a priori | 38.2   | -0.02 | 38.9   | -0.09 | 42.5   | -0.15 | 36.5   | 0.00  | 34.9   | 0.07  |
|                                      |               | KF-AQS   | 38.1   | -0.01 | 38.9   | -0.09 | 42.6   | -0.16 | 36.5   | 0.02  | 34.6   | 0.09  |
|                                      |               | KF-OMI   | 41.9   | -0.26 | 44.9   | -0.37 | 46.6   | -0.08 | 39.5   | -0.29 | 36.7   | -0.38 |
|                                      | 2005-<br>2009 | KF-AQS   | 38.0   | 0.07  | 38.9   | -0.17 | 42.9   | -0.48 | 36.6   | -0.08 | 34.0   | -0.01 |
|                                      |               | KF-OMI   | 43.2   | 0.19  | 45.9   | -0.06 | 46.1   | -0.17 | 41.6   | -0.16 | 39.8   | -0.20 |
|                                      | 2010-<br>2014 | KF-AQS   | 38.3   | -0.11 | 39.8   | 0.00  | 43.1   | -1.21 | 36.5   | 0.17  | 34.5   | 0.60  |
|                                      |               | KF-OMI   | 42.3   | -0.72 | 46.8   | -1.04 | 47.8   | -1.56 | 39.1   | -0.11 | 35.3   | 0.18  |
|                                      | 2015-<br>2020 | KF-AQS   | 38.0   | -0.07 | 38.2   | -0.29 | 41.8   | -0.11 | 36.5   | 0.53  | 35.0   | -0.31 |
| KF-OMI                               |               | 40.6     | -0.30  | 42.6  | -0.72  | 46.0  | -0.19  | 38.0  | 0.15   | 35.3  | -0.44  |       |

**Table. S3.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> concentrations in 2005-2020 over West Coast (Region #3) from observations (AQS and OMI) and a priori and a posteriori (KF) simulations. Grids with low and medium anthropogenic NO<sub>x</sub> emissions are excluded in West Coast.

| #4 Middle US                         |               |          | Annual |       | Spring |       | Summer |       | Autumn |       | Winter |       |
|--------------------------------------|---------------|----------|--------|-------|--------|-------|--------|-------|--------|-------|--------|-------|
|                                      |               |          | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend |
| TS4.1 surface<br>(sampled)           | 2005-<br>2020 | AQS      | 39.7   | -0.30 | 44.3   | -0.29 | 46.0   | -0.62 | 37.8   | -0.35 | 31.0   | -0.06 |
|                                      |               | a priori | 42.2   | -0.28 | 44.8   | -0.40 | 55.2   | -0.61 | 40.6   | -0.18 | 28.3   | 0.02  |
|                                      |               | KF-AQS   | 40.4   | -0.29 | 44.8   | -0.33 | 48.5   | -0.61 | 38.4   | -0.30 | 30.2   | -0.01 |
| TS4.2<br>surface                     | 2005-<br>2020 | a priori | 41.3   | -0.23 | 44.1   | -0.34 | 54.1   | -0.48 | 39.3   | -0.18 | 27.8   | 0.04  |
|                                      |               | KF-AQS   | 40.0   | -0.21 | 44.1   | -0.28 | 49.8   | -0.46 | 37.6   | -0.22 | 28.7   | 0.05  |
| TS4.3<br>trop. column<br>(convolved) | 2005-<br>2020 | OMI      | 39.0   | 0.02  | 40.8   | 0.11  | 44.4   | 0.16  | 37.2   | -0.06 | 33.8   | -0.12 |
|                                      |               | a priori | 38.2   | -0.10 | 39.5   | -0.11 | 46.3   | -0.17 | 36.1   | -0.06 | 31.0   | -0.05 |
|                                      |               | KF-OMI   | 39.4   | -0.13 | 41.8   | -0.13 | 46.4   | 0.00  | 37.2   | -0.14 | 32.1   | -0.24 |
| TS4.4<br>trop. Column                | 2005-<br>2020 | a priori | 39.3   | -0.08 | 40.8   | -0.19 | 46.1   | -0.18 | 36.6   | -0.02 | 33.9   | 0.04  |
|                                      |               | KF-AQS   | 38.9   | -0.07 | 40.8   | -0.17 | 45.1   | -0.17 | 36.2   | -0.02 | 33.9   | 0.05  |
|                                      |               | KF-OMI   | 41.5   | -0.24 | 44.4   | -0.33 | 47.7   | -0.09 | 38.5   | -0.21 | 35.4   | -0.34 |
|                                      | 2005-<br>2009 | KF-AQS   | 39.0   | -0.43 | 41.1   | -0.62 | 45.3   | -1.44 | 36.1   | -0.15 | 33.3   | -0.58 |
|                                      |               | KF-OMI   | 42.6   | -0.30 | 45.4   | -0.42 | 47.3   | -1.26 | 39.8   | -0.14 | 37.9   | -0.59 |
|                                      | 2010-<br>2014 | KF-AQS   | 39.4   | -0.60 | 41.6   | -1.20 | 45.9   | -1.37 | 36.4   | -0.31 | 34.0   | -0.49 |
|                                      |               | KF-OMI   | 41.8   | -0.88 | 45.7   | -1.47 | 48.8   | -1.25 | 38.3   | -0.50 | 34.2   | -0.81 |
|                                      | 2015-<br>2020 | KF-AQS   | 38.5   | 0.07  | 39.7   | 0.11  | 44.2   | 0.28  | 36.1   | 0.00  | 34.3   | -0.02 |
| KF-OMI                               |               | 40.3     | -0.12  | 42.4  | -0.23  | 47.1  | 0.19   | 37.6  | -0.25  | 34.2  | -0.13  |       |

**Table. S4.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> concentrations in 2005-2020 over Middle US (Region #4) from observations (AQS and OMI) and a priori and a posteriori (KF) simulations.

| #5 Southeast US                      |               |          | Annual |       | Spring |       | Summer |       | Autumn |       | Winter |       |
|--------------------------------------|---------------|----------|--------|-------|--------|-------|--------|-------|--------|-------|--------|-------|
|                                      |               |          | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend |
| TS5.1 surface<br>(sampled)           | 2005-<br>2020 | AQS      | 39.9   | -0.47 | 46.9   | -0.54 | 42.6   | -0.92 | 37.6   | -0.44 | 32.7   | -0.03 |
|                                      |               | a priori | 44.4   | -0.38 | 46.6   | -0.47 | 54.8   | -0.86 | 43.3   | -0.24 | 33.2   | 0.00  |
|                                      |               | KF-AQS   | 40.3   | -0.43 | 46.3   | -0.52 | 44.7   | -0.87 | 37.8   | -0.37 | 32.6   | -0.02 |
| TS5.2<br>surface                     | 2005-<br>2020 | a priori | 43.7   | -0.39 | 46.0   | -0.47 | 53.1   | -0.84 | 42.9   | -0.26 | 33.0   | -0.02 |
|                                      |               | KF-AQS   | 40.6   | -0.39 | 45.3   | -0.48 | 45.3   | -0.78 | 38.9   | -0.33 | 32.8   | 0.00  |
| TS5.3<br>trop. column<br>(convolved) | 2005-<br>2020 | OMI      | 42.7   | 0.06  | 44.5   | 0.06  | 48.9   | 0.09  | 40.4   | 0.00  | 36.9   | 0.07  |
|                                      |               | a priori | 42.8   | -0.11 | 45.3   | -0.13 | 49.3   | -0.21 | 40.1   | -0.08 | 36.6   | -0.02 |
|                                      |               | KF-OMI   | 43.6   | -0.10 | 46.8   | -0.13 | 49.8   | -0.02 | 41.0   | -0.13 | 36.9   | -0.14 |
| TS5.4<br>trop. Column                | 2005-<br>2020 | a priori | 44.3   | -0.19 | 47.4   | -0.34 | 48.1   | -0.26 | 40.0   | -0.07 | 42.2   | -0.09 |
|                                      |               | KF-AQS   | 43.5   | -0.18 | 47.1   | -0.34 | 46.1   | -0.23 | 39.2   | -0.08 | 42.1   | -0.09 |
|                                      |               | KF-OMI   | 45.8   | -0.28 | 49.8   | -0.42 | 49.4   | -0.17 | 41.6   | -0.20 | 42.7   | -0.34 |
|                                      | 2005-<br>2009 | KF-AQS   | 44.1   | -0.23 | 48.0   | -1.06 | 46.5   | -0.61 | 38.9   | -0.40 | 43.3   | -0.32 |
|                                      |               | KF-OMI   | 46.9   | -0.22 | 51.0   | -0.98 | 49.3   | -0.64 | 42.1   | -0.39 | 45.9   | -0.15 |
|                                      | 2010-<br>2014 | KF-AQS   | 44.3   | -1.05 | 48.7   | -1.53 | 47.3   | -1.83 | 40.1   | -0.97 | 40.9   | -0.94 |
|                                      |               | KF-OMI   | 46.6   | -1.24 | 51.8   | -1.69 | 50.8   | -1.74 | 42.4   | -1.03 | 40.5   | -1.07 |
|                                      | 2015-<br>2020 | KF-AQS   | 42.4   | 0.14  | 45.1   | 0.14  | 44.7   | 0.19  | 38.6   | -0.15 | 42.2   | 0.44  |
| KF-OMI                               |               | 44.2     | -0.06  | 47.2  | -0.13  | 48.3  | 0.04   | 40.6  | -0.39  | 41.8  | 0.24   |       |

**Table. S5.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> concentrations in 2005-2020 over Southeast US (Region #5) from observations (AQS and OMI) and a priori and a posteriori (KF) simulations.

| #1 Britain                           |               |            | Annual |       | Spring |       | Summer |       | Autumn |       | Winter |       |
|--------------------------------------|---------------|------------|--------|-------|--------|-------|--------|-------|--------|-------|--------|-------|
|                                      |               |            | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend |
| TS6.1 surface<br>(sampled)           | 2005-<br>2020 | AirBase    | 24.4   | 0.14  | 31.8   | 0.17  | 27.5   | -0.01 | 19.7   | 0.12  | 18.8   | 0.35  |
|                                      |               | a priori   | 29.9   | 0.10  | 36.0   | 0.11  | 38.7   | -0.10 | 25.2   | 0.08  | 19.9   | 0.31  |
|                                      |               | KF-AirBase | 26.0   | 0.09  | 32.9   | 0.11  | 30.7   | -0.07 | 21.3   | 0.07  | 19.1   | 0.28  |
| TS6.2<br>surface                     | 2005-<br>2020 | a priori   | 30.0   | 0.09  | 35.9   | 0.10  | 38.5   | -0.10 | 25.4   | 0.07  | 20.2   | 0.30  |
|                                      |               | KF-AirBase | 26.7   | 0.07  | 33.4   | 0.09  | 31.9   | -0.09 | 22.0   | 0.06  | 19.5   | 0.28  |
| TS6.3<br>trop. column<br>(convolved) | 2005-<br>2020 | OMI        | 35.9   | -0.05 | 37.8   | -0.31 | 37.2   | -0.17 | 34.1   | 0.04  | 34.5   | 0.30  |
|                                      |               | a priori   | 30.9   | -0.07 | 32.3   | -0.16 | 32.2   | -0.24 | 29.8   | 0.03  | 29.3   | 0.12  |
|                                      |               | KF-OMI     | 33.7   | -0.25 | 36.2   | -0.43 | 35.9   | -0.29 | 31.7   | -0.19 | 30.8   | -0.03 |
| TS6.4<br>trop. Column                | 2005-<br>2020 | a priori   | 30.1   | 0.00  | 32.9   | 0.01  | 31.8   | 0.02  | 27.9   | -0.02 | 27.8   | 0.01  |
|                                      |               | KF-AirBase | 29.7   | 0.00  | 32.6   | 0.01  | 31.0   | 0.02  | 27.5   | -0.02 | 27.7   | 0.01  |
|                                      |               | KF-OMI     | 34.7   | -0.49 | 39.8   | -0.60 | 36.9   | -0.07 | 30.9   | -0.52 | 30.6   | -0.71 |
|                                      | 2005-<br>2009 | KF-AirBase | 29.7   | -0.29 | 32.4   | 0.18  | 31.1   | -0.85 | 27.7   | -0.27 | 27.6   | -0.27 |
|                                      |               | KF-OMI     | 37.7   | -0.52 | 42.8   | 0.03  | 37.0   | -0.42 | 34.4   | -0.92 | 35.9   | -1.35 |
|                                      | 2010-<br>2014 | KF-AirBase | 29.5   | -0.16 | 32.8   | -0.67 | 30.3   | -0.04 | 27.3   | 0.29  | 27.5   | -0.40 |
|                                      |               | KF-OMI     | 34.2   | -0.88 | 40.7   | -1.52 | 37.0   | -0.41 | 29.9   | -0.25 | 28.3   | -1.00 |
|                                      | 2015-<br>2020 | KF-AirBase | 29.9   | 0.07  | 32.6   | 0.18  | 31.4   | 0.17  | 27.5   | -0.15 | 28.1   | -0.12 |
| KF-OMI                               |               | 32.5       | -0.35  | 36.6  | -0.52  | 36.7  | -0.45  | 28.7  | -0.42  | 28.2  | -0.18  |       |

**Table. S6.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> concentrations in 2005-2020 over Britain (Region #1) from observations (AirBase and OMI) and a priori and a posteriori (KF) simulations. Grids with low and medium anthropogenic NO<sub>x</sub> emissions are excluded in Britain.

| #2 Central EU                        |               |            | Annual |       | Spring |       | Summer |       | Autumn |       | Winter |       |
|--------------------------------------|---------------|------------|--------|-------|--------|-------|--------|-------|--------|-------|--------|-------|
|                                      |               |            | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend |
| TS7.1 surface<br>(sampled)           | 2005-<br>2020 | AirBase    | 28.0   | 0.21  | 35.5   | 0.14  | 38.3   | 0.17  | 20.5   | 0.22  | 17.7   | 0.33  |
|                                      |               | a priori   | 30.5   | 0.14  | 36.5   | 0.12  | 44.7   | 0.03  | 25.2   | 0.14  | 15.9   | 0.27  |
|                                      |               | KF-AirBase | 28.2   | 0.16  | 35.4   | 0.13  | 39.0   | 0.12  | 21.3   | 0.17  | 17.4   | 0.26  |
| TS7.2<br>surface                     | 2005-<br>2020 | a priori   | 30.6   | 0.14  | 36.5   | 0.12  | 44.8   | 0.03  | 25.2   | 0.14  | 15.9   | 0.27  |
|                                      |               | KF-AirBase | 28.3   | 0.16  | 35.4   | 0.12  | 39.1   | 0.12  | 21.3   | 0.17  | 17.3   | 0.25  |
| TS7.3<br>trop. column<br>(convolved) | 2005-<br>2020 | OMI        | 35.5   | -0.22 | 36.6   | -0.51 | 37.8   | -0.25 | 33.4   | -0.14 | 34.2   | -0.02 |
|                                      |               | a priori   | 31.6   | -0.13 | 32.9   | -0.26 | 34.4   | -0.23 | 30.2   | -0.02 | 28.9   | -0.02 |
|                                      |               | KF-OMI     | 34.0   | -0.31 | 36.2   | -0.52 | 37.5   | -0.29 | 31.9   | -0.22 | 30.5   | -0.20 |
| TS7.4<br>trop. Column                | 2005-<br>2020 | a priori   | 31.0   | 0.04  | 33.4   | 0.02  | 34.7   | 0.09  | 28.5   | -0.01 | 27.3   | 0.06  |
|                                      |               | KF-AirBase | 30.4   | 0.04  | 33.0   | 0.02  | 33.5   | 0.10  | 27.8   | -0.01 | 27.4   | 0.06  |
|                                      |               | KF-OMI     | 34.9   | -0.38 | 39.3   | -0.50 | 38.9   | 0.03  | 31.1   | -0.44 | 30.0   | -0.62 |
|                                      | 2005-<br>2009 | KF-AirBase | 30.2   | -0.24 | 32.7   | 0.07  | 33.2   | -0.50 | 27.9   | -0.48 | 27.0   | -0.28 |
|                                      |               | KF-OMI     | 37.4   | -0.36 | 41.6   | -0.12 | 38.5   | -0.13 | 34.1   | -0.83 | 34.8   | -1.25 |
|                                      | 2010-<br>2014 | KF-AirBase | 30.2   | -0.11 | 33.3   | -0.61 | 32.8   | -0.06 | 27.7   | 0.24  | 27.0   | -0.17 |
|                                      |               | KF-OMI     | 34.4   | -0.72 | 40.3   | -1.49 | 38.8   | -0.30 | 30.3   | -0.12 | 27.8   | -0.71 |
|                                      | 2015-<br>2020 | KF-AirBase | 30.8   | 0.11  | 33.0   | 0.19  | 34.5   | 0.07  | 27.8   | 0.13  | 27.9   | -0.12 |
| KF-OMI                               |               | 33.3       | -0.23  | 36.7  | -0.45  | 39.3  | -0.31  | 29.2  | -0.11  | 28.0  | -0.18  |       |

**Table. S7.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> concentrations in 2005-2020 over Central EU (Region #2) from observations (AirBase and OMI) and a priori and a posteriori (KF) simulations. Grids with low and medium anthropogenic NO<sub>x</sub> emissions are excluded in Central EU.

| #3 Western EU                        |               |            | Annual |       | Spring |       | Summer |       | Autumn |       | Winter |       |
|--------------------------------------|---------------|------------|--------|-------|--------|-------|--------|-------|--------|-------|--------|-------|
|                                      |               |            | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend |
| TS8.1 surface<br>(sampled)           | 2005-<br>2020 | AirBase    | 32.3   | 0.11  | 38.9   | 0.05  | 40.1   | -0.06 | 27.6   | 0.15  | 22.6   | 0.30  |
|                                      |               | a priori   | 35.9   | 0.02  | 40.6   | -0.12 | 44.9   | -0.10 | 32.9   | 0.03  | 25.2   | 0.23  |
|                                      |               | KF-AirBase | 32.5   | 0.05  | 38.6   | -0.02 | 40.2   | -0.10 | 28.4   | 0.07  | 23.0   | 0.25  |
| TS8.2<br>surface                     | 2005-<br>2020 | a priori   | 36.0   | 0.00  | 40.6   | -0.13 | 44.8   | -0.12 | 33.2   | 0.01  | 25.6   | 0.22  |
|                                      |               | KF-AirBase | 32.8   | 0.03  | 38.6   | -0.05 | 40.2   | -0.12 | 28.8   | 0.05  | 23.5   | 0.23  |
| TS8.3<br>trop. column<br>(convolved) | 2005-<br>2020 | OMI        | 36.3   | -0.16 | 37.8   | -0.29 | 41.6   | -0.10 | 34.7   | -0.07 | 31.0   | -0.21 |
|                                      |               | a priori   | 33.3   | -0.10 | 34.0   | -0.19 | 37.8   | -0.17 | 32.0   | -0.01 | 29.5   | -0.04 |
|                                      |               | KF-OMI     | 35.7   | -0.26 | 37.5   | -0.40 | 41.3   | -0.20 | 33.7   | -0.20 | 30.5   | -0.24 |
| TS8.4<br>trop. Column                | 2005-<br>2020 | a priori   | 32.5   | 0.02  | 34.1   | -0.02 | 37.0   | 0.03  | 30.0   | -0.01 | 28.7   | 0.09  |
|                                      |               | KF-AirBase | 31.8   | 0.03  | 33.6   | 0.00  | 35.9   | 0.03  | 29.3   | 0.00  | 28.5   | 0.09  |
|                                      |               | KF-OMI     | 36.4   | -0.36 | 40.0   | -0.44 | 41.4   | -0.02 | 32.7   | -0.41 | 31.2   | -0.53 |
|                                      | 2005-<br>2009 | KF-AirBase | 31.6   | -0.23 | 33.3   | -0.15 | 35.7   | -0.64 | 29.3   | -0.21 | 27.9   | -0.06 |
|                                      |               | KF-OMI     | 38.5   | -0.40 | 41.8   | -0.53 | 41.1   | -0.25 | 35.4   | -0.55 | 35.4   | -0.81 |
|                                      | 2010-<br>2014 | KF-AirBase | 31.7   | -0.25 | 33.8   | -0.60 | 35.5   | -0.38 | 29.2   | -0.14 | 28.4   | -0.14 |
|                                      |               | KF-OMI     | 36.2   | -0.79 | 41.1   | -1.19 | 41.6   | -0.54 | 32.1   | -0.47 | 29.2   | -0.67 |
|                                      | 2015-<br>2020 | KF-AirBase | 32.1   | -0.03 | 33.6   | -0.04 | 36.5   | -0.20 | 29.4   | -0.03 | 29.1   | 0.02  |
| KF-OMI                               |               | 34.8       | -0.32  | 37.6  | -0.57  | 41.4  | -0.45  | 31.1  | -0.35  | 29.3  | -0.09  |       |

**Table. S8.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> concentrations in 2005-2020 over Western EU (Region #3) from observations (AirBase and OMI) and a priori and a posteriori (KF) simulations.

| #4 Iberian Peninsula                 |               |            | Annual |       | Spring |       | Summer |       | Autumn |       | Winter |       |
|--------------------------------------|---------------|------------|--------|-------|--------|-------|--------|-------|--------|-------|--------|-------|
|                                      |               |            | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend |
| TS9.1 surface<br>(sampled)           | 2005-<br>2020 | AirBase    | 34.2   | 0.06  | 39.5   | 0.02  | 41.1   | -0.15 | 31.2   | 0.12  | 25.3   | 0.23  |
|                                      |               | a priori   | 40.3   | -0.06 | 43.9   | -0.21 | 48.4   | -0.18 | 38.1   | -0.01 | 30.6   | 0.13  |
|                                      |               | KF-AirBase | 35.2   | -0.02 | 40.0   | -0.08 | 42.1   | -0.20 | 32.5   | 0.02  | 26.2   | 0.17  |
| TS9.2<br>surface                     | 2005-<br>2020 | a priori   | 40.4   | -0.07 | 43.9   | -0.22 | 48.7   | -0.17 | 38.4   | -0.02 | 30.9   | 0.10  |
|                                      |               | KF-AirBase | 36.2   | -0.05 | 40.6   | -0.13 | 43.4   | -0.22 | 33.4   | 0.00  | 27.2   | 0.13  |
| TS9.3<br>trop. column<br>(convolved) | 2005-<br>2020 | OMI        | 37.8   | -0.12 | 39.1   | -0.20 | 45.4   | 0.05  | 35.7   | -0.22 | 30.9   | -0.12 |
|                                      |               | a priori   | 35.3   | -0.08 | 36.0   | -0.16 | 41.5   | -0.06 | 33.3   | -0.07 | 30.5   | -0.05 |
|                                      |               | KF-OMI     | 37.8   | -0.21 | 39.2   | -0.30 | 45.3   | -0.03 | 35.2   | -0.28 | 31.2   | -0.24 |
| TS9.4<br>trop. Column                | 2005-<br>2020 | a priori   | 34.2   | 0.02  | 35.0   | -0.06 | 40.7   | 0.03  | 31.3   | 0.00  | 29.9   | 0.09  |
|                                      |               | KF-AirBase | 33.6   | 0.02  | 34.5   | -0.04 | 39.7   | 0.02  | 30.6   | 0.00  | 29.5   | 0.10  |
|                                      |               | KF-OMI     | 38.1   | -0.30 | 40.8   | -0.39 | 45.3   | 0.01  | 34.2   | -0.36 | 32.0   | -0.46 |
|                                      | 2005-<br>2009 | KF-AirBase | 33.3   | -0.26 | 34.4   | -0.20 | 39.5   | -0.69 | 30.4   | -0.31 | 29.0   | 0.02  |
|                                      |               | KF-OMI     | 39.7   | -0.41 | 42.1   | -0.33 | 44.9   | -0.43 | 36.3   | -0.55 | 35.6   | -0.50 |
|                                      | 2010-<br>2014 | KF-AirBase | 33.6   | -0.38 | 34.7   | -0.63 | 39.4   | -0.42 | 30.7   | -0.68 | 29.7   | 0.10  |
|                                      |               | KF-OMI     | 38.2   | -0.86 | 42.0   | -1.18 | 45.6   | -0.44 | 34.0   | -0.97 | 30.4   | -0.37 |
|                                      | 2015-<br>2020 | KF-AirBase | 33.8   | 0.03  | 34.4   | -0.28 | 40.1   | 0.05  | 30.7   | 0.10  | 29.9   | 0.34  |
| KF-OMI                               |               | 36.7       | -0.23  | 38.6  | -0.74  | 45.4  | -0.07  | 32.7  | -0.29  | 30.2  | 0.20   |       |

**Table. S9.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> concentrations in 2005-2020 over Iberian Peninsula (Region #4) from observations (AirBase and OMI) and a priori and a posteriori (KF) simulations.

| #5 Apennine Peninsula                 |               |            | Annual |       | Spring |       | Summer |       | Autumn |       | Winter |       |
|---------------------------------------|---------------|------------|--------|-------|--------|-------|--------|-------|--------|-------|--------|-------|
|                                       |               |            | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend | Mean   | Trend |
| TS10.1 surface<br>(sampled)           | 2005-<br>2020 | AirBase    | 34.0   | 0.08  | 39.2   | -0.06 | 45.5   | -0.03 | 29.2   | 0.17  | 22.0   | 0.22  |
|                                       |               | a priori   | 41.8   | -0.05 | 45.3   | -0.30 | 53.9   | -0.10 | 39.0   | 0.00  | 29.1   | 0.20  |
|                                       |               | KF-AirBase | 35.3   | 0.04  | 40.1   | -0.09 | 46.4   | -0.04 | 31.0   | 0.11  | 23.7   | 0.18  |
| TS10.2<br>surface                     | 2005-<br>2020 | a priori   | 41.8   | -0.06 | 45.2   | -0.30 | 52.8   | -0.10 | 39.2   | -0.01 | 30.1   | 0.17  |
|                                       |               | KF-AirBase | 36.6   | -0.02 | 41.0   | -0.16 | 46.1   | -0.11 | 33.0   | 0.03  | 26.5   | 0.16  |
| TS10.3<br>trop. column<br>(convolved) | 2005-<br>2020 | OMI        | 38.7   | -0.13 | 39.5   | -0.25 | 46.7   | 0.02  | 36.5   | -0.20 | 31.9   | -0.15 |
|                                       |               | a priori   | 35.4   | -0.08 | 35.6   | -0.17 | 43.0   | -0.02 | 33.4   | -0.07 | 29.6   | -0.06 |
|                                       |               | KF-OMI     | 37.7   | -0.21 | 38.7   | -0.33 | 46.0   | -0.02 | 35.2   | -0.25 | 30.7   | -0.25 |
| TS10.4<br>trop. Column                | 2005-<br>2020 | a priori   | 34.9   | 0.04  | 35.3   | -0.10 | 44.1   | 0.12  | 31.8   | 0.02  | 28.5   | 0.09  |
|                                       |               | KF-AirBase | 34.0   | 0.04  | 34.5   | -0.08 | 42.7   | 0.11  | 30.8   | 0.03  | 28.1   | 0.10  |
|                                       |               | KF-OMI     | 38.2   | -0.26 | 40.3   | -0.42 | 47.6   | 0.11  | 34.2   | -0.29 | 30.8   | -0.46 |
|                                       | 2005-<br>2009 | KF-AirBase | 33.5   | -0.24 | 34.6   | -0.68 | 41.8   | 0.17  | 30.4   | -0.57 | 27.6   | -0.39 |
|                                       |               | KF-OMI     | 39.6   | -0.29 | 41.7   | -0.80 | 46.5   | 0.43  | 36.0   | -0.74 | 34.5   | -1.06 |
|                                       | 2010-<br>2014 | KF-AirBase | 34.1   | -0.28 | 34.9   | -0.58 | 42.8   | -0.51 | 31.0   | -0.02 | 27.6   | -0.28 |
|                                       |               | KF-OMI     | 38.3   | -0.81 | 41.5   | -1.19 | 48.2   | -0.67 | 34.0   | -0.45 | 28.7   | -0.79 |
|                                       | 2015-<br>2020 | KF-AirBase | 34.2   | 0.03  | 34.2   | -0.24 | 43.3   | -0.07 | 30.8   | 0.25  | 28.9   | 0.01  |
| KF-OMI                                |               | 37.0       | -0.19  | 38.0  | -0.70  | 48.0  | -0.16  | 32.9  | -0.01  | 29.5  | -0.11  |       |

**Table. S10.** Averages (with units ppb or DU) and trends (with units ppb yr<sup>-1</sup> or DU yr<sup>-1</sup>) of surface and tropospheric column O<sub>3</sub> concentrations in 2005-2020 over Apennine Peninsula (Region #5) from observations (AirBase and OMI) and a priori and a posteriori (KF) simulations.