# Supplementary tables

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Source** | **Emissions**  **(TgN/yr2)** | **Tropospheric O3 burden (TgO3/yr)** | **OPE**  **((mol O3**  **/mol N)/yr)** | **Surface mean**  **(ppbv/yr)** | **Population weighted mean (ppbv/yr)** |
| Total | (-8.77e-03/0.29) (0.06) | (0.52/0.92) (1.44e-05) | N/A | (0.06/0.09) (1.95e-06) | (0.15/0.18) (7.13e-08) |
| Stratosphere | N/A | (-0.19/0.20) (1.00) | N/A | (2.01e-03/0.03) (0.03) | (-0.02/6.33e-03) (0.18) |
| Aircraft | (0.02/0.03) (1.54e-07) | (0.19/0.37) (1.05e-07) | (-6.6e-03/0.01) (0.53) | (0.01/0.03) (7.13e-08) | (0.01/0.03) (2.76e-06) |
| Biogenic1 | (0.e+00/0.e+00) (1.00) | (-0.09/-0.03) (1.19e-04) | (-5.07e-03/-1.67e-03) (1.19e-04) | (-6.33e-03/-1.55e-03) (1.01e-03) | (-0.04/-0.02) (7.83e-04) |
| Biomass Burning | (-0.06/-0.01) (0.01) | (-0.29/-0.11) (4.68e-04) | (-0.01/-5.14e-04) (0.03) | (-0.03/-6.97e-03) (1.64e-03) | (-0.03/-0.01) (8.91e-05) |
| Lightning | (-0.02/-4.30e-03) (2.62e-03) | (-0.61/-0.23) (2.09e-04) | (-0.02/-6.08e-03) (1.29e-03) | (-0.03/-9.45e-03) (4.68e-04) | (-0.04/-0.02) (2.74e-04) |
| Extra Production | N/A | (4.54e-03/8.50e-03) (5.41e-06) | N/A | (2.39e-04/5.62e-04) (1.19e-04) | (4.93e-04/9.29e-04) (4.94e-05) |
| Anthropogenic | (0.03/0.35) (0.03) | (1.02/1.90) (2.69e-05) | (5.33e-03/9.15e-03) (5.41e-06) | (0.06/0.10) (1.38e-06) | (0.21/0.24) (9.60e-09) |
| Regional contribution to Anthropogenic component | | | | | |
| International Shipping | (0.06/0.11) (3.87e-06) | (0.15/0.47) (1.01e-03) | (-0.01/-4.68e-03) (4.68e-04) | (0.03/0.07) (1.44e-05) | (0.03/0.04) (3.87e-06) |
| North America | (-0.35/-0.30) (4.16e-09) | (-0.37/-0.30) (4.16e-09) | (0.01/0.01) (2.17e-08) | (-0.05/-0.04) (2.72e-09) | (-0.05/-0.04) (1.45e-08) |
| Europe | (-0.12/-0.10) (2.72e-09) | (-0.12/-0.10) (3.24e-08) | (1.63e-03/3.66e-03) (1.58e-04) | (-0.02/-0.02) (6.34e-09) | (-0.03/-0.02) (7.13e-08) |
| East Asia | (0.07/0.44) (0.01) | (0.22/0.60) (4.68e-04) | (-3.4e-03/3.76e-03) (0.83) | (0.02/0.05) (1.19e-04) | (0.05/0.08) (4.71e-07) |
| South Asia | (0.13/0.15) (4.16e-09) | (0.22/0.28) (3.24e-08) | (-0.01/-9.42e-03) (6.77e-07) | (0.02/0.02) (6.34e-09) | (0.10/0.12) (4.82e-08) |
| Russia, Belarus, Ukraine | (-0.02/-0.01) (6.77e-07) | (-0.02/-9.86e-03) (1.44e-05) | (5.89e-04/2.24e-03) (4.12e-03) | (-3.92e-03/-1.38e-03) (2.09e-04) | (-5.42e-03/-1.31e-03) (2.62e-03) |
| Middle East | (0.05/0.07) (4.71e-07) | (0.05/0.09) (1.38e-06) | (-0.01/-0.01) (2.24e-07) | (5.22e-03/8.81e-03) (1.95e-06) | (8.36e-03/0.02) (3.59e-04) |
| Rest of the World | (0.14/0.16) (9.60e-09) | (0.62/0.89) (3.26e-07) | (-0.01/-4.1e-03) (2.09e-04) | (0.04/0.05) (1.45e-08) | (0.07/0.08) (6.34e-09) |

1Since we prescribe an annually repeating seasonal cycle of biogenic/soil NOx emissions, the annual mean value is held constant leading to zero slope and p-value=1

\*p-value representing medium certainty (between 0.05 and 0.1)

\*\*p-value representing low certainty (>0.1)

**Table S1**: This table provides the 95th percentile confidence interval and p-values for the slopes reported in Table 3 for the NOx-tagged metrics. The first brackets show the 95th percentile confidence interval separated by a slash (“/”) and the second brackets report the p-value of the slope.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Field (Units)[Period]** | **Mean** | **Slope in units/yr (slope in %/yr)** | **95 % confidence interval of slope** | **p-value of slope** |
| Emission (TgN/yr)  [2000-2011] | 8.63 | 0.49(8.05) | (0.44/0.53) | 8.30E-06 |
| Emission (TgN/yr)  [2011-2018] | 9.96 | -0.44(-3.90) | (-0.54/-0.38) | 8.37E-04 |
| Tropospheric O3 burden (Tg) [2000-2011] | 15.63 | 0.66(5.37) | (0.59/0.75) | 1.56E-05 |
| Tropospheric O3 burden (Tg) [2011-2018] | 18.41 | -0.51(-2.67) | (-0.67/-0.24) | 9.37E-03 |
| OPE (mol O3/mol N) [2000-2011] | 0.53 | -7.89e-03 | (-0.01/-4.21e-03) | 2.03E-03 |
| OPE (mol O3/mol N) [2011-2018] | 0.54 | 0.01 | (6.4e-03/0.02) | 8.37E-04 |
| Surface mean (ppbv)  [2000-2011] | 1.5 | 0.05(3.91) | (0.04/0.05) | 8.30E-06 |
| Surface mean (ppbv)  [2011-2018] | 1.73 | -0.04(-2.03) | (-0.05/-0.02) | 0.02 |
| Population weighted mean (ppbv) [2000-2011] | 4.06 | 0.08(2.38) | (0.06/0.10) | 9.28E-05 |
| Population weighted mean (ppbv) [2011-2018] | 4.6 | \*\*0.02(0.38) | (-0.03/0.06) | 0.27 |

**Table S2**: The mean, slope, 95 % confidence interval (in brackets separated by a slash (“/”)), and p-value of various NOx-tagged metrics attributed to East Asian NOx emissions over the pre-2011 (2000-2011) and post-2011 (2011-2018) periods.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Source** | **Emissions**  **(TgC/yr2)** | **Tropospheric O3 burden (TgO3/yr)** | **OPE**  **((mol O3**  **/mol C)/yr)** | **Surface mean (ppbv/yr)** | **Population weighted mean (ppbv/yr)** |
| Total | (0.67/6.55) (9.63e-03) | (0.52/0.92) (1.44e-05) | (-1.29e-04/8.48e-05) (0.73) | (0.06/0.09) (1.95e-06) | (0.15/0.18) (7.13e-08) |
| Stratosphere | N/A | (-0.18/0.27) (0.78) | N/A | (3.91e-03/0.03) (7.84e-03) | (-0.02/8.67e-03) (0.53) |
| Aircraft | (2.31e-03/5.78e-03) (7.53e-06) | (3.20e-04/6.84e-04) (2.76e-06) | (-3.36e-05/8.41e-05) (0.33) | (2.22e-05/4.84e-05) (2.76e-06) | (2.63e-05/6.27e-05) (1.04e-05) |
| Biogenic | (-0.07/2.12) (0.06) | (-0.02/0.15) (0.11) | (-1.02e-04/4.16e-05) (0.67) | (4.69e-03/0.01) (7.83e-04) | (0.02/0.04) (2.69e-05) |
| Biomass Burning | (-2.85/1.11) (0.58) | (-0.22/0.06) (0.29) | (-1.27e-04/6.83e-05) (0.62) | (-0.01/0.01) (0.78) | (-0.01/0.01) (0.89) |
| Methane oxidation | (1.40/2.27) (1.38e-06) | (0.18/0.70) (1.01e-03) | (-1.8e-04/2.48e-05) (0.16) | (0.03/0.07) (6.65e-05) | (0.02/0.07) (1.58e-04) |
| Extra Production | N/A | (0.02/0.03) (6.77e-07) | N/A | (1.53e-03/2.48e-03) (2.76e-06) | (2.92e-03/6.01e-03) (1.97e-05) |
| Anthropogenic | (1.07/2.36) (3.59e-04) | (0.30/0.63) (1.58e-04) | (9.98e-05/2.94e-04) (2.74e-04) | (-2.11e-03/0.01) (0.14) | (0.08/0.11) (1.05e-07) |
| Regional contribution to Anthropogenic component | | | | | |
| International Shipping | (8.40e-03/0.02) (3.65e-05) | (-4.01e-03/-1.32e-03) (1.58e-04) | (-2.41e-03/-8.86e-04) (7.13e-08) | (-3.18e-04/-7.31e-05) (5.13e-03) | (-2.79e-04/1.34e-04) (0.53) |
| North America | (-2.22/-1.47) (4.16e-09) | (-0.20/-0.14) (4.16e-09) | (1.18e-04/1.88e-04) (1.95e-06) | (-0.03/-0.02) (6.34e-09) | (-0.03/-0.02) (1.45e-08) |
| Europe | (-0.78/-0.64) (6.34e-09) | (-0.09/-0.08) (9.60e-09) | (-7.84e-05/2.84e-06) (0.07) | (-0.02/-0.01) (2.17e-08) | (-0.02/-0.02) (1.05e-07) |
| East Asia | (0.54/2.51) (7.84e-03) | (0.12/0.34) (8.91e-05) | (1.17e-04/3.08e-04) (2.09e-04) | (0.01/0.03) (1.58e-04) | (0.05/0.07) (6.77e-07) |
| South Asia | (1.08/1.30) (2.72e-09) | (0.14/0.17) (2.17e-08) | (-9.17e-05/5.00e-05) (0.44) | (0.01/0.01) (9.60e-09) | (0.06/0.07) (4.82e-08) |
| Russia, Belarus, Ukraine | (-0.07/-0.04) (3.26e-07) | (-0.01/-8.50e-03) (7.13e-08) | (-1.81e-04/-3.46e-05) (2.08e-03) | (-1.59e-03/-9.28e-04) (2.24e-07) | (-2.4e-03/-9.23e-04) (1.58e-04) |
| Middle East | (-0.10/0.04) (0.21) | (-0.02/-6.54e-03) (0.01) | (-2.52e-04/-2.78e-05) (6.36e-03) | (-2.42e-03/6.12e-05) (0.06) | (-5.27e-03/5.51e-05) (0.05) |
| Rest of the World | (1.38/1.59) (1.45e-08) | (0.20/0.32) (1.95e-06) | (-1.19e-04/1.67e-04) (0.58) | (0.01/0.02) (4.71e-07) | (0.02/0.03) (6.34e-09) |

**Table S3**: Same as Table S1 but for RC-tagged metrics (in Table 5)

|  |  |  |
| --- | --- | --- |
| **Field (Units)**  **Annual mean: 2010** | **HTAP v3**  **(this study)** | **HTAP v2**  **(Butler et al. 2020)** |
| CH4 mixing ratio (ppb) | 1788 | 1760 |
| Total atmospheric CH4 burden (TgCH4) | 4980 | 4822 |
| Trop. CH4 oxidation rate (TgC/yr) | 443.53 | 410 |
| Trop. CH4 lifetime (years) | 8.42 | 8.82 |
| Trop. O3 burden (Tg O3) | 319.73 | 319 |
| Trop. O3\_X\_CH4 burden (Tg O3) | 133.31 | 113 |
| OPE-CH4 (mol O3/ mol C) | 0.075 | 0.07 |

**Table S4**: Comparison of various CH4-related metrics between this study and Butler et al. 2020, for the year 2010. The tropospheric CH4 lifetime reported as 7.59 years was calculated wrongly as the tropospheric burden of CH4 divided by the tropospheric CH4 oxidation rate. We therefore recalculate the tropospheric CH4 lifetime, as the global CH4 burden divided by the tropospheric CH4 oxidation rate (mentioned in section 3.2.2), to be 8.82 years in the year 2010 simulation performed by Butler et al., 2020.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Quantity (units)** | **Mean** | **Slope in units/yr**  **(Slope in %/yr)** | **95% confidence interval** | **p-value** |
| Prescribed CH4 conc. (ppm) | 1.79 | 7.24e-03(0.41) | (7.24e-03/0.41) | 9.60E-09 |
| CH4 oxidation rate (TgC/yr) | 437.98 | 1.81(0.42) | (1.81/0.42) | 1.38E-06 |
| CH4 lifetime (years) | 8.54 | -0.013 | (-0.01/-0.16) | 4.12E-03 |
| Trop. OH conc.  (Air mass weighted)  (105 molec/cm3) | 12.63 | \*\*0.01(0.08) | (0.01/0.08) | 0.23 |
| Trop. OH conc.  (CH4-reaction weighted)  (105 molec/cm3) | 18.94 | 0.02(0.12) | (0.02/0.12) | 4.12E-03 |

\*\*trend with low certainty (p-value > 0.1)

**Table S5**: The mean, slope, 95 % confidence interval (in brackets separated by a slash (“/”)), and p-value of various CH4-related quantities over the 2000-2018 period shown in Fig. 4.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Field (Units)[Period]** | **Mean** | **Slope in units/yr (slope in %/yr)** | **95 % confidence interval** | **p-value** |
| Emission (TgC/yr)  [2000-2011] | 92.42 | 3.54(4.93) | (2.74/3.92) | 8.30E-06 |
| Emission (TgC/yr)  [2011-2018] | 101.42 | -2.82(-2.60) | (-4.22/-1.39) | 8.37E-04 |
| Tropospheric O3 burden (Tg) [2000-2011] | 10.25 | 0.42(5.07) | (0.36/0.50) | 1.56E-05 |
| Tropospheric O3 burden (Tg) [2011-2018] | 12 | \*\*-0.17(-1.39) | (-0.31/0.01) | 0.11 |
| OPE (mol O3/mol C) [2000-2011] | 0.03 | \*\*7.25e-05 | (-1.32e-04/2.79e-04) | 0.37 |
| OPE (mol O3/mol C) [2011-2018] | 0.03 | 4.23e-04 | (2.3e-04/5.98e-04) | 9.37E-03 |
| Surface mean (ppbv)  [2000-2011] | 0.98 | 0.04(4.63) | (0.03/0.04) | 8.30E-06 |
| Surface mean (ppbv)  [2011-2018] | 1.13 | -0.02(-1.53) | (-0.03/-8.80e-03) | 9.37E-03 |
| Population weighted mean (ppbv) [2000-2011] | 2.34 | 0.07(3.72) | (0.06/0.08) | 5.21E-05 |
| Population weighted mean (ppbv) [2011-2018] | 2.81 | \*\*0.02(0.59) | (-0.02/0.07) | 0.54 |

\*\*trend with low certainty (p-value > 0.1)

**Table S6**: Same as Table S2 but for RC-tagged metrics attributed to East Asian anthropogenic NMRC emissions.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Tag** | **Emissions**  **(TgN/yr)** | **Trop O3 burden (TgO3)** | **OPE**  **(mol O3**  **/mol C)** | **Surface mean (ppbv)** | **Population weighted mean (ppbv)** |
| Explicitly tagged regions within the Rest of the World tag in our NOx-tagged simulation | | | | | |
| Central Asia | 0.38 (0.77) | 0.75 (0.24) | 0.59 (28.89) | 0.15 (0.58) | 0.34 (1.02) |
| Mexico and Central America | 1.37 (2.79) | 8.47 (2.67) | 1.81 (88.93) | 0.65 (2.55) | 0.78 (2.35) |
| North Africa | 0.60 (1.21) | 1.65 (0.52) | 0.82 (40.24) | 0.27 (1.05) | 0.45 (1.36) |
| South-East Asia | 1.76 (3.58) | 11.78 (3.71) | 1.98 (97.28) | 0.68 (2.67) | 0.68 (2.07) |
| Southern Hemisphere regions\* | 3.10 (6.31) | 17.26 (5.43) | 1.63 (80.11) | 2.02 (7.94) | 0.70 (2.13) |

\*Includes southern hemisphere regions: Sahel Africa, Pacific-Australia-New Zealand and South America.

**Table S7**: 2000-2018 Mean contributions from explicitly tagged regions to the “Rest of the World” tag in our NOx-tagged simulation discussed this study.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Tag** | **Emissions**  **(TgN/yr2)** | **Trop O3 burden (TgO3/yr)** | **OPE**  **(mol O3**  **/mol C/yr)** | **Surface mean (ppbv/yr)** | **Population weighted mean (ppbv/yr)** |
| Explicitly tagged regions within the Rest of the World tag in our NOx-tagged simulation | | | | | |
| Central Asia | 8.52e-03(2.90) | 0.01(1.63) | -4.10E-03 | 2.68e-03(2.13) | 5.22e-03(1.71) |
| Mexico and Central America | \*\*2.28e-03(0.17) | 0.05(0.60) | 8.91E-03 | \*\*6.85e-04(0.11) | \*\*2.14e-04(0.03) |
| North Africa | 0.02(5.52) | 0.05(3.93) | -8.20E-03 | 7.29e-03(3.50) | 0.01(3.20) |
| South-East Asia | 0.05(4.19) | 0.25(2.39) | -0.02 | 0.01(2.33) | 0.01(2.10) |
| Southern Hemisphere regions\*\*\* | 0.06(2.57) | 0.30(2.02) | -5.27E-03 | 0.02(1.20) | 0.01(2.09) |

\*0.5<p<0.1 (medium certainty)

\*\*p≥0.1 (low certainty)

\*\*\*Includes southern hemisphere regions: Sahel Africa, Pacific-Australia-New Zealand, and South America.

**Table S8**: Theil-Sen estimator/slope of trends in contributions from explicitly tagged regions to the “Rest of the World” tag in our NOx-tagged simulation discussed this study. Trend slope and p-value estimation has been summarized in section 2.3. The 95 % confidence interval and p-value for each of these estimated trends is provided in Table S10. In brackets is the trend in %/year.

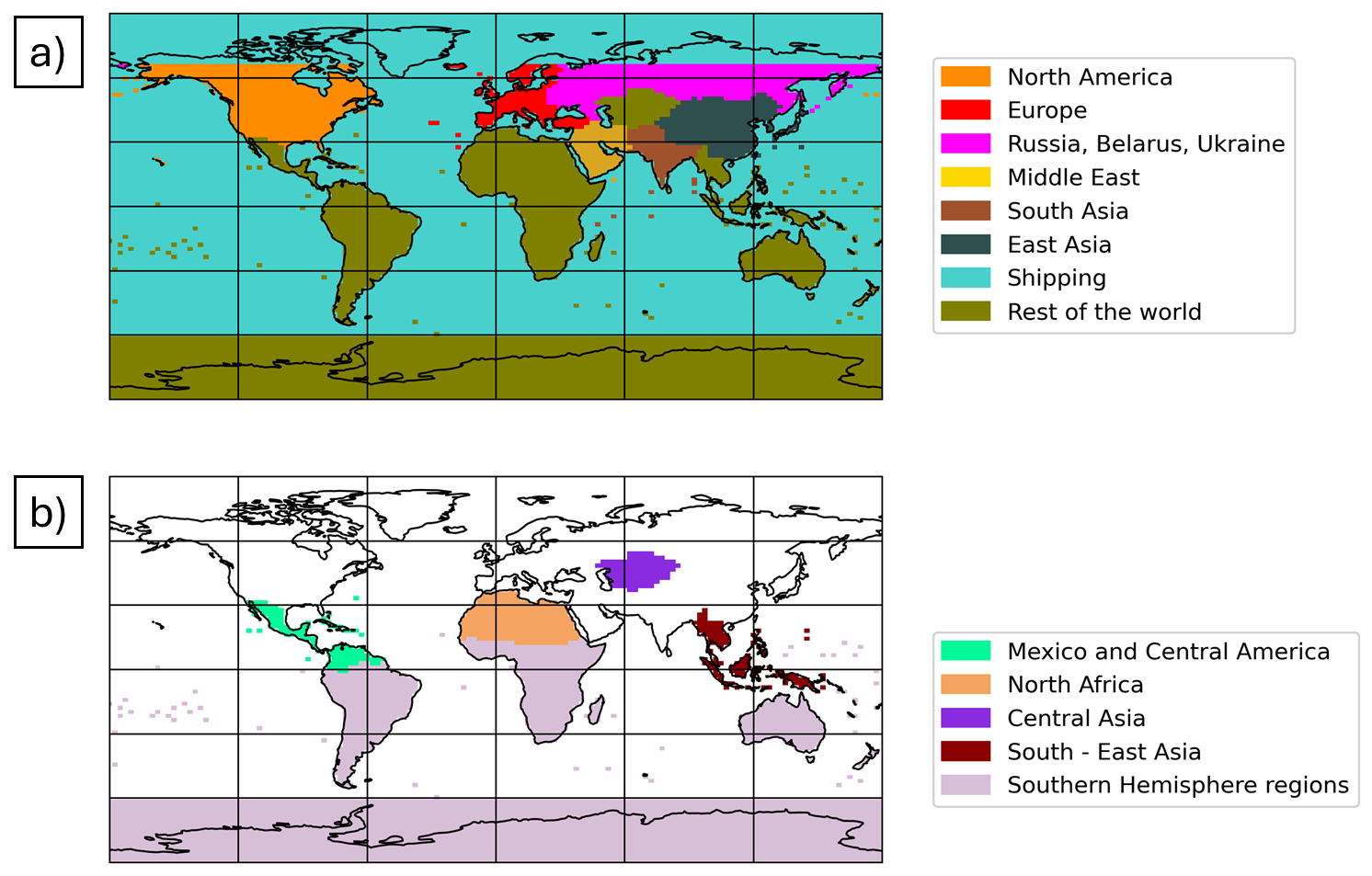
|  |  |  |
| --- | --- | --- |
| **Region** | **Population** | **Population percentage** |
| Total | 7954963550 | 100 |
| Europe | 645060000 | 8.1089 |
| North America | 378064000 | 4.752555 |
| East Asia | 1686480000 | 21.20035 |
| Russia, Belarus, Ukraine | 205763000 | 2.586599 |
| South Asia | 1882710000 | 23.66711 |
| Middle East | 278477000 | 3.50067 |
| Rest of the World | 2875973800 | 36.1532 |
| Explicitly tagged regions in our NOx-tagged simulation within the “Rest of the World” tag | | |
| Southeast Asia | 714549000 | 8.98243 |
| Mexico and Central America | 341845000 | 4.297254 |
| North Africa | 310292000 | 3.900609 |
| Central Asia | 77050900 | 0.968589 |
| Southern Hemisphere regions\* | 1432236900 | 18 |

\*Includes southern hemisphere regions: Sahel Africa, Pacific-Australia-New Zealand, and South America.

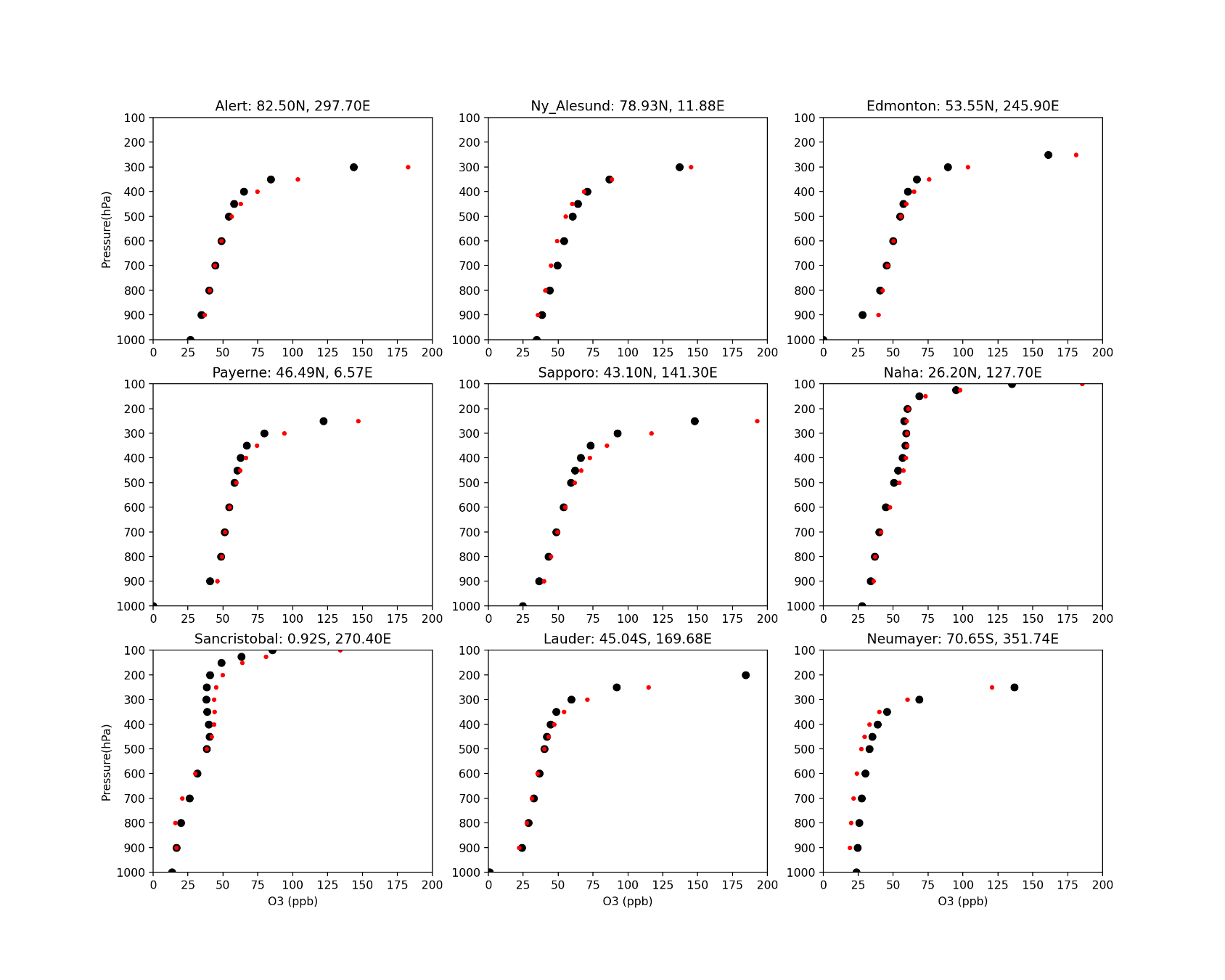
**Table S9**: Population at various regions considered in this study. These numbers are approximate numbers derived from re-gridding a fine resolution dataset to a coarser resolution grid used in our study.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Tag** | **Emissions**  **(TgN/yr2)** | **Trop O3 burden (TgO3/yr)** | **OPE**  **(mol O3**  **/mol C/yr)** | **Surface mean (ppbv/yr)** | **Population weighted mean (ppbv/yr)** |
| Explicitly tagged regions within the Rest of the World tag in our NOx-tagged simulation | | | | | |
| Central Asia | (6.44e-03/0.01) (6.65e-05) | (6.17e-03/0.02) (6.07e-04) | (-6.03e-03/-2.14e-03) (8.91e-05) | (1.94e-03/3.33e-03) (2.69e-05) | (3.89e-03/6.81e-03) (1.44e-05) |
| Mexico and Central America | (-3.28e-03/5.17e-03) (0.40) | (0.02/0.07) (5.13e-03) | (4.82e-03/0.01) (1.29e-03) | (-7.79e-04/2.33e-03) (0.44) | (-2.14e-03/1.83e-03) (0.58) |
| North Africa | (0.02/0.02) (4.16e-09) | (0.04/0.05) (2.17e-08) | (-0.01/-5.8e-03) (3.87e-06) | (6.69e-03/7.77e-03) (2.17e-08) | (9.34e-03/0.01) (3.26e-07) |
| South-East Asia | (0.05/0.06) (2.72e-09) | (0.17/0.33) (2.69e-05) | (-0.04/-5.81e-03) (1.64e-03) | (0.01/0.02) (2.24e-07) | (0.01/0.01) (4.82e-08) |
| Southern Hemisphere regions\* | (0.06/0.07) (1.54e-07) | (0.26/0.34) (1.54e-07) | (-7.86e-03/-2.48e-03) (1.01e-03) | (0.02/0.03) (6.77e-07) | (0.01/0.01) (2.17e-08) |

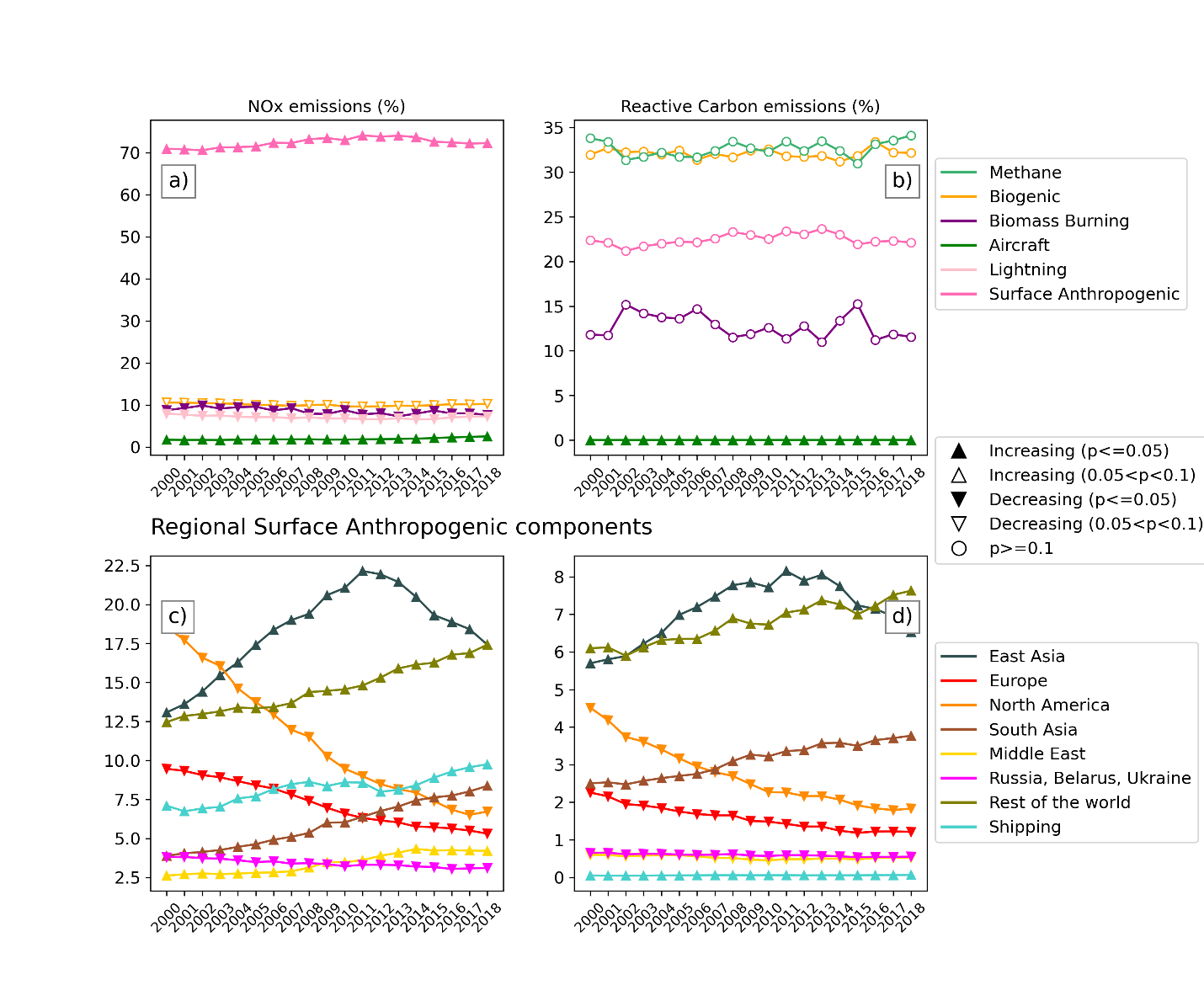
**Table S10**: This table provides the 95th percentile confidence interval and p-values for the slopes reported in Table S8 for the NOx-tagged metrics attributed to the explicitly tagged regions within the “Rest of the World” tag in our NOx-tagged simulation. The first brackets show the 95th percentile confidence interval separated by a slash (“/”) and the second brackets report the p-value of the slope.



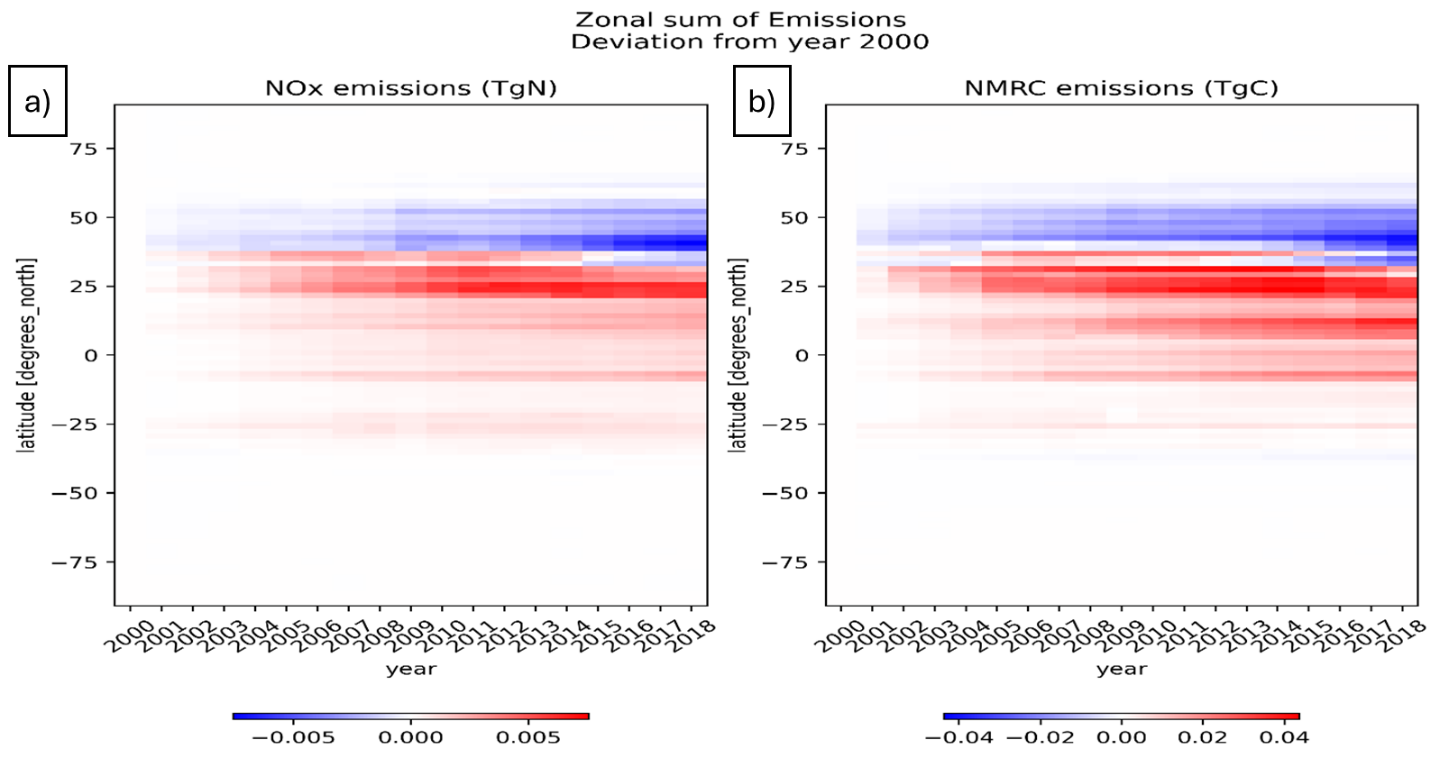
**Fig. S1**: a) Regions considered as regional anthropogenic tags in our simulations (NOx and RC tagged), b) Explicitly tagged regions within the “Rest of the World” tag in our NOx-tagged simulation.



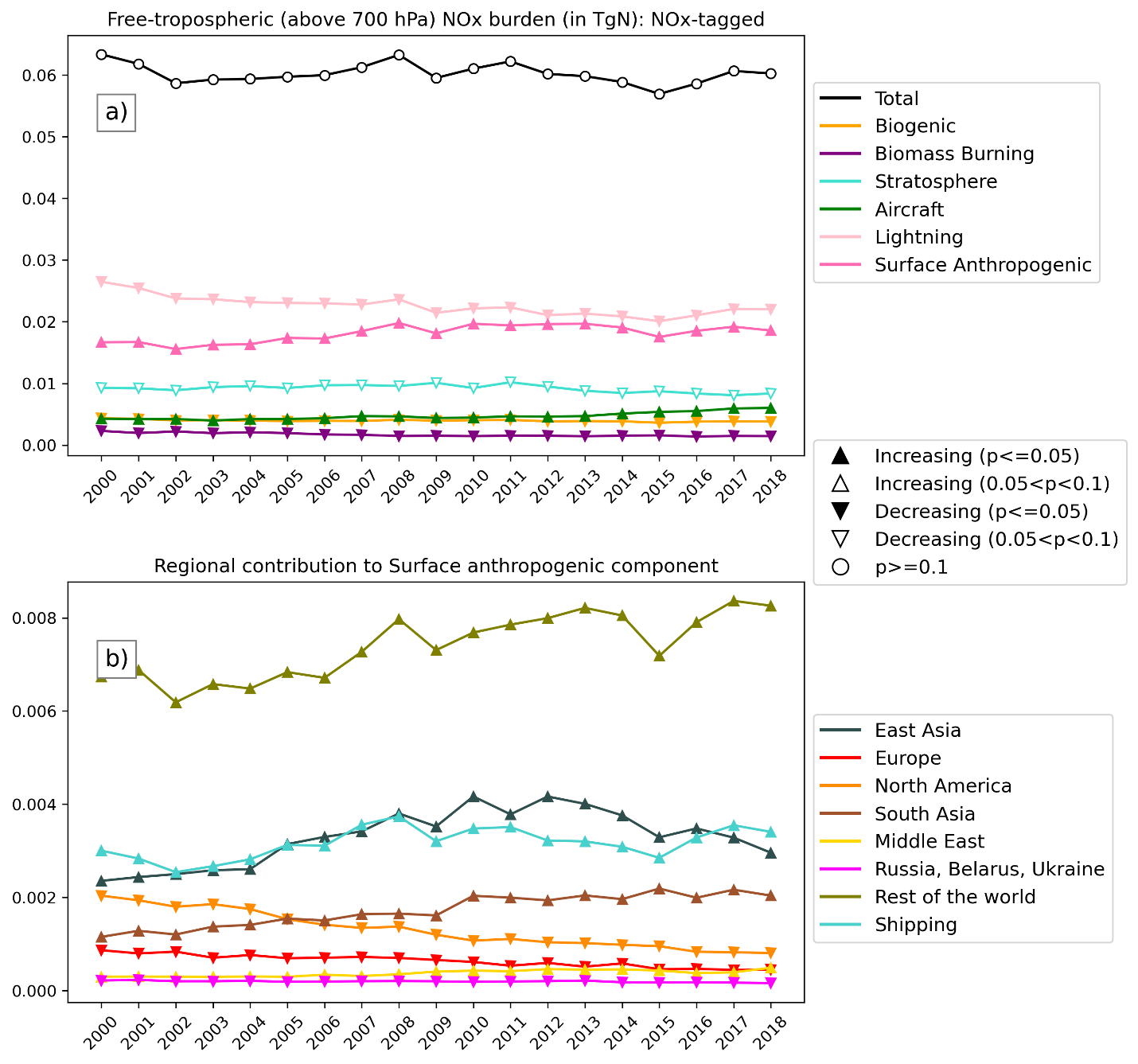
**Fig. S2**: Vertical profile comparison of annual mean tropospheric O3 mixing ratio at nine ozonesonde stations (coordinates stated as the title) from the model (red dots) for the 2000-2010 period and ozonesonde climatology (black dots) for the 1995-2010 period. The nine selected ozonesonde stations are latitudinally representative across the northern and southern hemispheres, as evaluated in Zhang et al., 2016.



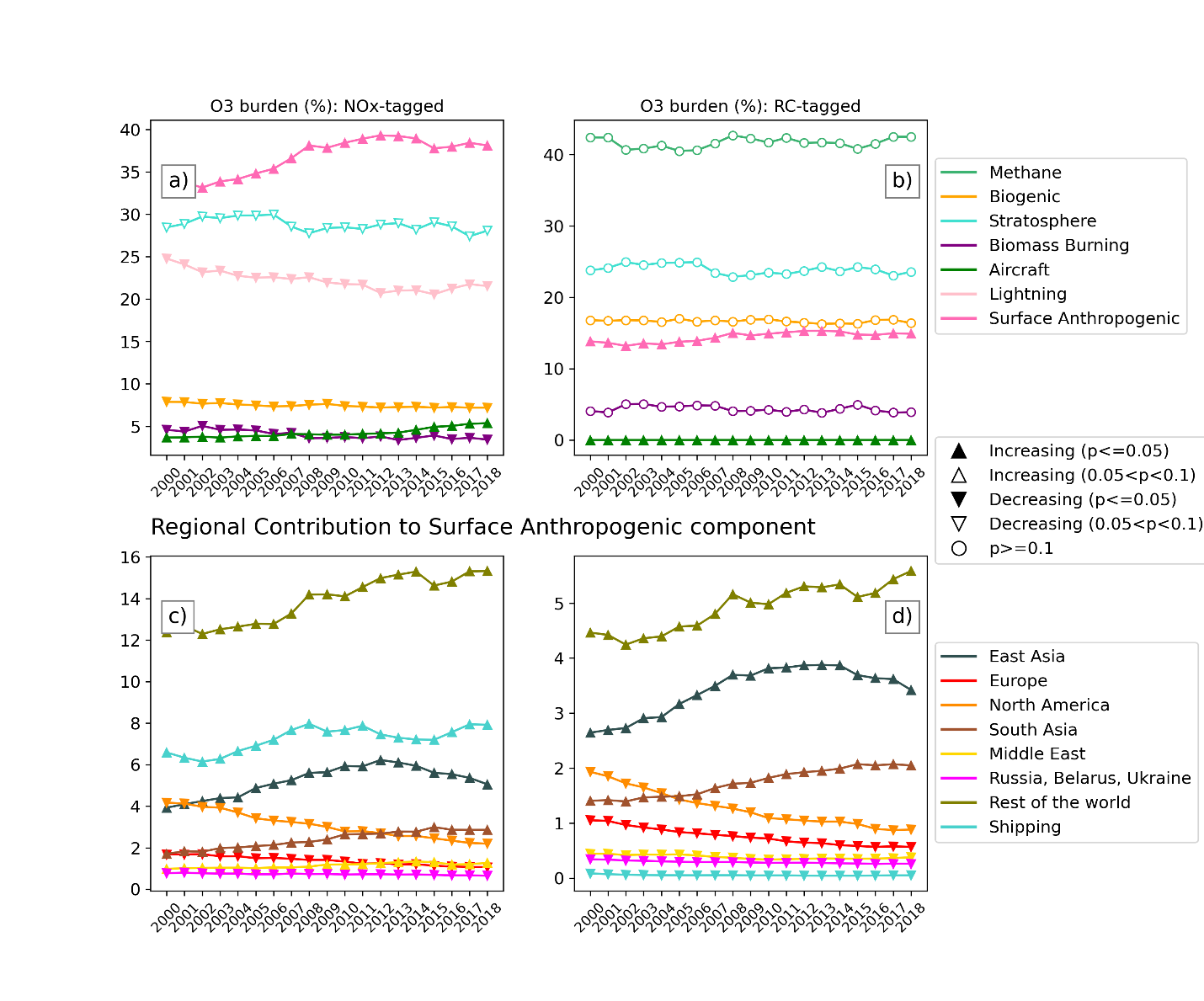
**Figure S3**: Relative contribution of each of the tagged emissions (in %) to the global annual total emissions (Black lines shown in Fig. 3 of the main text) of NOx (left panels) and reactive carbon (right panels).



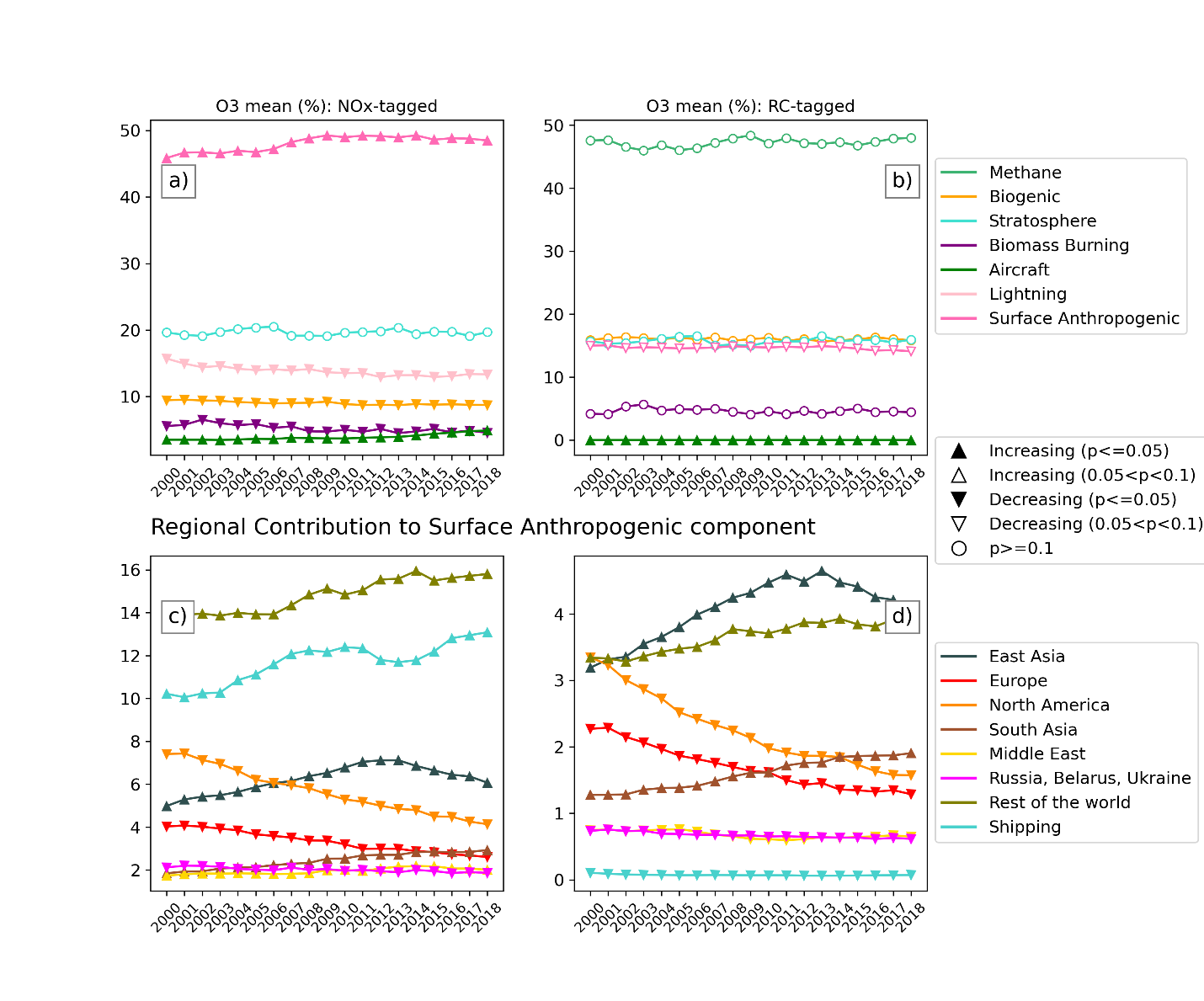
**Fig. S4**: Deviation from year 2000 in the zonal sum of annual anthropogenic emissions of a) NOx and b) NMRC emissions.



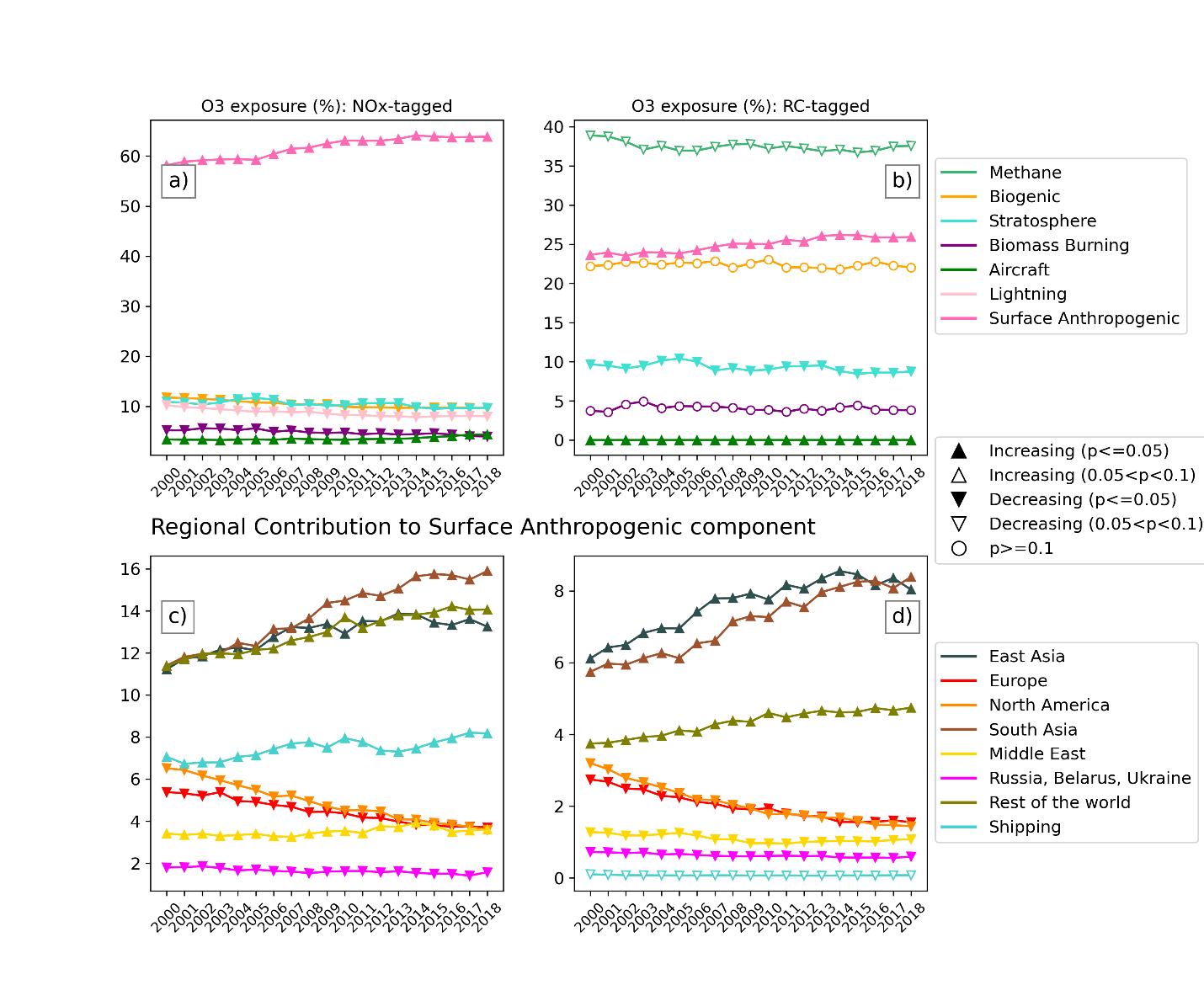
**Fig. S5**: Timeseries of free tropospheric NOx burden (above 700 hPa and within the troposphere; in TgN) over the 2000-2018 period. Shown are the total simulated free tropospheric NOx burden and the contributions from the NOx-tagged simulation. Bottom panels are the regional contributions to the total anthropogenic component (dark pink line) shown in the top panels. The symbols indicate the sign and certainty of trend in the plotted quantity.



**Figure S6**: Relative contribution of each of the tagged components (in %) to the total annual mean tropospheric ozone burden (Black lines shown in Fig. 5 of the main text) from the NOx-tagged (left panels) and RC-tagged (right panels) simulations.



**Figure S7**: Relative contribution of each of the tagged components (in %) to the total global annual area-weighted mean surface ozone (Black lines shown in Fig. 8 of the main text) from the NOx-tagged (left panels) and RC-tagged (right panels) simulations.



**Figure S8**: Relative contribution of each of the tagged components (in %) to the total global annual population-weighted mean surface ozone (Black lines shown in Fig. 9 of the main text) from the NOx-tagged (left panels) and RC-tagged (right panels) simulations.