The Impact of Climate Mitigation Measures on Near Term Climate Forcers

S. T. Turnock\textsuperscript{1}, S. Smith\textsuperscript{2} and F. M. O’Connor\textsuperscript{1}

\textsuperscript{1} Met Office Hadley Centre, Fitzroy Road, Exeter, Devon, UK;

\textsuperscript{2} Joint Global Change research Institute, Pacific Northwest National Laboratory, College Park, MD 20740, USA

Supplementary Material

Figure S1 – The range of global annual total emissions of a) CO\textsubscript{2}, b) NO\textsubscript{x}, c) CO, d) NMVOCs, e) SO\textsubscript{2}, f) BC and g) OC in the CMIP6 (blue) and CMIP5 (green) scenario. h) Shows the range of global CH\textsubscript{4} abundance in the CMIP5 and CMIP6 scenarios. Dashed lines show emission total at which future scenarios depart from the historical emissions in 2000 for CMIP5 and 2015 for CMIP6. The global emission totals used in this study from REF 2050 (red) and RCP4.5 (dark green) are shown by the stars. Annual carbon emissions from the Global Carbon budget (Le Quéré et al., 2018) have been included from 2000 to 2018 on a). The observed global CH\textsubscript{4} concentrations from NOAA (Dlugokencky, 2019) are included on h). Note that the high end of the SSP (blue) range in this figure represents futures with high air pollutant emissions and lower technological advancement, while the lower end of the range represents either strong climate policy scenarios (where in fossil fuel consumption is greatly reduced) or scenarios with strong air pollutant controls and higher technological advance (see Rao et al. 2017). Emission co-benefits in the SSP scenarios potentially span a wide range, but are generally smaller than the full range shown which represents bounding scenarios.
Figure S2: Regional definitions used in this study, previously defined in the Hemispheric Transport of Air Pollutants (HTAP) project.

Figure S3: Annual and seasonal mean differences in surface PM$_{2.5}$ components in the 2050s between RCP4.5 and REF. Stippling shows differences that are statistically significant at the 95% confidence interval. SO4 – Sulphate, OM – organic matter, BC – Black carbon.
Figure S4: Annual and seasonal mean differences in surface $O_3$ and $PM_{2.5}$ the 2050s between RCP4.5 and REF4.5 (co-emission change only). Stippling shows differences that are statistically significant at the 95% confidence interval.

Figure S5: Annual and seasonal mean differences in surface $O_3$ and $PM_{2.5}$ components in the 2050s between REF4.5 and REF (climate change only). Stippling shows differences that are statistically significant at the 95% confidence interval.
Figure S6: Annual and seasonal mean differences in tropospheric O$_3$ burden in the 2050s between RCP4.5 and REF. Stippling shows differences that are statistically significant at the 95% confidence interval. For these calculations the tropopause is defined as a O$_3$ concentration of 150 ppbv.

Figure S7: Annual and seasonal mean differences in aerosol optical depth (AOD) in the 2050s between RCP4.5 and REF. Stippling shows differences that are statistically significant at the 95% confidence interval.
Figure S8: Annual mean difference in a) CO$_2$, b) CH$_4$, c) aerosol and d) ozone all-sky effective radiative forcing (ERF) over the period 2050 to 2000 between the RCP4.5 2050 and REF 2050 simulations.
Table S1: The percentage difference in annual mean global and regional total air pollutant anthropogenic, shipping and biomass burning emissions for sulphur dioxide (SO$_2$), nitrogen oxides (NO$_x$ = NO + NO$_2$), carbon monoxide (CO), non-methane volatile organic compounds (NMVOCs), black carbon (BC), and organic carbon (OC). For global CH$_4$ percentage differences are calculated from changes in global abundances. Differences are between RCP4.5 2050s and BASE 2000, and RCP4.5 2050 and REF 2050.

| Region          | SO$_2$ | NO$_x$ | CO  | NMVOCs | BC | OC  | SO$_2$ | NO$_x$ | CO  | NMVOCs | BC | OC  |
|-----------------|--------|--------|-----|--------|----|-----|--------|--------|-----|--------|----|-----|
| Global          | -53    | -21    | -18 | -12    | -21| -25 | -23    | -9     | -20 | -16    | -24| -31 |
| Europe          | -82    | -65    | -74 | -31    | -62| -40 | -23    | -10    | -8  | -4     | -14| -14 |
| North Africa    | +164   | +63    | +105| +41    | +80| +80 | -29    | -12    | -3  | -1     | -9 | -2  |
| Southern Africa | +55    | +18    | +13 | +2     | +11| 0   | -28    | -9     | -11 | -10    | -13| -13 |
| South America   | -19    | -13    | -47 | -17    | -41| -54 | -12    | -17    | -49 | -19    | -38| -54 |
| North America   | -87    | -71    | -69 | -7     | -31| +18 | -27    | -26    | -67 | -49    | -60| -68 |
| Central America | -54    | -13    | -34 | -13    | -28| -48 | -16    | -14    | -39 | -18    | -35| -51 |
| Central Asia    | -29    | -6     | +24 | +20    | -19| -17 | -21    | -16    | -22 | -20    | -37| -40 |
| Russia Bel Ukr  | -73    | -43    | -1  | +5     | -32| -5  | -24    | -30    | -52 | -47    | -60| -46 |
| Middle East     | -60    | +1     | +19 | +4     | -10|     | -8     | -5     | -2  | -2     | -12| -6  |
| East Asia       | -80    | -53    | -51 | -46    | -57| -74 | -25    | -14    | -11 | -2     | -26| -15 |
| South Asia      | +41    | +105   | +74 | +31    | +43| +6  | -30    | -13    | +6  | +1     | -5 | +6  |
| South East Asia | -35    | -12    | -57 | -47    | -40| -69 | -27    | -15    | -10 | -8     | -17| -22 |
| Pacific Aus NZ  | -49    | -24    | -14 | -3     | -9 | -3  | -14    | 0      | +2  | 1      | +1 | +1  |
| Ocean           | -43    | -3     | -1  | +3     | -5 | -11 | -15    | +14    | +1  | +5     | -9 | -5  |

Global CH$_4$ +5 -24

Table S2: Details of model simulations conducted in this study quantify effective radiative forcing for a 30 year averaging period

| Simulation    | Time Period | Air Pollutant Emissions                                                                 | SST/Sea Ice | Radiation Interactions |
|---------------|-------------|----------------------------------------------------------------------------------------|-------------|------------------------|
| O$_3$ ERF BASE | 2000s       | All from historical dataset in 2000s                                                   | 2000s       | O$_3$ only             |
| O$_3$ ERF RCP4.5 | 2050s     | All 2000s except O$_3$ precursor emissions and CH$_4$ from RCP4.5 in 2050              | 2000s       | O$_3$ only             |
| O$_3$ ERF REF  | 2050s       | All 2000s except O$_3$ precursor emissions and CH$_4$ from REF in 2050                 | 2000s       | O$_3$ only             |
| CH$_4$ ERF BASE | 2000s     | All from historical dataset in 2000s                                                   | 2000s       | CH$_4$ only            |
| CH$_4$ ERF RCP4.5 | 2050s    | All 2000s except CH$_4$ from RCP4.5 in 2050                                            | 2000s       | CH$_4$ only            |
| CH$_4$ ERF REF  | 2050s       | All 2000s except CH$_4$ from REF in 2050                                               | 2000s       | CH$_4$ only            |
| AERO ERF BASE  | 2000s       | All from historical dataset in 2000s                                                   | 2000s       | ari and aci only*      |
| AERO ERF RCP4.5 | 2050s     | All 2000s except aerosol precursors from RCP4.5 in 2050                                 | 2000s       | ari and aci only*      |
| AERO ERF REF   | 2050s       | All 2000s except for aerosol precursors from REF in 2050                                | 2000s       | ari and aci only*      |
| BC ERF RCP4.5  | 2050s       | All 2000s except for Black Carbon aerosol precursors from RCP4.5 in 2050                | 2000s       | ari and aci only*      |
| BC ERF REF     | 2050s       | All 2000s except for Black Carbon precursors from REF in 2050                           | 2000s       | ari and aci only*      |
| CO$_2$ ERF RCP4.5 | 2050s | All 2000s except CO$_2$ from RCP4.5 in 2050                                           | 2000s       | All                    |
| CO$_2$ ERF REF | 2050s       | All 2000s except CO$_2$ from REF in 2050                                               | 2000s       | All                    |

* ari – aerosol radiation interactions, aci – aerosol cloud interactions
Table S3 – Annual mean change in regional population weighted surface O$_3$ and PM$_{2.5}$ concentrations between different simulations to calculate the total impact of carbon mitigation measures (RCP4.5 – REF), and those solely from co-emissions (RCP4.5 – REF4.5) and climate (REF4.5 – REF).

| Region                      | RCP4.5 - REF | RCP4.5 – REF4.5 | REF4.5 – REF | RCP4.5 - REF | RCP4.5 – REF4.5 | REF4.5 – REF |
|-----------------------------|--------------|-----------------|--------------|--------------|-----------------|--------------|
| Europe                      | -1.5         | -1.6            | +0.1         | -0.2         | -0.2            | 0.0          |
| North Africa                | -1.1         | -1.6            | +0.6         | +0.4         | -1.7            | -2.1         |
| Southern Africa             | -1.6         | -1.9            | +0.3         | +0.4         | -0.4            | -0.8         |
| South America               | -1.1         | -1.1            | 0.0          | 0.0          | -0.1            | 0.0          |
| North Pole                  | -1.7         | -1.7            | +0.1         | -0.1         | -0.1            | 0.0          |
| North America               | -2.1         | -2.1            | +0.1         | -0.3         | -0.3            | 0.0          |
| Central America             | -1.5         | -1.8            | +0.2         | -0.1         | -0.3            | -0.2         |
| Central Asia                | -1.9         | -2.0            | +0.1         | -2.4         | -2.3            | 0.0          |
| Russia Belarus Ukraine      | -1.6         | -1.7            | +0.1         | -0.6         | -0.7            | -0.1         |
| Middle East                 | -1.8         | -2.5            | +0.7         | -0.1         | -1.6            | -1.5         |
| East Asia                   | -2.0         | -2.3            | +0.3         | -1.8         | -1.5            | +0.4         |
| South Asia                  | -1.4         | -1.7            | +0.3         | -1.2         | -1.3            | -0.1         |
| South East Asia             | -1.8         | -1.8            | 0.0          | -0.3         | -0.5            | -0.2         |
| Pacific Australia New Zealand | -0.3       | -0.6            | +0.4         | 0.0          | -0.3            | -0.3         |
| Ocean                       | -1.4         | -1.8            | +0.4         | -0.2         | -0.4            | -0.3         |
| Global                      | -1.6         | -1.9            | +0.3         | -0.5         | -0.9            | -0.3         |
Table S4 – Seasonal mean change in regional population weighted surface O₃ and PM₂.₅ concentrations between different simulations to calculate the total impact of carbon mitigation measures (RCP4.5 – REF), and those solely from changes in co-emissions (RCP4.5 – REF4.5) and climate (REF4.5 – REF).

| Region | DJF | MAM | JJA | SON | DJF | MAM | JJA | SON | DJF | MAM | JJA | SON | DJF | MAM | JJA | SON | DJF | MAM | JJA | SON |
|--------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| EUR    | -1.6 | -3.2 | -1.1 | +0.2 | -1.9 | -3.5 | -1.2 | -0.2 | +0.4 | +0.3 | +0.1 | -0.8 | +0.3 | -0.2 | -0.3 | -0.4 | -0.5 | -1.0 | +0.6 | +0.1 | +0.2 |
| NAF    | -0.1 | -2.1 | -1.0 | +0.4 | -1.9 | -2.8 | -1.3 | +0.4 | +0.8 | +0.8 | +0.3 | +1.9 | +2.1 | -1.2 | -1.2 | -1.9 | -1.6 | -1.9 | -1.5 | +3.8 | +3.8 | +0.7 | +0.3 |
| SAF    | -2.1 | -0.9 | -1.7 | -1.4 | -2.4 | -1.5 | -2.0 | -1.6 | +0.2 | +0.6 | +0.3 | +1.3 | +2.2 | -0.7 | -1.2 | +1.0 | -0.3 | -1.0 | -1.2 | +0.3 | +2.5 | +0.4 | +0.1 |
| SAM    | -0.6 | -0.5 | -1.9 | -1.2 | -0.6 | -0.6 | -1.9 | -1.2 | 0.0  | +0.1 | 0.0  | 0.0  | 0.0  | 0.0  | 0.0  | 0.0  | 0.0  | 0.0  | 0.0  | 0.0  | 0.0  | 0.0  | -0.2 |
| NOP    | -0.4 | -1.8 | -3.5 | -1.0 | -0.8 | -2.1 | -3.4 | -0.6 | +0.4 | +0.3 | 0.0  | -0.1 | -0.1 | -0.2 | -0.1 | -0.2 | -0.2 | -0.1 | 0.0  | +0.1 | +0.1 | -0.1 | -0.1 |
| NAM    | -0.3 | -2.6 | -3.6 | -1.6 | -0.5 | -2.6 | -3.9 | -1.5 | +0.2 | 0.0  | +0.2 | -0.1 | -0.3 | -0.4 | -0.2 | -0.2 | -0.3 | -0.3 | -0.3 | 0.0  | 0.0  | 0.1  | -0.0 |
| MCA    | -1.1 | -3.1 | -0.9 | -1.1 | -1.4 | -3.2 | -1.4 | -1.1 | +0.3 | +0.1 | +0.5 | +0.0 | -0.4 | +0.1 | +0.5 | -0.5 | -0.4 | -0.4 | -0.4 | 0.0  | 0.0  | -1.0 | -0.3 |
| CAS    | -0.3 | -2.2 | -3.6 | -1.4 | 0.0  | -2.1 | -4.2 | -1.5 | -0.3 | -0.1 | +0.6 | +0.1 | -2.7 | -2.3 | -1.9 | -2.6 | -2.2 | -2.0 | -2.3 | -2.8 | -0.5 | +0.4 | +0.2 |
| RBU    | +0.1 | -1.9 | -3.7 | -1.0 | +0.1 | -2.0 | -3.7 | -1.2 | 0.0  | +0.1 | +0.1 | +0.2 | -0.4 | -0.5 | -1.0 | -0.6 | -0.5 | -0.7 | -0.9 | -0.6 | +0.1 | +0.3 | -0.1 | 0.0  |
| MDE    | -1.3 | -2.1 | -2.3 | -1.7 | -1.2 | -2.8 | -3.8 | -2.2 | -0.1 | +0.7 | +1.5 | +0.5 | +0.3 | -0.2 | -0.5 | 0.0  | -1.6 | -2.7 | -1.5 | -0.6 | +1.8 | +2.6 | +0.9 | +0.6 |
| EAS    | +0.1 | -3.2 | -3.2 | -1.6 | 0.0  | -3.5 | -3.3 | -2.2 | +0.2 | +0.3 | +0.1 | +0.6 | -1.7 | -2.4 | -1.1 | -2.0 | -0.9 | -1.8 | -1.1 | -2.0 | -0.8 | -0.6 | +0.1 | 0.0  |
| SAS    | -0.5 | -1.8 | -1.9 | -1.5 | 0.0  | -2.4 | -2.6 | -1.9 | -0.5 | +0.6 | +0.7 | +0.4 | -2.4 | -0.2 | -0.6 | -2.1 | -2.1 | -1.0 | -0.1 | -2.2 | -0.3 | +1.1 | -0.5 | +0.1 |
| SEA    | -2.0 | -1.8 | -1.9 | -1.6 | -2.1 | -1.4 | -1.8 | -2.0 | +0.1 | -0.3 | -0.1 | +0.4 | -0.4 | -0.3 | -0.4 | -0.2 | -0.5 | -0.4 | -0.4 | -0.7 | +0.1 | +0.1 | 0.0  | +0.5 |
| PAN    | +0.3 | +0.1 | -0.6 | -0.8 | -0.1 | -0.2 | -0.8 | -1.3 | +0.4 | +0.3 | +0.3 | +0.5 | -0.1 | 0.0  | +0.1 | 0.0  | -0.3 | -0.1 | -0.5 | -0.1 | +0.1 | +0.1 | +0.6 | +0.1 |
| OCN    | -1.3 | -1.5 | -1.6 | -1.2 | -1.4 | -2.1 | -2.1 | -1.7 | +0.1 | +0.5 | +0.5 | +0.6 | -0.2 | +0.3 | -0.2 | -0.6 | -0.2 | -0.3 | -0.4 | -0.7 | +0.1 | +0.6 | +0.2 | +0.1 |
| Global | -0.9 | -1.8 | -2.2 | -1.4 | -0.9 | -2.2 | -2.6 | -1.7 | 0.0  | +0.4 | +0.4 | +0.3 | -0.6 | +0.2 | -0.6 | -1.2 | -0.6 | -0.8 | -0.7 | -1.3 | +0.1 | +1.0 | +0.1 | +0.1 |

EUR – Europe, NAF – North Africa, SAF – South Africa, SAM – South America, NOP – North Pole, NAM – North America, MCA – Central America, CAS – Central Asia, RBU – Russia Belarus Ukraine, MDE – Middle East, EAS – East Asia, SAS – South Asia, SEA – South East Asia, PAN – Pacific Australia New Zealand, OCN – Ocean, South Pole
Table S5 – Ozone budget terms for each simulation

| Scenario      | Production | Loss | Net Production | Deposition | Inferred Strat-Trop exchange | Lifetime (Days) | Burden (Tg) |
|---------------|------------|------|----------------|------------|------------------------------|-----------------|-------------|
| BASE          | 1710       | 1460 | 256            | 336        | 79                           | 60              | 300         |
| RCP4.5        | 1770       | 1560 | 202            | 320        | 118                          | 58              | 305         |
| REF           | 1890       | 1680 | 217            | 336        | 119                          | 56              | 313         |
| REF4.5        | 1860       | 1630 | 224            | 339        | 115                          | 58              | 316         |
| RCP4.5 – REF  | -120       | -120 | -15            | -16        | -1                           | 2               | -8          |
| RCP4.5 – REF4.5 (Emissions Only) | -90 | -70 | -22 | -19 | +3 | 0 | -11 |
| REF4.5 – REF (Climate Only) | -30 | -50 | +7 | +3 | -4 | 2 | +3 |

Table S6: Population weighted mean number of days exceeding the O₃ and PM₂.₅ World Health Organization Air Quality Guideline values (2005) within each region for the BASE 2000, RCP4.5 2050 and REF 2050 scenarios.

| Region     | O₃ BASE 2000 | O₃ RCP4.5 2050 | O₃ REF 2050 | O₃ RCP4.5 2050 – REF 2050 | PM₂.₅ BASE 2000 | PM₂.₅ RCP4.5 2050 | PM₂.₅ REF 2050 | PM₂.₅ RCP4.5 2050 – REF 2050 |
|------------|--------------|----------------|--------------|---------------------------|-----------------|-------------------|-----------------|-----------------------------|
| Global     | 164          | 165            | 185          | -20                       | 90              | 92                | 98              | -6                          |
| Europe     | 121          | 96             | 123          | -27                       | 3               | 2                 | 2               | 0                           |
| Africa     | 219          | 238            | 250          | -12                       | 193             | 242               | 244             | -2                          |
| Southern   | 74           | 136            | 157          | -21                       | 110             | 133               | 135             | -2                          |
| Africa     | 1            | 0              | 3            | -3                        | 1               | 3                 | 1               | +2                          |
| South      | 0            | 0              | 0            | 0                         | 0               | 0                 | 0               | 0                           |
| America    | 149          | 46             | 76           | -30                       | 0               | 1                 | 1               | 0                           |
| North      | 103          | 92             | 120          | -28                       | 11              | 16                | 13              | +3                          |
| Central    | 167          | 162            | 182          | -20                       | 34              | 43                | 56              | -13                         |
| Asia       | 70           | 32             | 54           | -22                       | 1               | 0                 | 1               | -1                          |
| Russia Bel | 289          | 293            | 301          | -8                        | 110             | 101               | 104             | -3                          |
| Middle     | 174          | 111            | 158          | -47                       | 158             | 21                | 32              | -11                         |
| East Asia  | 274          | 318            | 321          | -3                        | 160             | 195               | 210             | -15                         |
| Asia Pacific Aus | 117 | 114          | 142          | -28                       | 46              | 24                | 25              | -1                          |
| NZ         | 0            | 0              | 0            | 0                         | 0               | 0                 | 0               | 0                           |
| Region                  | Aerosols (aci) |   | Aerosols (ari) |   |
|-------------------------|----------------|---|----------------|---|
|                         | RCP4.5         | REF | Diff           | RCP4.5 | REF | Diff |
| Global                  | 0.93 +/- 0.11  | 0.66 +/- 0.10  | +0.26  | 0.47 +/- 0.06  | 0.37 +/- 0.05  | +0.10 |
| Europe                  | 2.94 +/- 0.36  | 2.87 +/- 0.34  | +0.06  | 1.98 +/- 0.20  | 1.55 +/- 0.17  | +0.43 |
| North Africa            | 0.09 +/- 0.22  | 0.40 +/- 0.26  | -0.31  | 0.14 +/- 0.18  | 0.44 +/- 0.24  | -0.30 |
| Southern Africa         | -0.19 +/- 0.08 | -0.61 +/- 0.10 | +0.42  | 0.10 +/- 0.11  | -0.07 +/- 0.06 | +0.17 |
| South America           | 0.27 +/- 0.09  | -0.08 +/- 0.07 | +0.34  | 0.32 +/- 0.09  | 0.08 +/- 0.08  | +0.24 |
| North Pole              | 1.17 +/- 0.46  | 0.68 +/- 0.26  | +0.49  | 0.52 +/- 0.18  | 0.27 +/- 0.14  | +0.26 |
| North America           | 2.82 +/- 0.39  | 2.09 +/- 0.35  | +0.73  | 1.31 +/- 0.18  | 1.14 +/- 0.16  | +0.18 |
| Central America         | 0.85 +/- 0.10  | 0.66 +/- 0.15  | +0.19  | 0.76 +/- 0.10  | 0.60 +/- 0.07  | +0.16 |
| Central Asia            | 1.50 +/- 0.21  | 1.04 +/- 0.21  | +0.46  | 1.06 +/- 0.37  | 1.00 +/- 0.21  | +0.06 |
| Russia Belarus Ukraine  | 2.91 +/- 0.51  | 1.84 +/- 0.46  | +1.07  | 1.89 +/- 0.32  | 1.02 +/- 0.31  | +0.87 |
| Middle East             | 1.02 +/- 0.21  | 1.26 +/- 0.19  | -0.24  | 1.12 +/- 0.20  | 1.31 +/- 0.20  | -0.19 |
| East Asia               | 2.09 +/- 0.39  | 1.98 +/- 0.24  | +0.11  | 2.13 +/- 0.18  | 2.09 +/- 0.16  | +0.04 |
| South Asia              | 0.44 +/- 0.20  | -0.19 +/- 0.17 | +0.63  | 0.51 +/- 0.25  | -0.13 +/- 0.20 | +0.64 |
| South East Asia         | 0.82 +/- 0.14  | 0.46 +/- 0.17  | +0.36  | 0.79 +/- 0.14  | 0.56 +/- 0.24  | +0.23 |
| Pacific Australia New Zealand | 0.66 +/- 0.14 | 0.53 +/- 0.11  | +0.13  | 0.49 +/- 0.19  | 0.50 +/- 0.13  | -0.01 |
| South Pole              | -0.01 +/- 0.08 | -0.01 +/- 0.09 | -0.01  | -0.02 +/- 0.06 | -0.02 +/- 0.08 | 0.00 |
| Ocean                   | 0.89 +/- 0.06  | 0.65 +/- 0.06  | 0.24   | 0.35 +/- 0.04  | 0.28 +/- 0.04  | +0.06 |
S1 Scenario Description

REF is a future pathway of intermediate economic development and population growth with no climate mitigation measures included, where total radiative forcing is nearly 7 W m\(^{-2}\) by 2100 (Thomson et al., 2011). Assumptions under REF include increasing energy and food consumption and decreased forest cover, with fossil fuels dominating the global energy supply. Controls to air pollution emissions are applied based on regional economic development (Smith et al., 2011). RCP4.5 is based on the same pathway of population growth and economic development as REF but applies a carbon price to emissions across all sectors (including terrestrial) to stabilise carbon dioxide (CO\(_2\)) concentrations at 525 ppm in 2100 and achieve a radiative forcing of 4.5 W m\(^{-2}\) (Thomson et al., 2011). Global energy demand is reduced slightly in RCP4.5, fossil fuels are replaced by renewables and there is an increase in bioenergy crops and biofuels that decrease GHG concentrations. Carbon capture and storage is applied to both bio-energy and fossil fuels to reduce total carbon emissions.

S2 Model Description

We have used the Global Atmosphere 7.0 (GA7.0; Walters et al., 2017) configuration of the Hadley Centre Global Environment Model version 3 (HadGEM3 - (Hewitt et al., 2011)) to simulate present and future air pollutant concentrations. GA7.0 includes the aerosol microphysics scheme GLOMAP-mode (Mann et al., 2010; Mulcahy et al., 2018) from the United Kingdom Chemistry and Aerosol (UKCA) model which is coupled to chemical mechanisms for both the troposphere (O’Connor et al., 2014) and stratosphere (Morgenstern et al., 2009). The model is used in an atmosphere-only configuration at a horizontal resolution of 1.875\(^{\circ}\) by 1.275\(^{\circ}\) (~140 km at mid-latitudes). It is set up to perform timeslice experiments centred on the 2000s or the 2050s, where prescribed concentrations of well-mixed GHGs (including CH\(_4\)), ozone depleting substances, sea ice (SI) distributions and sea surface temperatures (SSTs) are used from each scenario. The land-use and vegetation distribution is kept constant between the 2000s and the 2050s (i.e. no change in land-use is assumed which fixes emission of biogenic volatile organic compounds (BVOCs)) but other natural sources e.g. dust and sea salt are interactive and vary according to the climate state.

The chemical reactions within HadGEM3-UKCA for both the troposphere and stratosphere are described in Morgenstern et al., (2009) and O’Connor et al., (2014). Tropospheric chemistry reactions are included for odd-oxygen (Ox), nitrogen (NOy), hydrogen (HOx = OH + HO\(_2\)), CO and methane, as well as other short chain non-methane volatile organic compounds (VOCs). It includes the Mainz Isoprene Mechanism (Pöschl et al., 2000; O’Connor et al., 2014). The chemistry scheme has been extended to include additional reactions related to aerosols for sulphur (Mann et al., 2010) and monoterpenes (Spracklen et al., 2006). Online photolysis rates are calculated using the Fast-JX photolysis scheme (Telford et al., 2013). Physical loss mechanisms of dry and wet deposition are included within the model for gas-phase and aerosol species.

Aerosols are included the model in the form of a two-moment modal scheme (Mann et al., 2010; Mulcahy et al., 2018) with five log-normal modes and four chemical components (sulphate, black carbon, organic matter, sea-salt). Secondary organic aerosol (SOA) is included at a fixed 13% yield rate from products of monoterpane oxidation (Spracklen et al., 2006) but is increased by a factor of 2 to represent SOA formed from other VOCs e.g. isoprene. Ammonium nitrate is not currently included within the aerosol scheme. Natural aerosol emissions of dust, sea-salt, dimethyl sulphide (DMS), biogenic VOCs and tropospheric volcanoes are included within the model. Mineral dust aerosol is simulated using the 6 bin mass only scheme of Woodward, (2001) and depends mainly on wind speed and soil moisture. Emissions of sea-salt are based on the surface wind speed using the parameterisation of (Gong, 2003). Emission of DMS are calculated using a wind-speed dependent air-sea exchange parameterisation (Liss and Merlivat, 1986) based on monthly sea-water concentrations fields (Kettle and Andreae, 2000). Mineral dust, sea-salt and DMS emissions are interactive in the model.
and vary depending on the meteorology simulated in the model. Whereas, prescribed monthly mean fields are used to represent biomass burning, monoterpene emissions from vegetation (Guenther et al., 1995) and tropospheric volcanoes (Andres and Kasgnoc, 1998).

Changes to the Earth’s radiative balance are calculated using the SOCRATES radiative transfer scheme within the model (Edwards and Slingo, 1996; Manners et al., 2015; Walters et al., 2017). A single-call radiation configuration is used in this study to calculate an effective radiative forcing (ERF) over a 30 year period in accordance with the definition in Forster et al., (2016). ERFs for the individual components CO₂, CH₄, O₃ and aerosols are calculated using paired experiments where SSTs and sea ice concentrations are held fixed at year 2000 values but air pollutant precursor emissions or abundances are varied between 2000 and 2050 in accordance with the values in the RCP4.5 and REF scenarios (see Table 1). In these simulations the radiation scheme only sees perturbations from the individual components to diagnose a direct ERF for CH₄, O₃ and aerosols. The ERF for CH₄ and aerosols comes from changes in CH₄ abundance and aerosol precursor emissions, whereas for the O₃ ERF this is from changes in O₃ precursor emissions and CH₄ abundance together. This configuration allows changes in the radiation balance, including rapid adjustment to clouds from aerosols, to feedback back onto the meteorology. Both aerosol radiation (ari) and aerosol cloud (aci) interactions are included within this definition of aerosol ERF.

Population weighted mean concentrations of O₃ and PM₂.₅ have been calculated by re-gridding HadGEM3-UKCA output onto population datasets for 2000 and 2050. Year 2000 population data was obtained from the Gridded Population of the World Version 4 in March 2018 (GPWv4 - http://sedac.ciesin.columbia.edu/data/collection/gpw-v4/documentation). For 2050 we downloaded population projections from the International Futures (IFs) project (http://www.ifs.du.edu/ifs/index.aspx) in September 2018, using IFs version 7.31. These datasets were used as they provide global grid maps of historical and future population change that can be used in the calculation of population weighted means.

**S3 Model Evaluation**

Surface PM₂.₅ data is provided from three main monitoring networks located in North America (Interagency Monitoring of Protected Visual Environments, IMPROVE), Europe (European Monitoring and Evaluation Programme, EMEP) and Asia/Pacific (Asia-Pacific Aerosol Database, A-PAD). There is generally not a long record of PM₂.₅ observations, with most measurements starting post-2000 (apart from the IMPROVE network). The TOAR database provides globally gridded maps of all surface observations available between 1970 and 2015 for model evaluation purposes. The observations are predominately based in North America and Europe with other sites located in East Asia and at remote locations around the globe. Observations of surface O₃ that are classified as being ‘rural’ in the TOAR database are considered representative of background ozone levels, which dominate radiative forcing from tropospheric ozone, and are more appropriate to compare to simulated values obtained at the horizontal resolution of the model. Observed values have been compared to simulated concentrations from the lowest model level that have been linearly interpolated to the corresponding measurement location.

The comparison of surface O₃ concentrations across regions shows that the model slightly overpredicts annual mean concentrations in the northern hemisphere (by a factor of 1 to 1.5) and underpredicts them (by a factor of ~1.5) in the southern hemisphere (apart from Southern Africa, Australia and New Zealand where there is a slight overprediction), in a similar way to other global models (Young et al., 2018). A clear seasonal discrepancy is highlighted in the surface O₃ model biases, with the model underestimating wintertime and overestimating summertime observations. These differences could be due to the coarse model resolution as well as errors in emissions, the representation of volatile organic compounds in the chemical mechanism and/or uncertainties in loss processes for O₃ (particularly dry
The comparison of the Daily Maximum 8 hour mean values (DM8H) at a monthly mean timescale shows a consistent model underprediction in most regions. This implies that the model under-represents the elevated daily surface \(O_3\) concentrations during pollution episodes, which is expected when using a global model driven by decadal mean monthly emissions. This means that simulated values at this higher temporal resolution are likely to be conservative.

The NMBFs for PM\(_{2.5}\) show that HadGEM3-UKCA generally underpredicts annual mean surface PM\(_{2.5}\) concentrations across the regions for which observations are available. Summertime PM\(_{2.5}\) concentrations tend to be better represented by the model but there is a consistent low model bias in wintertime possibly due to the absence and/or under-representation of emission sources (e.g. local dust sources) and aerosol formation processes (e.g. absence of nitrate aerosols or underrepresentation of secondary organic aerosol formation – see section S2 of the supplementary). The model observational biases in HadGEM3-UKCA for surface PM\(_{2.5}\) are similar to those identified before (Turnock et al., 2015) and in other global and regional models (Glotfelty et al., 2017; Solazzo et al., 2017; Im et al., 2018). There is also a slight low model bias in the daily mean PM\(_{2.5}\) evaluation across most regions. However, the low model bias becomes particularly large when elevated daily mean PM\(_{2.5}\) concentrations are observed across South and East Asia, although the number of observation sites used in this evaluation is quite low. This indicates that the model, like for \(O_3\), under-represents elevated daily mean PM\(_{2.5}\) concentrations that are indicative of pollution episodes. Like for surface \(O_3\), the magnitude of similar daily events in the future will also be underestimated by the model but the simulated values will provide a conservative estimate of the change.

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