**Air Quality and Climate Connections**

**Supplemental Material**

Arlene M. Fiore

Department of Earth and Environmental Sciences and Lamont-Doherty Earth Observatory

of Columbia University, Palisades, NY, 10964

Vaishali Naik

University Corporation for Atmospheric Research (UCAR) & NOAA Geophysical Fluid Dynamics Laboratory, Princeton, NJ 08540

Eric M. Leibensperger

Center for Earth and Environmental Science, SUNY Plattsburgh, Plattsburgh, NY 12901

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**SUPPLEMENTAL TEXT**

**S1: Emissions**

*Historical emissions.* In preparation for the IPCC AR5 report (IPCC, 2013a), a consistent set of historical emissions of air pollutants (NOx, CO, NMVOCs, SO2, NH3, OC, BC, and CH4) was developed for use in CCMs, and for generating NTCF atmospheric constituent distributions for use in GCMs. Lamarque et al. (2010) combined several existing regional and global anthropogenic and biomass burning emission inventories for the year 2000 with long-term global emission datasets to produce monthly, sectoral, gridded dataset covering the 1850-2000 period, referred to as the Atmospheric Chemistry-Climate Model Intercomparison Project (ACCMIP) inventory.

Granier et al. (2011) compared and evaluated several emission inventories, including the ACCMIP dataset, over the 1980-2010 period – a time period covered at least partially by most of the inventories (e.g., Figures S1 and S2). At the global scale, there is consensus across inventories of anthropogenic emissions of CO, NOx, and SO2 for most years and of BC up to 2000, but this agreement does not indicate low uncertainty. Global inventories differ at the regional scale, especially for CO, BC and SO2 (e.g., on average a factor of 2 differences across inventories for BC emissions in India over the 1980-2005 period).

For the U.S.A., Figure S1 shows a difference of more than a factor of two between the highest BC emissions (J&L inventory) and the lowest (Bond inventory) in the 1980s, but all inventories agree to within 50% in 2000. For CO, emissions provided by the EPA show large differences versus other inventories prior to 1995. U.S. emissions of BC, CO, and SO2 were declining at the start of the period, and both SO2 and NOx decreases accelerated in the mid to late 1990s (Figure S1). These trends are consistent with an inventory built upon the latest activity and emission control data (Xing et al., 2013).

Substantial discrepancies remain in our understanding of the global CH4 emission budget. Constraints exist on the total CH4 sink by tropospheric oxidation (e.g., Montzka et al., 2011), and the atmospheric burden and growth rate are well approximated by the global surface network of CH4 measurements (Dlugokencky et al., 2011 and references therein). Together, knowledge of the CH4 oxidation sink, atmospheric burden, and growth rates provide fairly strong constraints on total global CH4 emissions. The sectoral distribution of emissions, however, is poorly understood, with large discrepancies remaining in bottom-up and top-down estimates (Kirschke et al., 2013). Uncertainties in the global emissions of NH3, originating predominantly from agriculture (Bouwman et al., 1997; Paulot et al., 2014), have not been thoroughly analyzed; Lamarque et al. (2013a) note possible errors in the NH3 ACCMIP inventory, including for the U.S.A.

*Future emissions.* Early efforts to project global anthropogenic air pollutant emissions, specifically NOx, CO, NMVOCs, and SO2, involved developing gridded emissions following the IPCC Special Report on Emission Scenarios (SRES) scenarios (Nakicenovic et al., 2000). The SRES scenarios encompassed several specific socioeconomic storylines with each one simulated by a different “marker” Integrated Assessment Model (IAM). The SRES scenarios covered a wide range of demographic, economic, and technological driving forces of emissions but did not explicitly include any climate policy. Emissions of GHGs including ozone depleting substances, and short-lived air pollutants, with the exception of carbonaceous aerosols, were provided globally and within four world regions (Nakicenovic et al., 2000). Individual modeling groups gridded the SRES emissions or made largely ad-hoc assumptions regarding future trajectories of unspecified species, which resulted in diverse trends and spatial distributions of projected air pollutant concentrations and forcings (Shindell et al., 2008; CCSP 2008).

The SRES scenarios were developed by the IPCC and then sequentially applied in GCM simulations. The systematic development of future scenarios for air pollutants and GHGs in a consistent manner for coordinated international climate modeling first occurred in the early 2010s (van Vuuren et al., 2011; Amman et al., 2013). In that effort, the Representative Concentration Pathway (RCP) emission trajectories were designed to target four levels of radiative forcing (RF) in the year 2100 and were developed by the research community in a ‘parallel process’ that enabled GCMs to project future climate change while the IAMs developed socioeconomic trajectories consistent with the RCPs (Moss et al., 2010). Unlike the SRES scenarios, the RCPs explicitly explore the impact of different potential approaches to mitigating climate change in addition to a no-climate-policy scenario (RCP8.5), and span the full range of published climate scenarios. Figure S2 shows the emissions for individual air pollutant species under the four RCP scenarios described in the main text.

**S2:** **Climate Change**

*Observations: Surface Temperature and Precipitation.* Global mean surface temperature has increased by +0.85 °C (linear trend with 95% confidence intervals of 0.65 to 1.05 °C) from 1880 to 2012, and the number of warm days and nights has increased since about 1950, while the number of cold days and nights has decreased (Hartmann et al., 2013). The U.S. National Climate Assessment (Melillo et al., 2014) reports that U.S. average temperature has increased by +0.7 to +1.1°C (+1.3 to +1.9 °F) since 1895, with most of the increase occurring since 1970. Increases in temperature are widespread across the United States with the notable exception of the Southeastern and South-Central regions where temperature measurements indicate either a cooling or an absence of waming over the 20th Century, referred to as the “warming hole” (Melillo et al., 2014; Hartmann et al., 2013). Proposed mechanisms involve regional circulation changes and interactions with the hydrologic cycle resulting from specific patterns of sea surface temperature variability (Meehl et al., 2012) or from regional aerosol forcing (Leibensperger et al. 2012a, Yu et al., 2014).

On average over mid-latitude land areas, precipitation has increased since the middle of the 20th century; heavy precipitation has increased in frequency or intensity over North America (≥66% probability), with heavier precipitation events increasing over Central North America (Hartmann et al., 2013) particularly over the Central and Midwestern U.S. regions (Groisman et al., 2005). Annual mean precipitation is higher in 1991-2012 versus 1901-1960 over much of the United States, however there are regional differences with increases in the Central and Northeast regions and a combination of areas with increases or decreases in the Southeast and Southwest U.S. (Melillo et al., 2014).

*Model Evaluation.* Taken together (i.e., averaging across multiple models), current generation GCMs and CCMs capture the observed changes from pre-industrial to present-day, including the more rapid globally averaged warming in the second half of the 20th century, cooling induced by volcanic eruptions, the spatial patterns of temperature change (global spatial pattern correlation of r ~ 0.99), and changes in the frequency of extreme warm and cold days (Flato et al., 2013). The observed temperature change over the 20th century, however, is produced in GCMs/CCMs with various combinations of RF from GHGs and NTCFs (Kiehl, 2007), which implies a wide range of future warming in response to GHGs as aerosols are reduced under air pollution control programs. Simulating regional precipitation remains a challenge (model vs. observed global spatial pattern correlation r = 0.82), with models generally underestimating the sensitivity of extreme precipitation to temperature variability (Flato et al., 2013). Major uncertainties remain in simulating clouds, including their interactions with aerosols (Flato et al., Boucher et al., 2013). GCMs generally simulate more intense and frequent extreme precipitation events in response to rising greenhouse gases (Flato et al., 2013) but with regional variations due to internal variability and interactions with aerosols (Kirtman et al., 2013).

*21st century projections of climate change.* Three major sources of uncertainty surround climate change projections. First, the specific scenario for GHGs and other climate-relevant species (the “climate forcing” trajectory) contributes to uncertainty. Consideration of multiple possible trajectories for GHGs and climate forcing agents produced by IAMs based on assumptions about the driving forces (Nakicenovic et al., 2000; Moss et al. 2010, van Vuuren et al. 2011), provides a measure of the uncertainty associated with the choice of scenario. Second, the simulated climate impact, global or regional, of an imposed forcing somewhat depends on the particular model setup used to estimate the response (“model response uncertainty”). This component of uncertainty has the potential to be narrowed as scientific understanding and its representation in climate models improves (Hawkins and Sutton, 2009; Hawkins and Sutton, 2010). Third, internally generated climate variability (chaos in the climate system; see Kirtman et al., 2013) confounds detection of the anthropogenic climate change signal. Due to internal variability, it is expected that (1) climate models will not reproduce the exact observed state of the planet in any given year, and (2) a set of climate models, or a set of simulations in a single model with different initial conditions will generate diverse responses to identical climate forcing. In other words, another model realization with the exact same forcing but with slightly different initial conditions may generate a different meteorological response. For these reasons, climate modeling approaches use multiple realizations of the climate system generated by multiple climate models. For an individual climate model, a set of simulations with identical climate forcing but slightly different initial conditions is termed an “ensemble”, with each individual simulation referred to as one “ensemble member”. Consideration of this variability is particularly critical for projecting climate for the near term when the (forced) climate change signal is small relative to (unforced) climate variability (Kirtman et al., 2013). The required number of ensemble members, or simulation years, needed to detect the forced signal depends on the size of the forced signal, the quantity (*i.e.,* climate variable such as temperature or precipitation), region, and time frame of interest, with more realizations needed for quantities with large intrinsic variability, smaller regions, smaller forced signals and closer time periods (Deser et al., 2012; Hawkins and Sutton, 2010; Kirtman et al., 2013). Natural variability in temperature is generally smaller than in precipitation, and the near-term signal of anthropogenic influence on temperature is clearest in the tropics where natural variability is low (Flato et al., 2013).

IPCC (2013a) assesses GCMs and CCMs forced with RCP scenarios to project near-term (1986-2005 to 2016-2035) global mean surface temperature changes of +0.3 to +0.7 °C, and assigns over a 50% probability of reaching 1°C above 1850-1900, and ≤10% probability of crossing 1.5 °C (Table S2), with land warming more rapidly than oceans (Kirtman et al., 2013). Over the U.S.A., temperature is projected to increase during the 21st century, though the rate of warming may vary by region (Melillo et al., 2014; Kirtman et al., 2013; M. Collins et al., 2013) and will fluctuate with natural variability (Deser et al., 2012). Models project drying over Texas and much of the Southwest (Melillo et al., 2014; Seager et al. 2007). Precipitation is projected to increase in much of the northern U.S., particularly in winter and spring in response to rising GHGs (Melillo et al., 2014).

**S3. Evaluation of ACCMIP and AeroCom models used to estimate RF**

*O3*

Uncertainties in the O3 RF estimates associated with the particular RTM scheme used are within ±10% (Stevenson et al., 2013). Some evidence indicates that CCMs and CTMs, while qualitatively reproducing observed features, underestimate century-scale lower tropospheric baseline O3 trends at northern mid-latitudes (Parrish et al., 2014; Cooper et al. 2014). While this finding might imply an underestimate of the RF from tropospheric O3 estimated with these models (Mickley et al., 2001), it is unclear whether trends in these lower tropospheric observations are relevant for O3 RF, which is strongest in the upper troposphere. Furthermore, the observations are sparse and the trends they record may reflect regional climate variability rather than emission trends.

Parameterizations based on sensitivities of surface O3 to precursor emissions diagnosed in a coordinated set of simulations with 14 CTMs indicate that models capture some of the observed global increases in surface O3 over the past three to four decades, though they generally underestimate the increase at continental sites (Wild et al., 2012). While these discrepancies could indicate problems with the emission inventories, they could alternatively suggest a role for changing natural sources or climate (Wild et al., 2012). Indeed, Lin et al. (2014) emphasize the importance of considering climate variability when interpreting observed long-term trends at the Mauna Loa site, one of the sites used for model evaluation by Wild et al. (2012). Young et al. (2013) evaluated ACCMIP models with tropospheric ozonesonde measurements and satellite data; the model ensemble mean captures these observations, including the seasonal cycle, with some exceptions in the tropical upper troposphere. Comparison with satellite-based (OMI/MLS) tropospheric O3 columns suggests that the global mean is also well represented by the ACCMIP models, but tends to be biased high in the northern hemisphere and low in the southern hemisphere (Young et al., 2013).

*PM.*

Satellite products of aerosol optical depth (AOD), the vertically integrated extinction of solar radiation by scattering and absorption, are currently one of the best benchmarks for modeled aerosol distributions and optical properties. Shindell et al. (2013) report that the spatial patterns in total AOD are broadly captured by the ACCMIP models, but generally biased low over Asia. The multi-model mean global average AOD (0.14) is slightly lower than retrievals from the MODIS (0.16) and MISR (0.17) satellite instruments (Shindell et al., 2013), though it is important to note the many uncertainties involved in the retrievals. The ACCMIP models capture AOD over large continental-sized regions as measured by surface AeroNet sun photometers, but the ability of individual models to simulate smaller regional features varies, and they differ in their estimates for the relative importance of specific PM components (Shindell et al., 2013). The ACCMIP models generally overestimate AOD at AeroNet sites between 40-60°N, implying an overestimate of anthropogenic sulfate (Shindell et al., 2013). In contrast, absorbing AOD (AAOD), a measure of the amount of absorption by dust, BC, and BrC, is strongly underestimated in many regions compared to those derived from the OMI satellite instrument, with a factor of 2 low bias globally and poor spatial pattern correlations (Bond et al. 2013; Shindell et al., 2013). While BC atmospheric burdens in the ACCMIP models varied by a factor of 3, their projected increases in pre-industrial to present-day atmospheric burdens by factors of 2.5-3 are consistent with the ACCMIP emission inventory increase of a factor of 2.5.

Regional forcing estimates for BC deposition on bright surfaces (e.g., snow and ice) can exceed 10 W m-2 (Zhao et al., 2014). The common modeling practice of prescribing deposition rates and decoupling those rates from snowfall, however, biases surface albedo RF estimates high by a factor of 1.5-2.5 (Doherty et al., 2014). Model estimates of the BC surface albedo effect are also sensitive to simulated BC spatial distributions, snow albedo parameterizations, and modeled snow cover (Lee et al., 2013). BC deposition to snow and ice surfaces is a particularly effective agent of climate change with ties to Arctic (Hansen and Nazarenko, 2004; Koch et al., 2009; Flanner et al., 2009), North American (Qian et al., 2009), and Himalayan snow and ice melt (Ramanathan and Carmichael, 2008; Menon et al., 2010), which amplify regional temperature increases, some of which may occur in response to BC RF.

Four ACCMIP models included SOA (though generally not its aqueous formation pathways or BrC), estimating 1850 to 2000 RFs of -0.07, -0.03, -0.01, and +0.32 W m-2. The simulated large positive RF from SOA occurs due to imposed land-use changes, which lowers SOA abundance compared to the pre-industrial era (Shindell et al., 2013). The AeroCom models report a range of organic aerosol burdens (biogenic and anthropogenic) ranging from 0.6 to 3.8 Tg, but tend to underestimate observations in urban (62%), remote (15%), and marine (30%) environments (Tsigaridis et al., 2014).

**S4. Sensitivity of Atmospheric Chemistry, Sources, and Sinks to Regional Climate Change and Variability**

We outline here the current understanding for several climate-sensitive atmospheric reactions, sources and sinks. These processes serve as feedbacks on the climate system when they alter emissions of NTCFs or CO2, and influence the sensitivity of regional air pollution to climate change and variability. We note where additional research is needed to build confidence in projections of air pollutant responses to future climate change.

*Atmospheric chemistry*. Three thermally sensitive factors (isoprene emissions, peroxyacetyl nitrate, and water vapor) have long been recognized as strong controls on the response of O3 to meteorology, including on daily time scales (e.g., Trainer et al., 1987; Chameides et al., 1988; Sillman and Samson, 1995; Johnson et al., 1999; Jacob et al., 1995; Steiner et al., 2006). Under warmer conditions, less NOx is locked up in PAN, and instead contributes to local O3 formation (e.g, Sillman and Samson, 1995). A warmer atmosphere holds more water vapor, leading to increased chemical O3 destruction in the remote (low-NOx) atmosphere reducing background O3 levels (IPCC, 2013a). In high-NOx polluted regions where O3 production is limited by the supply of HOx radicals, higher water vapor can increase O3 (e.g., Jacobson, 2008), though the importance of this factor varies locally, with regional-scale O3-water vapor relationships more likely to reflect synoptically-driven correlations (Jacob and Winner, 2009; Camalier et al., 2007). Isoprene emissions increase with temperature and react rapidly to produce O3 (see below). Doherty et al. (2013) demonstrate that the sum of increases in these three individual factors: PAN decomposition, water vapor, and isoprene emissions, captures roughly half (r2 = 0.52) of the spatial variance in the annual mean surface O3 response to climate change in a CCM.

Understanding of isoprene oxidation chemistry remains incomplete (Paulot et al., 2009). Alternative treatments of isoprene-NOx interactions affect the global O3 burden (Ito et al., 2009) and the sign of the local surface O3 response to an increase in isoprene emissions (e.g., Fiore et al., 2005; Weaver et al., 2009), with a strong sensitivity to assumptions regarding organic nitrate chemistry (Browne and Cohen, 2012). Based on laboratory and field measurements Mao et al. (2013a) conclude that the local surface O3 response to isoprene emissions is positive, as isoprene does not titrate OH (necessary for the isoprene-O3 reaction to serve as a sink for O3).

The individual PM components respond differently to meteorological changes. Higher temperatures and humidity can enhance gas-phase sulfate formation, though aqueous phase sulfate production decreases under cloud-free conditions, and there is less partitioning of nitrate and volatile organics into the aerosol phase (e.g., Jiang et al., 2010; Liao et al., 2006; Aw and Kleeman, 2003; Pye et al., 2009; Racherla and Adams, 2006; Kleeman, 2008). For example, measurements at two sites during wintertime PM episodes in the Midwest indicate over a factor of two increase in nitrate associated with air stagnation, attributed at least partially to stronger partitioning towards the aerosol phase (Stanier et al., 2012). PM also varies with oxidant levels, including those induced by remote emissions (Unger et al., 2006; Leibensperger et al., 2011). Major uncertainties surround model chemical mechanisms for oxidation of isoprene and other biogenic volatile organic compounds (BVOC), and the ensuing impacts on O3 and SOA (e.g., Lelieveld et al., 2008; Carlton et al., 2009; Paulot et al., 2009; Hoyle et al., 2011; Murphy et al., 2012; Mao et al., 2013a). New mechanistic understanding is emerging on aqueous phase SOA production (e.g., Fu et al., 2009; Carlton et al., 2010; Ervens et al., 2011; McNeill et al., 2012, 2015; He C. et al., 2013). Finally, uncertainties persist regarding heterogeneous chemistry of aerosols and their influence on tropospheric composition and regional air quality (e.g., Jacob, 2000; MacIntyre and Evans, 2010; Mao et al., 2013b).

*Anthropogenic Emissions*. Higher anthropogenic NOx emissions associated with meeting electricity demands to power air conditioning on the hottest days have been estimated to produce at least one-third of the observed O3 dependence on temperature, with an estimated ~2.5-4% °C-1 increase in power plant NOx emissions for 1997-2011 (He et al., 2013). Bloomer et al. (2009) and others have documented the decreasing response of O3 to temperature as NOx emission controls have been implemented since the late 1990s. Loughner et al. (2014) report that in 1980, there were twice as many days with O3 above the current 75 ppb NAAQS level as the number of hot days (defined as maximum temperature **≥** 90 °F) whereas from 2009-2013 there were fewer high-O3 days (above 75 ppb) than hot days. With model simulations, Loughner et al. (2014) further demonstrate that emission controls phased in since 2002 prevented 3 to 11 high-O3 days during the hot July of 2011 throughout Maryland. Figure 6a shows this improvement in air quality: despite similar temperature excursions throughout the record, July mean maximum daily average 8-hour (MDA8) O3 levels did not exceed 65 ppb from 2003-2014.

*Natural Sources.* We discuss here natural sources that are climate-sensitive and expected to influence O3 or PM2.5 in some U.S. regions. These changes in natural sources can also alter global O3 and PM distributions, serving as a feedback to climate change. Some models project O3 enhancements from higher natural sources such as stratosphere-to-troposphere and lightning NOx in a warmer climate (e.g., Stevenson et al., 2013; Young et al., 2013). While these climate-change induced increases in natural sources are uncertain, they influence O3 in the mid-to-upper troposphere where the O3 lifetime is longer and it is more effective as a GHG. The accompanying changes in RF may not be evident from consideration of global mean changes in O3 burdens as one of the most robust impacts of climate warming is to decrease lower tropospheric O3, which could offset larger upper tropospheric natural sources in terms of the global mean burden.

*Biogenic NMVOC emissions:* Biogenic NMVOC includes isoprene and terpenes, which are strongly temperature dependent, and some also respond strongly to variations in sunlight and soil moisture (e.g., Guenther et al., 2006). Emissions in CCMs and CTMs are typically parameterized using spatial maps of base emissions that are adjusted according to the local temperature and sunlight, with some approaches incorporating additional factors such as soil moisture or leaf age (Guenther et al., 2006; 2012) or tying emissions directly to photosynthesis in a dynamic vegetation land model (Unger et al., 2013). Rising CO2 may suppress isoprene emission (Rosenstiel et al., 2003), opposing the influence of rising temperatures (Pacifico et al., 2012). The species-specificity of isoprene emissions (Guenther et al., 1995) further implies that any changes in the distribution of particular plant species, whether induced by a changing climate (e.g., Sanderson et al., 2003) or land-use (e.g., Lathiere et al., 2010; Purves et al., 2004; Wiedinmyer et al., 2006; Heald et al., 2008, Wu et al., 2012), will alter emissions and the local air quality. Huang et al. (2015) estimate that monthly isoprene emissions vary by over 30% from year to year, and that Texas droughts in 2006 and 2011 reduced isoprene and monoterpene emissions by as much as 24%. Formaldehyde, a short-lived intermediate of isoprene oxidation, has been retrieved from satellite instruments since the mid-1990s and offers some bounds on monthly and inter-annual variability in isoprene emissions over the United States (Abbot et al., 2003; Palmer et al., 2006; Millet et al., 2008). Where isoprene is abundant, O3 is strongly NOx-limited (e.g., Jacob et al., 1995), such that rising isoprene emissions are unlikely to offset the O3 decreases resulting from NOx reductions projected under the RCPs (Figure S2). SOA production from biogenic emissions is expected to vary with climate (Heald et al., 2008; Jiang et al., 2010) but yields remain uncertain and likely vary with local factors including organic aerosol loading, NOx, the degree of oxidation including through aqueous pathways (e.g., Carlton et al., 2009; McNeill, 2015). We emphasize the important distinction between biogenic and natural, since over half of SOA derived from biogenic VOC emissions may be controllable via anthropogenic emissions (e.g., Carlton and Turpin, 2013; Xu et al., 2015).

*Wildfires:* Wildfires contribute to high-PM2.5 events, particularly in the western United States (e.g., Jaffe et al., 2008a) where they are the major driver of inter-annual variability in OC (Spracklen et al., 2007). From a policy perspective, these events can be screened from counting towards attainment determinations as exceptional events (e.g., Taubman et al. 2004; see also Fiore et al., 2014a). Nevertheless, they do pose a direct threat to public health, with fire conditions linked with a 50% higher probability of people seeking emergency care as compared to non-fire conditions (Thelen et al., 2013). The contribution of wildfires to O3, including during individual events, is under debate (e.g., Jaffe and Wigder, 2012; see Fiore et al., 2014b and references in their Table 1).

Wildfires have been increasing in the Western U.S. with spring and summer temperatures (Westerling et al., 2006). While global fire emission inventories exist based upon space-based products of area burned (e.g., Duncan et al., 2003; van der Werf et al., 2006; 2010; Kaiser et al., 2012; Wiedinmyer et al., 2011), computationally efficient predictive capability such as needed to project air pollutants with CCMs is only beginning to be developed (e.g., Pechony and Shindell, 2009), as are mechanistic representations of fires in vegetation models (e.g., Flannigan et al., 2009). Regression models that account for current and antecedent meteorology and a parameterization that considers temperature, precipitation, relative humidity, Santa Ana winds and geographical dependence of wildfires all project increases (ranging from 10% to doubling depending on the region and approach) in area burned by mid-21st century, as well as the possibility of a longer fire season in southwestern California due to warmer and drier conditions on Santa Ana days in November (Yue et al., 2014). Related approaches have been used to project area burned, combined with wildfire emission factors, in CTMs that project increases in western U.S. OC and BC levels in surface air of 46-70% and 20-27%, respectively by mid-century (Yue et al., 2013; Spracklen et al., 2009). Hurteau et al. (2014) considered additional factors including changing population distributions and development densities, and hydrologic variables obtained by downscaling temperature and precipitation for two scenarios from three GCMs for each 1/8° grid cell over California. With this approach, they find wildfire emission increases of 19-101% (median of 56%) over the 21st century, with the largest increases in northern California (Hurteau et al., 2014).

Jaffe et al. (2008b, 2011) point out that summertime mean O3 is enhanced in the Western U.S.A. in years with high wildfires. This relationship may reflect a common underlying relationship with temperature rather than a direct impact of wildfires on O3. Zhang et al. (2014) find little response of Western U.S. O3 in their model to year-to-year changes in wildfire emissions, but instead find that warmer years are associated with deeper mixing depths, enabling higher O3 levels from the free troposphere to reach the surface in warmer years. Uncertainty remains, however, in the possible role of PAN transport and subsequent O3 production from fire plumes in this region (Zhang et al., 2014).

*Wetland and permafrost CH4*: Globally, emissions from wetlands drive much of the observed inter-annual variability in atmospheric CH4 (Kirschke et al., 2013), and are expected to increase in a warmer climate, though the magnitude and regional distribution are uncertain (Ciais et al., 2013). O’Connor et al. (2010) review parameterizations for climate-sensitive wetland emissions that have been incorporated into GCMs and CCMs, some of which consider changing areal extents for wetlands. Only two of the ACCMIP models (Figure 10) included changes in CH4 emissions from wetlands in their projections since the majority fixed CH4 abundances either directly or through a lower boundary condition (Lamarque et al., 2013b). None of the ACCMIP models included a possible, but highly uncertain, CH4 source from thawing permafrost, which may increase as the Arctic climate continues to warm (e.g., Schuur et al., 2015). Increases in atmospheric CH4 will raise baseline tropospheric O3, thus serving as a positive feedback on climate, and raising ground-level O3 globally.

*Soil NOx:* Soil NOx emissions vary with temperature and precipitation, with pulsing induced by precipitation following dry spells (Yienger and Levy, 1995). Some CTMs and CCMs include a climate-dependent parameterization based on Yienger and Levy (1995). Estimates with updates to that parameterization, including new fertilizer application data, indicate that soil NOx enhances MDA8 O3 in surface air over the U.S. Great Plains by 3 ppb on average in June, with weather-dependent inter-annual variability increasing this value to 5 ppb in June of 2006 (Hudman et al., 2010).

*Lightning NOx:* Lightning activity is expected to change in a warming climate (Price, 2013; Williams, 2005). The tropospheric O3 and CH4 burdens and resulting climate impacts (note that NOx increases O3 but decreases CH4) are more sensitive to changes in lightning NOx than anthropogenic NOx (e.g., Wild et al., 2001; Labrador et al., 2005; Murray et al., 2013), though surface O3 is typically much more sensitive to regional anthropogenic NOx emissions. Some studies, however, find lightning NOx influences of up to 10-14 ppb in U.S. surface air (Kaynak et al., 2008; Zhang et al., 2014). Present-day simulations of the lightning NOx source can be constrained with flash rates observed by satellite instruments (Murray et al., 2012) or by assimilating multiple species retrieved from satellite (Miyazaki et al., 2014). Model parameterizations based on convective cloud top heights (Price and Rind, 1992), convective mass fluxes or convective precipitation (Allen et al., 2002) generally project increased lightning NOx in warmer climates (e.g., John et al., 2012; Lei et al., 2012; Young et al., 2013, Murray et al., 2014), one model with a microphysically-based representation of lightning NOx projects decreases due to declining numbers of ice crystals (Jacobson and Streets, 2009).

*Dust****:*** Dust emissions, both anthropogenic and natural, depend strongly on climate, particularly on changes in the hydrological cycle as reviewed by Ginoux et al. (2012). Models (CTMs and CCMs) often include climate-dependent dust emissions (e.g., Ginoux et al., 2001; Lamarque et al., 2012). Studies conflict, however, in projections of the dust response to climate change (Mahowald and Luo, 2003; Tegen et al., 2004; Myhre et al., 2013).

*Downward Transport from the Stratosphere*: The western U.S. is downwind from an active region for stratosphere-to-troposphere O3 transport in spring (e.g., Skerlak et al. 2014), which can contribute to O3 events above the NAAQS level (Langford et al., 2009; Lin et al., 2012a). Several modeling studies suggest increased stratosphere-to-troposphere flux under climate change associated with the combination of stratospheric O3 recovery and a change in stratospheric circulation, which may increase the downward flux of O3 at northern mid-latitudes (e.g., Hegglin and Shepherd, 2009; Kawase et al., 2011). New evidence from satellite data exploits natural climate variability to support the model-based increases in stratosphere-to-troposphere O3 exchange with climate warming (Neu et al., 2014). Observations during a field campaign over the central United States in summer 2012 also suggest that thunderstorms may enable fine-scale transport of stratospheric O3 into the troposphere that is not resolved in current CCMs (Pan et al., 2014).

*Dry and Wet Deposition.* Both wet and dry deposition remove PM and O3 precursors. The suppression of O3 dry deposition by vegetation under drought conditions has been implicated in contributing to extreme pollution events (Solberg et al., 2008) and has been projected to change with regional climate (Andersson and Engardt, 2010). The representation of dry deposition varies widely in current global models (Hardacre et al., 2014), reflecting a lack of clear process-based understanding from observations (e.g., Fowler et al., 2009; val Martin et al., 2014). Changes in wet deposition are tied to precipitation changes, discussed in the main text.

**S5. Evaluating Models Used To Study U.S. Air Quality Responses To Emissions Or Meteorology**

Computational advances now enable global CCMs and CTMs to perform decadal and centennial simulations at 1°x1° or 2°x2° horizontal resolution (e.g., Lamarque et al., 2013b and references therein), with resolutions comparable to those of RCMs and RCTMs possible for shorter periods (e.g., Lin et al., 2012ab; Zhang et al., 2014). Pfister et al. (2014) demonstrated that high-resolution models may simulate different mean states as they spatially refine simulations relative to a coarse resolution configuration, but that the coarse- and high-resolution versions both project similar changes about their respective mean states. Similar findings have been noted for present-day applications of coarse- versus high-resolution models, as well as the decoupling of model capability to represent temporal (e.g., day-to-day) variations versus mean O3 levels at individual monitoring sites (e.g., Fiore et al., 2003; 2014a). These findings imply that bias-correction or statistical downscaling methods to spatially refine projections from global models may provide useful information at the local scale (see also Hall, 2014), though urban-rural differences not represented at the coarse scale should be considered.

*Surface O3.* A systematic positive bias in summertime eastern U.S. surface O3 plagues many regional and global models (e.g., Murazaki and Hess, 2006; Nolte et al., 2008; Fiore et al., 2009; Reidmiller et al. 2009; Naik et al., 2013a; Brown-Steiner et al., 2015). val Martin et al. (2014) attribute a portion of this bias in some models to an erroneous implementation of dry deposition. Despite mean state biases, these models generally capture the salient features of O3 pollution episodes, including their areal extent and duration (Fiore et al., 2003), as well as year-to-year variability (Schnell et al., 2014), indicating that they represent the underlying processes controlling the build-up of air pollution events, and are thus suitable tools for studying how air pollution events will change as climate and emissions evolve.

Observed U.S. air quality responses to emission controls implemented over recent decades provide key tests for the CCMs and CTMs used to project future air quality in response to proposed emission control programs (e.g., Table S1). NO2 columns retrieved from satellite instruments and ground-level NO2 measurements from the U.S. Air Quality System both indicate an average decrease of 38% in U.S. tropospheric NO2 columns from 2005 to 2013, along with a changing amplitude of the NO2 seasonal cycle in response to declining NOx emissions (Lamsal et al., 2015). “Dynamic evaluation” of emission-response relationships tests model skill at simulating the observed differences due to meteorology and emission shifts from one year to another (e.g., Gilliland et al., 2008; Nolte et al., 2008). Prior studies have attributed eastern U.S. decreases in various O3 metrics over recent decades to NOx emission controls (Frost et al., 2006; Gégo et al., 2007; Bloomer et al., 2009; 2010; Kang et al., 2013; Napelenok et al., 2011; Zhou et al., 2013; Figures 6, 10 and 11). The highest observed surface O3 levels decrease most (Cooper et al., 2012; Rieder et al., 2013), broadening the seasonal cycle to a spring-summer maximum in polluted regions where summertime peaks were typically observed during the 1990s (Clifton et al., 2014; Cooper et al., 2014). The overall O3 distribution is thus more narrow, particularly as the lowest concentrations are increasing in many U.S. regions (e.g., Cooper et al., 2012; 2014; Simon et al., 2014). CTMs and CCMs generally represent the observed summertime decreases, wintertime increases, and larger declines on the highest (and warmest) days in response to NOx emission reductions (e.g., Clifton et al., 2014; Rieder et al., 2015; Brown-Steiner et al., 2015).

Historically observed relationships between relevant meteorological variables and air quality (Lin et al., 2001; Bloomer et al., 2009; Tai et al., 2010) provide tests for model responses to changing meteorology. An evaluation of the O3-temperature relationship reveals more success in capturing observed relationships over the Northeast and Midwest than over the mid-Atlantic (Rasmussen et al., 2012). Tawfik and Steiner (2013) find that O3 in the Southeast correlates strongly with surface drying (evaporative fraction) suggesting that regional O3-temperature relationships respond to differences in the soil moisture-atmosphere coupling regime. The higher model skill in the Northeast thus likely reflects the more accurate simulation of large-scale synoptic conditions, which shape the O3-temperature relationship in the Northeast, relative to the land-atmosphere couplings responsible for surface drying in the Southeast. One study demonstrates a dependence of simulated U.S. O3-temperature relationships and extreme O3 on the number of vertical levels in the CCM, cloud cover, photolysis, isoprene emissions, and the model meteorology (Brown-Steiner et al., 2015).

*Surface PM*. Ambient concentrations and deposition of PM2.5 components have been observed for decades (Lehmann et al., 2007). Models (e.g., Pozzoli et al., 2011; Leibensperger et al., 2012b) and observations (e.g., Sickles and Shadwick, 2015) attribute the observed eastern U.S. decline in sulfate concentrations to SO2 emission controls. Leibensperger et al. (2012b) also showed that a CTM reproduces the lack of a trend in ammonium wet deposition but indicates little trend in nitrate deposition despite decreasing observations, suggesting poor model representation of emission trends, and possibly the sulfate-nitrate-ammonium system. Decadal and longer records from satellite offer new opportunities to evaluate air quality trends globally (Martin, 2008; Streets et al., 2013). Boys et al. (2014) infer a decrease from 1999 to 2012 in eastern U.S. PM2.5 of -0.37±0.13 μg m-3 yr-1 from satellite data as compared to -0.38±0.06 μg m-3 yr-1 from ground-based sites, attributed to decreasing sulfate-nitrate-ammonium aerosol.

Models generally capture surface distributions of BC but show large discrepancies with remote observations (Q. Wang et al., 2014; X. Wang et al., 2014), reflecting uncertainties in emissions, aging mechanisms, optical properties (when assessed by AAOD), and wet scavenging (Bond et al., 2013). Over the U.S.A., BC decreases of 1-5% yr-1 are estimated for 1990 to 2004 (Murphy et al., 2011), but models generally fail to capture these trends (Koch et al., 2011; Leibensperger et al., 2012b). Simulating OC remains problematic (Kanakidou et al., 2005; Tsigaridis et al., 2014), especially in the southeastern U.S.A. (Ford and Heald, 2013), though improvement occurs with updated SOA mechanisms (Carlton et al., 2010). Uncertainties in SOA stem from the anthropogenic and biogenic precursor NMVOC emissions and the subsequent atmospheric chemistry (Hallquist et al., 2009).

**S6. Observed Relationships Between Air Pollutants and Meteorology and Statistical Downscaling Approaches**

Numerous statistical methods exist to remove the influence of meteorology on observed air pollutant trends in order to discern the efficacy of pollution control programs (e.g., Porter et al., 2001). The U.S. EPA has begun to provide both the raw observed and weather-adjusted trends in summer mean O3 and annual PM2.5 (<http://www.epa.gov/airtrends/reports.html>; <http://www.epa.gov/airtrends/weather.html>). Long-term observations thus contain information regarding the response of air pollution to variability in meteorology, which may offer insights to the response to climate change.

One method to estimate future changes in air quality combines GCM or RCM projections of regional climate change with observed relationships between air pollution and meteorology (Statistical Downscaling in Table S1). Many of the observed relationships between a single meteorological variable and an air pollutant, however, reflect the net response to air pollution meteorology, atmospheric chemistry and sources and sinks. For example, the strong observed correlation between O3 and temperature in many polluted U.S. regions (Figure 6) reflects several processes (Figure 2;see also Weaver et al., 2009; Rasmussen et al. 2012). These include: (1) the impact of temperature on reaction rates, particularly on the thermal suppression of peroxyacetyl nitrate (PAN) formation which leads to additional NOx available to produce O3 locally (e.g., Sillman and Samson 1995; Steiner et al., 2010); (2) the impact of temperature on precursor availability, including from anthropogenic NOx (higher electricity demand; He H et al., 2013), and biogenic NMVOC (Guenther et al., 1995; Steiner et al., 2006; Andersson and Endgart, 2010); see also Supplemental Text 4, and (3) the underlying dependence of extreme temperature and pollution on air pollution meteorology, including cloud-free conditions with abundant radiation needed for photochemistry (Logan, 1989; NRC, 1991).

Over the Southeastern U.S.A., the O3-temperature correlation is weaker than in the Northeast (e.g., Camalier et al., 2007). In this region, surface drying, expressed as evaporative fraction, has been shown to correlate better with O3 than temperature, specific humidity, or radiation, which may reflect a fundamental shift in the soil moisture-atmosphere coupling regime between the Northeast and Southeast U.S.A. (Tawfik and Steiner, 2013). The present-day relationship between O3 and temperature has been quantified (Bloomer et al., 2009; Rasmussen et al., 2013), but the processes controlling this relationship are unlikely to scale simply with temperature (Steiner et al., 2010; Kirtman et al., 2013; Tawfik and Steiner, 2013).

Statistical downscaling approaches that identify the underlying drivers of observed relationships, such as stagnation events (Leibensperger et al., 2008; Tai et al., 2012ab; Thishan Dharshana et al., 2012), proximity to the summertime mid-latitude jet (Barnes and Fiore, 2013), or surface drying (Tawfik and Steiner, 2013) may be more reliable. At present, however, this approach is limited by GCM (or CCM) skill of projecting changes in the frequency, duration, and intensity of regional air stagnation (particularly those events associated with atmospheric blocking (e.g., Christensen et al., 2013), the jet latitude (e.g., Barnes and Polvani, 2013), and land-atmosphere couplings (Dirmeyer et al., 2013). Statistical downscaling approaches based solely on air pollution meteorology can be confounded by the dependence of present-day relationships on the chemical regime (e.g., availability of NOx and VOC) (Bloomer et al. 2009; Rasmussen et al., 2012, 2013; Steiner et al. 2006, 2010; Figure 6). For example, Rasmussen et al. (2013) illustrate the strong dependence of the O3-temperature relationship on precursor emissions for two urban airsheds in California, adding this information to O3 isopleth plots, which indicate the efficacy of possible O3 precursor control strategies. Future changes in the balance between local-to-regional and background pollutant levels such as due to changing water vapor, global CH4 or changes in stratospheric O3 influx (Kirtman et al., 2013; Lamarque et al., 2011; Kawase et al., 2011; Clifton et al., 2014) may also complicate projections based on present-day relationships between air pollutants and meteorological conditions. The response of PM2.5 will vary by region and in time with the major components of PM2.5. For example, formation of the nitrate component of PM2.5 is inhibited at warmer temperatures, and the regional PM2.5 response may be dominated by climate-sensitive sources such as wildfires, dust, and biogenic precursors of organic carbon (Jacob and Winner, 2009; Dawson et al. 2014; Supplemental Text S4).

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**Table S1.** Summary of Methods used to study air quality-climate connections

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Approach** | **Example questions** | **Advantages** | **Limitations** | **Example References** |
| *Methods applied to study air pollutant impacts on the climate system* | | | | |
| Radiative Transfer Model (RTM) forced with distributions of atmospheric constituents from a Chemistry-Climate Model (CCM), or a Global Chemistry-Transport Model (GCTM) | What is the radiative forcing (RF) from changes in one or more atmospheric constituents (or to a change in emissions)? | Enables attribution of RF to changes in individual atmospheric constituents (or emission perturbations) | Inputs such as cloud distributions in the RTM are not necessarily consistent with the simulation of the atmospheric constituents. The simulations used to estimate changes in atmospheric distributions of constituent(s) are often limited to a few years of meteorology. | Feng et al., 2013;  Fry et al., 2012; 2013; 2014; Fuglestvedt et al., 1999; Naik et al., 2005;  Stevenson et al., 2013; Wild et al., 2001. |
| Global CCM driven by fixed (monthly-varying) sea surface temperatures, (also called “Radiative Flux Perturbations”) | What is the effective radiative forcing (ERF) from a particular atmospheric species or emitted compound? | As above but includes rapid adjustmentsa such as aerosol-cloud interactions | Land warming complicates the calculation. | Boucher et al., 2013;  Hansen et al., 2005; Lohmann et al., 2010; Shindell et al., 2013;  Unger et al., 2010. |
| Regression of net energy imbalance onto the change in GMSTb in a transient climate simulation | As above | As above | Estimate can be confounded by natural variability or time-varying feedbacks. | Boucher et al., 2013; Gregory et al., 2004. |
| General Circulation Models (GCMs) | How will climate evolve? What is the climate response to a given forcing agent? | Long, multi-ensemble simulations allow separation of climate change signal from variability | Prescribed ozone and aerosol fields drive evolution of climate system, introducing inconsistencies in the distributions of chemical and meteorological variables; calculating RF from a single constituent requires double calls to the radiation code. | Typical IPCC-class models used to project changes in climate variables, e.g. Collins, M. et al., 2013; Flato et al., 2013; Leibensperger et al., 2012b. |
| Regional Climate Models (RCMs) | As above but for a targeted region | Finer resolution; more complex representation of processes | Limited by availability of boundary conditions from the parent GCM and the GCM skill at representing large-scale circulation. | Gustafson and Leung, 2007; Trail et al., 2013. |
| Global CCMs, either coupled to a full ocean model, or driven by sea surface temperatures and sea ice archived from a GCM | How does the climate system respond to changes in air pollutants? | Consistent simulation of meteorology and air pollutants; represents at least some two-way interactions between air pollutants and climate | Computational expense from chemistry implies coarser resolution than possible with GCMs; simplistic or absent representations of some processes; multiple sensitivity simulations required for source attribution; calculating RF from a single constituent requires double calls to radiation code. | Bellouin et al., 2011; Collins et al., 2011; Jones et al. 2011; Lamarque et al., 2012; Levy et al., 2013; Naik et al., 2013a; Pawson et al., 2008; Pozzoli et al., 2008;  Rotstayn et al., 2013; Shindell et al., 2006; 2012; 2013; Szopa et al. 2012; Watanabe et al., 2011; Zhang et al., 2012. |
| Regional CCMs | How does regional climate respond to local changes in air pollutants (or to global forcings via changing boundary conditions)? | As above but with higher resolution and/or more complex description of processes such as for resolving clouds | As for RCMs, plus boundary conditions for chemical and meteorological variables are not necessarily consistent. | Kalina et al., 2014; Mashayekhi and Sloan, 2014; Morrison 2012; Thompson and Eidhammer, 2014. |
| Nested grid models (e.g., one-way global through urban scales) | How do air pollutants or specific sectors affect local-to regional climate? | Fully consistent simulation that resolves processes at the relevant scale in the region of interest | Limited by ability to resolve relevant processes, or if two-way couplings between scales are important. | Jacobson et al., 2007; Jacobson, 2008. |
| Reduced-complexity climate (and carbon cycle or earth system) models | How does GMST (or another climate response) evolve under a wide range of scenarios? | Emulates GCMs for rapid calculation of GMST or other climate response to multiple emission scenarios | Simple representation of climate system may not properly account for couplings in the system such as chemical feedbacks. | Meinshausen et al., 2011; Rogelj et al., 2014; Shoemaker et al., 2013; Shoemaker and Schrag, 2013; Smith and Mizrahi, 2013; Unger et al., 2010. |
| Analytical formulae | What is the RF from changes in a well-mixed GHG? | Simple relationship between equilibrium RF to a change in abundance (or emissions) | As above. | Ramaswamy et al., 2001. |
| Simple box models | What is the GMST response to different RF scenarios? | Can incorporate multiple time scales to estimate transient and steady-state GMST changes for a variety of scenarios | As above; neglects possible dependence of GMST response to the spatial distribution of RF and thus is unlikely to approximate well the response to short-lived air pollutants. | Boucher and Reddy, 2008;  Held et al., 2010; Pierrehumbert, 2014; Shindell et al., 2012. |
| Adjoint methods in a GCTM | Which emissions (species, region, sector) contribute to RF by a particular atmospheric constituent (regional, global)? | Receptor-oriented framework allows attribution to multiple factors in a single simulation; complementary to forward modeling source attributions | Assumes linear system; by design targets a single quantity (‘receptor’). | Henze et al. 2012. |
| *Methods applied to study climate impacts on air pollution* | | | | |
| Observed relationships between chemical and meteorological variables | How does air pollution respond to changing weather? | Observational constraints on models | Does not directly provide information on how climate will change, and may not correctly identify the underlying drivers of the correlation. | Bloomer et al., 2009; Dawson et al. 2014; Jacob and Winner, 2009; Rasmussen et al., 2012; Tai et al., 2010, 2012a. |
| Statistical downscaling using observed relationships (above) and climate change projections | How will air pollution respond to changes in regional or global climate? | Can be applied to a large suite of GCM physical climate change simulations (or their emulators) | Assumes stationarity in the air pollutant response to the projected meteorological variable. | Holloway et al., 2008; Tai et al., 2012b. |
| CTMs driven by perturbed meteorological variables | How does air pollution respond to changes in a meteorological driver? | Develops process-level understanding of air pollutant response to changes in a single factor; helps diagnose responses in more complex models | Changes in air pollutants do not feed back on the climate system. | Section 4 of Jacob and Winner, 2009; Rasmussen et al., 2013; Steiner et al., 2006. |
| Global CTM driven by meteorology from a GCM (or observed for present-day source attributions) | How do changes in climate alter air quality? | Sensitivity simulations separate role of changes due to meteorology versus emissions or chemistry. | As above, plus often limited to a few years of meteorology from the parent GCM, complicating attribution to climate change. | Jacob and Winner, 2009; West et al., 2013; Wu et al. 2008ab; Weaver et al., 2009. |
| Regional CTM driven by meteorology from a GCM or RCM | As above, but with a region-specific focus. | As above, may also consider region-specific feedbacks or processes | As above but with additional dependence on chemical and meteorological boundary conditions which may not be consistent. | Gao et al., 2013; He et al., 2014; Jacob and Winner, 2009; Kelly et al., 2012; ; Penrod et al., 2014;Trail et al., 2014; Weaver et al., 2009 |
| Global CCMs | How do changes in climate alter air quality? | As above but with a consistent simulation of meteorology and pollutants | Coarse resolution; simplistic or absent representations of some processes, particularly interactions with the biosphere. | Clifton et al., 2014; Doherty et al., 2013; Fang et al., 2013; Fiore et al., 2012; Jacobson and Streets, 2009; Lamarque et al., 2011; Rieder et al., 2015; Young et al., 2013. |
| Regional CCMs | As above, but with region-specific focus. | As above but with higher resolution and/or more complex description of processes | Boundary conditions for chemical and meteorological variables not necessarily consistent. | Shalaby et al., 2012. |
| Global-to-regional nested CCM (one-way) | As above. | As above but avoids inconsistent boundary conditions | Limited by ability to resolve relevant processes, or if two-way couplings between scales are important. | Jacobson, 2008 |

a. Rapid adjustments are not dependent on the change in temperature, and thus do not act as a feedback to a change in temperature. They include processes such as cloud changes, including those induced by interactions with aerosols, as well as lapse rate changes, geographic temperature variations, and changes in the biosphere that are not direct responses to temperature; some rapid adjustments occur for many atmospheric constituents, including CO2. See Boucher et al. (2013) for a thorough explanation.

b. Global Mean Surface Temperature

**Table S2:** Global annual mean temperature changes for the 2081–2100 period relative to 1850-1900 as projected with GCMs and CCMs (first four rows), excerpted from M. Collins et al. (2013). Shown are temperature changes for each RCP scenario (mean, ±1 standard deviation and 5 to 95% ranges obtained by multiplying the model ensemble standard deviation by 1.64), assuming that 0.61°C warming has occurred prior to 1986–2005 (third column). The final three columns show the percentage of models projecting 2081–2100 temperatures above levels of 1°C, 2°C, and 4°C for each RCP scenario. The last row shows the global annual mean temperature change for the 2016-2035 relative to 1986-2005 assessed as likely (≥66%) by Kirtman et al. (2013).

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Time period** | **Scenario** | **ΔT (°C)** | **ΔT > +1.0 (°C)** | **ΔT > +2.0 (°C)** | **ΔT > +4.0 (°C)** |
| 2081-2100 | RCP2.6 | 1.6 ± 0.4 (0.9, 2.3) | 94% | 22% | 0% |
|  | RCP4.5 | 2.4 ± 0.5 (1.7, 3.2) | 100% | 79% | 0% |
|  | RCP6.0 | 2.8 ± 0.5 (2.0, 3.7) | 100% | 100% | 0% |
|  | RCP8.5 | 4.3 ± 0.7 (3.2, 5.4) | 100% | 100% | 62% |
| 2016-2035 versus 1986-2005 | All RCPs considered | +0.3 to +0.7 °C | 50% |  |  |

**Table S3.** Estimates of the impact of climate and emission change on O3 and PM2.5 air quality in the U.S.A. as derived from various studies applying modeling approaches described in Table S1. Regions are abbreviated: Northeast (NE), Southeast (SE), Midwest (MW), Great Plains (GP), Northwest (NW), and Southwest (SW) following the regions defined in Melillo et al. (2014), and InterMountain West (IMW) following Clifton et al. (2014). Regional ranges are subject to uncertainties as most numbers were obtained by reading them from maps provided in the papers.

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| --- | --- | --- | --- | --- | --- | --- |
| **Surface O3 change (ppb or as noted)** | **Reference and**  **Modeling Approach** | **Scenario** | **Time horizon** | **Metric Reported** | **Climate change (holding air pollutant emissions constant)** | **Climate + emissions change** |
| Combined ranges from studies cited in Table 2 from Jacob and Winner (2009) and Figure 7 of Weaver et al. (2012)  GCM-CTM; RCM-RCTM | Various (SRES A1B, A1FI, A2, B1) | Various, mainly end of century | JJA (or July) mean MDA8 | NE: up to +8  MW: -2 to +3  Western US (mainly CA and NV): -2 to +3  Gulf Coast (mainly TX/LA): -8 to +4  SE: -6 to +5 | N/A |
| Ranges from studies cited in Figure 3 of Fiore et al. (2012); GCM-CTM; RCM-RCTM | IS92a (2030);  SRES A1B, A2 (2050) | 2030, 2050 | Various | Entire U.S.  -0.4 to + 1.2 (2030),  -6 to +10 (2050) | N/A |
| Avise et al. (2009)  RGCM-RCTM | A2 | 5 separate Julys from 2045-2054 vs. 1990-1999 | Spatially averaged July mean MDA8 | EPA Regionsa:  1-3 combined: +4  4: -5  5: +1  6: -6  7: -1  8: +0  9: +0  10: -1 | EPA Regionsa:  1-3 combined: +12  4: +3  5: +7  6: +3  7: +5  8: +9  9: +12  10: +7 |
| Clifton et al. (2014)  CCM | RCP4.5, RCP8.5 | 2091-2100 vs. 2006-2015 | Monthly, 3-ensemble member meanb | NE (JJA): +1 to +2 (RCP4.5); +3 (RCP8.5)  IMW (Jun-Nov): -4 to -1 (both scenarios); little change rest of year | NE JJA: -15 (RCP4.5); -10 (RCP8.5); DJF: up to +5 (RCP4.5); up to +20 (RCP8.5).  IMW JJA: up to -15 (RCP4.5); little change (RCP8.5); DJF: up to -4 (RCP4.5); up to +14 (RCP8.5). |
| Doherty et al. (2013)  3 CCMs | SRES A2 | 2095 vs. 2000 | Annual mean; spatial range over 3 CCMs | NE: -1 to +6  SE: -2 to +6  MW: -1 to +4  GP: -2 to +4  NW: -4 to 0  SW: -2 to +1 | N/A |
| Gao et al. (2013)  Kim et al. (2015)  GCM/RCM-RCTM | RCP4.5, RCP8.5  (SSTs used to drive GCM are taken from the closest SRES scenario) | 2057-2059 vs. 2001-2004 | Spatial range of seasonal mean MDA8b | N/A | JJA: NE, SE, SW: Mostly < -10; GP, NW: < -10 to 0; MW: mostly < -10 to -6; urban areas change little or increase (RCP4.5)  NW/GP: little change; MW, NE: mostly -6 to -0; SE: -10 to 0; urban areas increase (RCP8.5)  DJF: NE, SE, MW: 0 to > +10; GP, SW: -4 to 0; NW: -2 to +2; urban areas: +5 to > +10 (RCP4.5)  SW: +2 to +6; SE, GP: +2 to +7; NE, MW: +5 to >+10; urban areas >+10s (RCP8.5) |
| Hedegaard et al. (2013)  GCM-RCTM | RCP4.5 | 2090-2099 vs. 1990-1999 | in annual mean | Spatial range over contiguous U.S.: up to +20%, highest in NE | Spatial range over contiguous U.S.: -10% to < -20% |
| Lei et al. (2012)  CCM | B1, A1B, A1FI | 2048-2052 vs. 1998-2002 | Jun-Aug MDA8 mean | N/A | NE: -20 to -5 (B1, A1B), +10 to +20 (A1FI).  SE: -20 to -5 (B1, A1B), +10 to +20 (A1FI); MW: -20 to -5, (B1, A1B), +10 to +20 (A1FI).  GP: -10 to 0 (B1, A1B), +5 to 20 (A1FI).  NW: -10 to +5 (B1, A1B), +10 to +20 (A1FI).  SW: -20 to 0, (B1, A1B), +10 to +20 (A1FI). |
| Kelly et al. (2012)  RCM-RCTM | SRES A2 climate, RCP6.0 emissions | 2041-2050 vs. 1997-2006 | JJA MDA8 | NE: 0 to +6; SE: 0 to +6;  MW: 0 to +7; GP: -1 to +4; NW: -1 to +2;  SW: -1 to >+7; largest increases in urban areas | NE, MW: -25 to -5.  SE: -35 to -5.  GP, NW, SW: mostly -15 to 0; small regions with larger decreases; increases only in Los Angeles (>+15 ppb). |
| Penrod et al. (2014)  RCM-RCTM | SRES A1B | 2026-2030 vs. 2001-2005 | Seasonal mean MDA8b | N/A | NE: DJF: 0 to +4; JJA: < -5 to -2.  SE: DJF: -3 to +3; JJA: <-5 to -1 except coastal FL/LA increases up to +5.  MW: DJF: 0 to +4; JJA: <-5 to +4.  GP: DJF: -3 to +2; JJA: <-5 to +1 except >+5 in Houston. NW: DJF: -2 to +1; JJA: -3 to +1.  SW: DJF: -3 to +4; JJA: <-5 to -1 except urban areas increase. |
| Pfister et al. (2014)  GCM-RCCM | SRES A2 climate, RCP8.5 emissions | 2046-2058 vs. 1996-2008 | JJA MDA8 | N/A | NE: -25 to -20.  SE: -25 to -15.  MW: -25 to -9.  GP: -20 to 0.  NW: -20 to -5.  SW: -20 to -5. |
| Rieder et al. (2015)  CCM | RCP4.5; mean of 3 ensemble members | 2026-2035, 2045-2055, 2091-2100 vs. 2006-2015 | 10%, 50%, 90% of regional  JJA MDA8 distribution | NE:  2030s: +1, +1, +1  2050s: 0,+1, +1  2090s: +1, +2, +1  Spatial range of mean:  -1 to +3 (2050s)  -1 to +4 (2090s).  Spatial range of 90% MDA8:  -2 to +3 (2050s)  -3 to +4 (2090s) | NE:  2030s: -4, -9, -14  2050s: -6, -16, -20  2090s: -11, -23, -30  Spatial range of mean:  -20 to -5 (2030s)  -25 to -5 (2050s)  -35 to -10 (2090s)  Spatial range of 90% MDA8:  -25 to -5 (2030s)  -35 to -10 (2050s)  -40 to -20 (2090s) |
| Trail et al. (2014)  GCM/RCM-RCTM | RCP4.5 climate; NEI 2005 emissions are projected to the future with the EPA MARKAL 9R model | 2048-2052 vs. 2006-2010 | Summer MDA8  (reported spring and fall too) | NE: -1 to +4  SE: 0 to +4  MW: -5 to +2  GP: -4 to +4 (up to +7 in TX cities)  NW: -4 to +3  SW: -5 to +4 | NE: -12 to -3  SE: -12 to -3  MW: -10 to -1  GP: -8 to +3;  NW: -6 to -1  SW: <-12 to +1 |
| val Martin et al. (2015)  CCM | RCP4.5, RCP8.5 | 9-year time slice simulations for 2050s vs. 2000s | Annual mean MDA8 | NE: 0 to +3 (RCP4.5); +2 to +5 (RCP8.5).  SE: 0 to +5 (both)  MW: +0 to +3 (RCP4.5); 0 to +4 (RCP8.5)  GP: -1 to +2 (RCP4.5); 0 to +2 (RCP8.5)  NW: 0 to +1 (RCP4.5); +1 to +2 (RCP8.5)  SW: -1 to +2 (RCP4.5); 0 to +3 (RCP8.5) | NE: -10 to -2 (RCP4.5); -5 to +1 (RCP8.5)  SE: -10 to -5 (RCP4.5); -4 to -1 (RCP8.5)  MW -7 to 0 (RCP4.5); -4 to +4 (RCP8.5)  GP: -5 to 0 (RCP4.5); -2 to +5 (RCP8.5)  NW: -5 to 0 (RCP4.5); 0 to +5 (RCP8.5)  SW: -8 to -5 (RCP4.5); -2 to + 2 (RCP8.5) |
| Gonzalez-Abraham et al. (2014)  GCM/RCM-RCTM | SRES A1B;  US anthropogenic emissions of NOx and SO2 decrease while those for other precursors increase globally | 2050s vs. 2000s; five representative summers selected from 1995-2004 vs. 2045-2054 | JJA MDA8 | NE, SE, MW: up to +9 GP: -1 to +6  NW: -2 to +2  SW: -3 to +4 | NE: -6 to +6  SE: -4 to +4  MW, GP: up to +12 and decrease up to -2 over northeast TX  NW: -3 to +5  SW: -6 to +9 |
| Fann et al. (2015)  GCM/RCM-RCTM | RCP8.5  RCP6.0, two different GCMs are employed for each scenario | 2025-2035 vs. 1995-2005 | May-Septembermean MDA8 | Spatial range of contiguous U.S.: -5 to -1 ppb, increases in some regions including central U.S. and California | N/A |
|  |  |  |  |  |  |  |
| **Surface PM2.5 (µg m-3, or as noted)** | Avise et al. (2009)  RGCM-RCTM | A2 | 5 separate Julys from 2045-2054 vs. 1990-1999 | Spatially averaged July mean | EPA Regions:  1-3 combined: +0.2  4: -3  5: -1  6: -1  7: -1  8: +0  9: -0.4  10: -0.2 | EPA Regions:  1-3 combined: +4  4: +1  5: +3  6: +2  7: +3  8: +2  9: +2  10: +2 |
| Hedegaard et al. (2013) | RCP4.5 | 2090-2099 vs. 1990-1999 | annual mean | Spatial range over contiguous U.S.: -15% to +15% | Spatial range over contiguous U.S.: -90% to -60% |
| Kelly et al. (2012)  RCM-RCTM | SRES A2 climate, RCP6.0 emissions | 2041-2050 vs. 1997-2006 | Summer mean | NE: +0.1 to 0.9  SE: +0.1 to 1.3  MW: +0.3 to 1.1  GP: -0.5 to +0.5  NW: +0 to +0.5;  SW: -0.5 to +0.5; up to +1.5 in Los Angeles | NE: -7 to 0  SE: -6 to +2  MW: -7 to +1 but >+7 in Chicago, IL  GP: -3 to +1  NW: -3 to +1  SW:-1 to +1 except larger increases and decreases near Los Angeles, CA |
| Penrod et al. (2014)  RCM-RCTM | SRES A1B | 2026-2030 vs. 2001-2005 | Summer and winter 24-hour average | N/A | NE: DJF: -2 to 0; JJA: < -5 to -1  SE: DJF:-5 to 0; JJA: <-5 to 0 MW:DJF:-2 to +1; JJA: < -5to 0  GP: DJF: -3 to 0 except up to -5 in Houston, TX; JJA: -5 to +1  NW: DJF, JJA: -2 to 0 but <-5 in Portland, OR  SW: DJF: -2 to 0; JJA: mostly -4 to +1. |
| Trail et al. (2014)  GCM/RCM-RCTM | RCP4.5 climate; NEI 2005 emissions are projected to the future with the EPA MARKAL 9R model | 2050 vs. 2010 | Annual and 4 seasons average | NE: -0.5 to +1.5  SE: -2 to +1.5  MW: -4 to +2.5  GP: -2 to -0.5  NW: -1 to +1  SW: -1.5 to +3 | NE: -4 to -1  SE: -4 to +1  MW: -4 to +1  GP: -2 to +1  NW: -1.5 to +0.5  SW: -1 to +0.5 |
| val Martin et al. (2015)  CCM | RCP4.5, RCP8.5 | 2050 vs. 2000 | Annual mean | NE: -1 to 0 (both)  SE: -1 to 0 (RCP4.5);  -0.5 to 0 (RCP8.5)  MW: -2 to 0 (RCP4.5); -1 to 0 (RCP8.5)  GP: -2 to +0 (RCP4.5); -0.5 to 0 (RCP8.5)  NW: -0.5 to 0 (both)  SW: -0.5 to +0.5 (both) | NE: -8 to -2 (RCP4.5); -10 to -2 (RCP8.5)  SE: -8 to -2 (RCP4.5); -8 to -4 (RCP8.5)  MW: -10 to -3 (RCP4.5); -10 to -2 (RCP8.5)  GP: -6 to -1 (RCP4.5); -6 to 0 (RCP8.5)  NW: -2 to 0 (both)  SW: -5 to 0 (RCP4.5); -6 to 0 (RCP8.5) |
| Gonzalez-Abraham et al. (2014)  GCM/RCM-RCTM | SRES A1B;  US anthropogenic emissions of NOx and SO2 decrease while those for other precursors increase globally | 2050s vs. 2000s; five representative summers selected from 1995-2004 vs. 2045-2054 | JJA mean | NE: -0.4 to +0.6  SE: -0.4 to 0.8  MW: 0 to +0.4  GP: -0.4 to +0.4  NW: +0.2 to +0.8  SW: +0 to +0.8 | NE: -4 to +2 (large changes concentrated in the NY and DC areas)  SE: -0.4 up to 4  MW: -0.6 to >2 (decreases in urban centers)  GP: -0.6 up to 2  NW: -0.6 to + 0.2 (increases only in and around Seattle) SW: -0.6 up to +2 (large increases in and around LA and San Diego) |

a EPA regions are shown at http://www2.epa.gov/aboutepa#pane-4

b Full-year results were reported, but we focus on the contrast between winter and summer since spring and fall changes lie in between.

**Table S4.** Atmospheric Abundances and Effective Radiative Forcing (ERF) for selected decades and NTCFs, compared to CO2, for scenarios with aggressive climate policy (RCP2.6; left, with no shading) vs. no climate policy (RCP8.5; right, with gray shading)a

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  | **2000** | **2010** | | **2030** | | **2050** | | **2100** | |
| **Global Atmospheric Abundances** | | | | | | | | | |
| **CO2** (ppm)b | 369\* | 388\* | | 431 | 449 | 433 | 541 | 421 | 936 |
| **CH4** (ppb) b | 1773\* | 1798\* | | 1600 | 2132 | 1452 | 2740 | 1254 | 3751 |
| **Trop. O3** (Tg)c | 337±23 | N/A | | 319±22 | 357±26 | N/A | N/A | 276±25 | 395±36 |
| **Sulfate** (Tg S) b,d | 1.55 | 1.57 | 2.54 | 1.21 | 1.44 | 0.94 | 1.20 | 0.71 | 0.94 |
| **BC** (Tg) b,d | 0.164 | 0.170 | 0.170 | 0.144 | 0.153 | 0.103 | 0.127 | 0.068 | 0.099 |
| **Effective Radiative Forcing (W/m2)** | | | | | | | | | |
| **CO2**b | 1.51 | 1.80 | | 2.34 | 2.56 | 2.49 | 3.56 | 2.22 | 6.49 |
| **CH4** b | 0.47 | 0.48 | | 0.42 | 0.62 | 0.36 | 0.80 | 0.27 | 1.08 |
| **Trop. O3b** | 0.36 | 0.40e | | 0.32 | 0.44 | N/A | N/A | 0.17 | 0.60 |
| **Total aerosolb** | -1.17±0.28 |  | | N/A | -0.91±0.22f | N/A | N/A | N/A | -0.12±0.03f |

\*measured.

aRCPs are shown by sector in Figure S2 and discussed in the main text (Emissions).

bFrom IPCC (2013b).

c15-model mean ± standard deviation from ACCMIP models reported in Table 1 of Young et al. (2013).

dStandard deviation across models is about 50%. 2000 and 2010 results are based on 18 models; RCP 2.6 and RCP8.5 estimates are based on 12 and 16 models, respectively. The sulfate and BC loadings do not necessarily rank with the RCPs as they do for the GHGs where higher concentrations occur in higher RCPs; see main text (Emissions)). For example, BC is highest in RCP4.5 through 2050, though some of these differences could reflect different numbers of models used in the estimates (19 for RCP4.5 versus 13 for RCP2.6) and the wide ranges in modeled distributions even with the same anthropogenic emissions (see main text and Supplemental Text S3).

e2011; the IPCC AR5 assessed value (Myhre et al., 2013).

fRCP8.5 only; see main text Figure 4a.

**Table S5**. Relative decrease in global anthropogenic SO2 and BC emissions between 2010 and 2030 for various scenarios reported in Table S5 of Rogelj et al. (2014). The year 2010 emission levels are 5.48 Tg C and 49.1 Tg S for BC and SOx, respectively (Table S4 of Rogelj et al., 2014).

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Scenario** | **Anthropogenic SO2** | | | **Anthropogenic BC** | | |
|  | REF | 450a | ABCMb | REF | 450 | ABCM |
| UNEP/WMO REF | -1% | -30% | -2% | -2% | -11% | -78% |
| Frozen legislationc | -2% | -38% |  | +5% | -15% |  |
| CLEd | -45% | -63% |  | -54% | -64% |  |
| Stringent SO2 controls | -65% | -75% |  | -71% | -78% |  |
| MFRe | -72% | -79% |  | -77% | -83% |  |

a450 refers to controls on CO2 emissions to limit atmospheric CO2 abundances to 450 ppm.

bABCM refers to All Black Carbon Measures in UNEP/WMO (2011) and Shindell et al. (2012),

roughly equal to BC reductions under MFR.

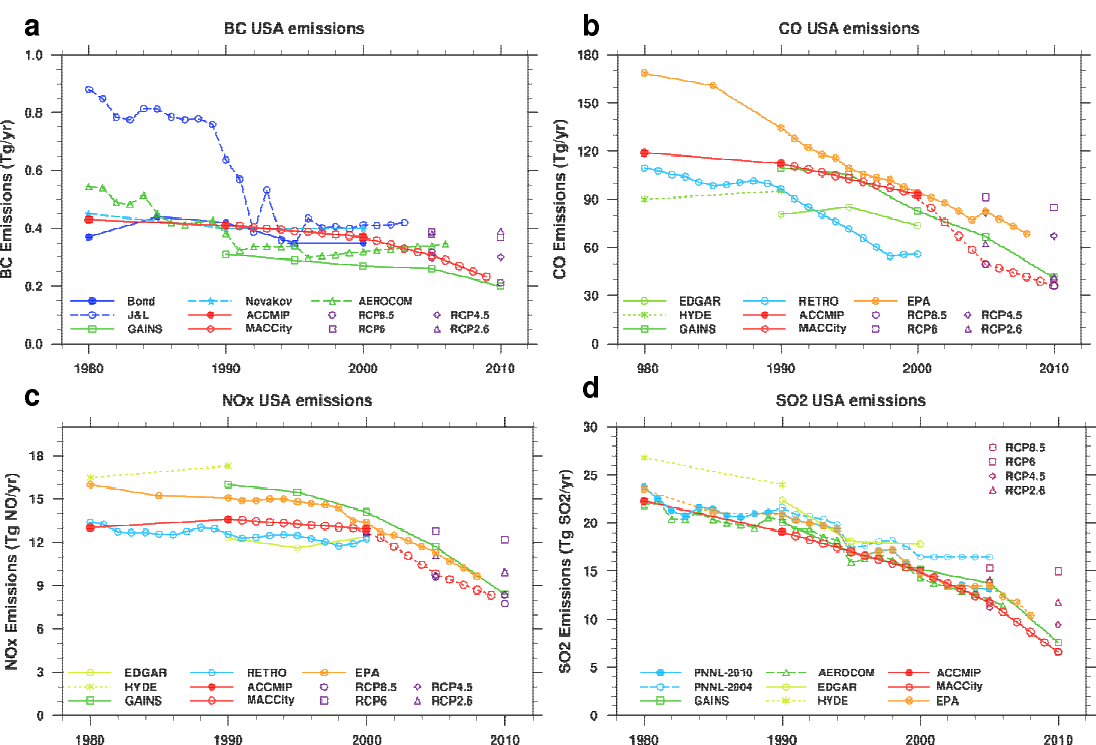
cFrozen legislation assumes that no air pollution controls are applied beyond 2005 (Rogelj et al, 2014).

dCLE is the Current Legislation Emissions scenario which assumes air pollution legislation

around the globe is implemented in the coming decades (see main text (Emissions) and Dentener et al., 2005).

eMFR is the Maximum Feasible Reductions possible with available technology applied globally

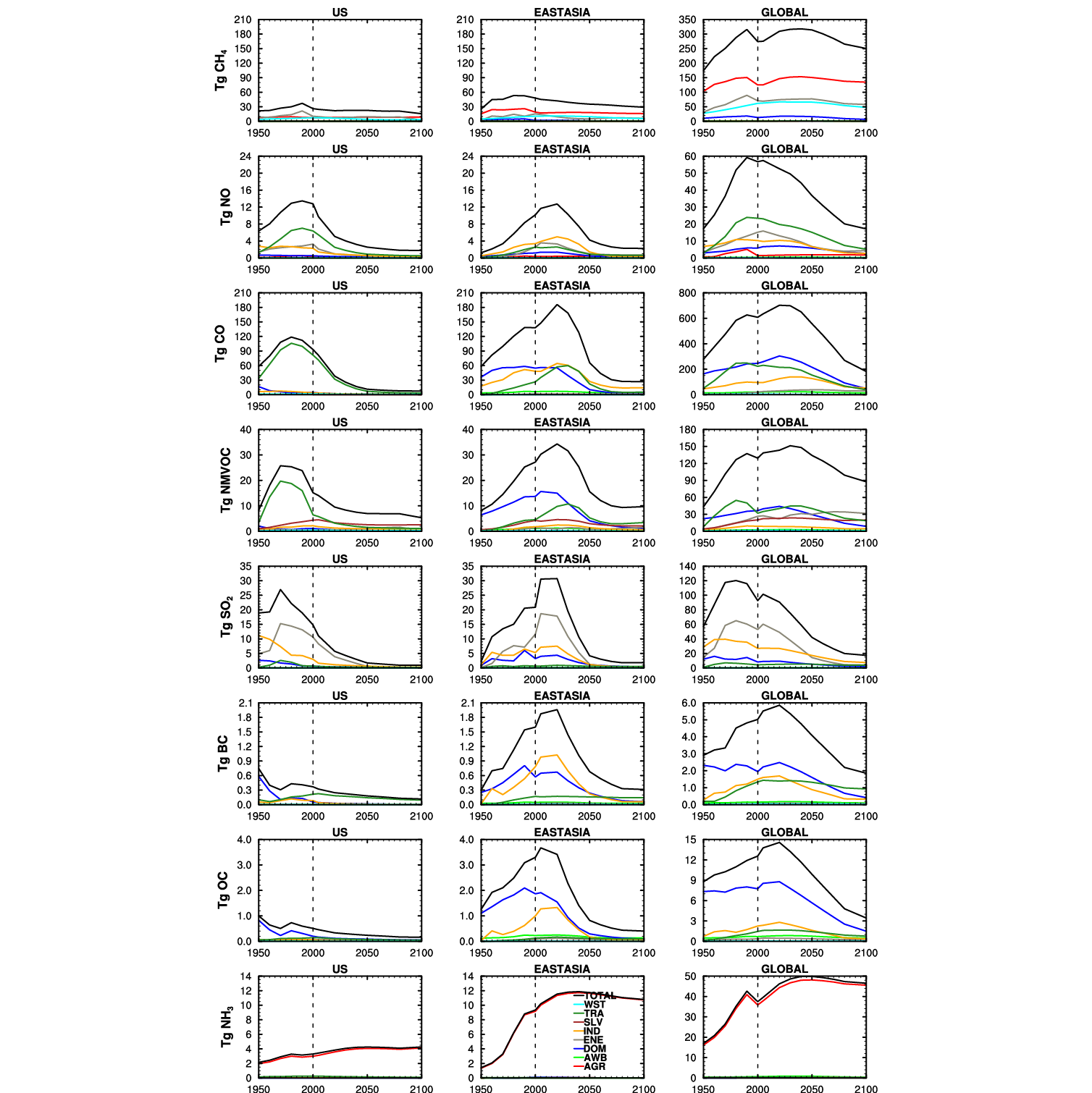
(see main text (Emissions) and Dentener et al., 2005).



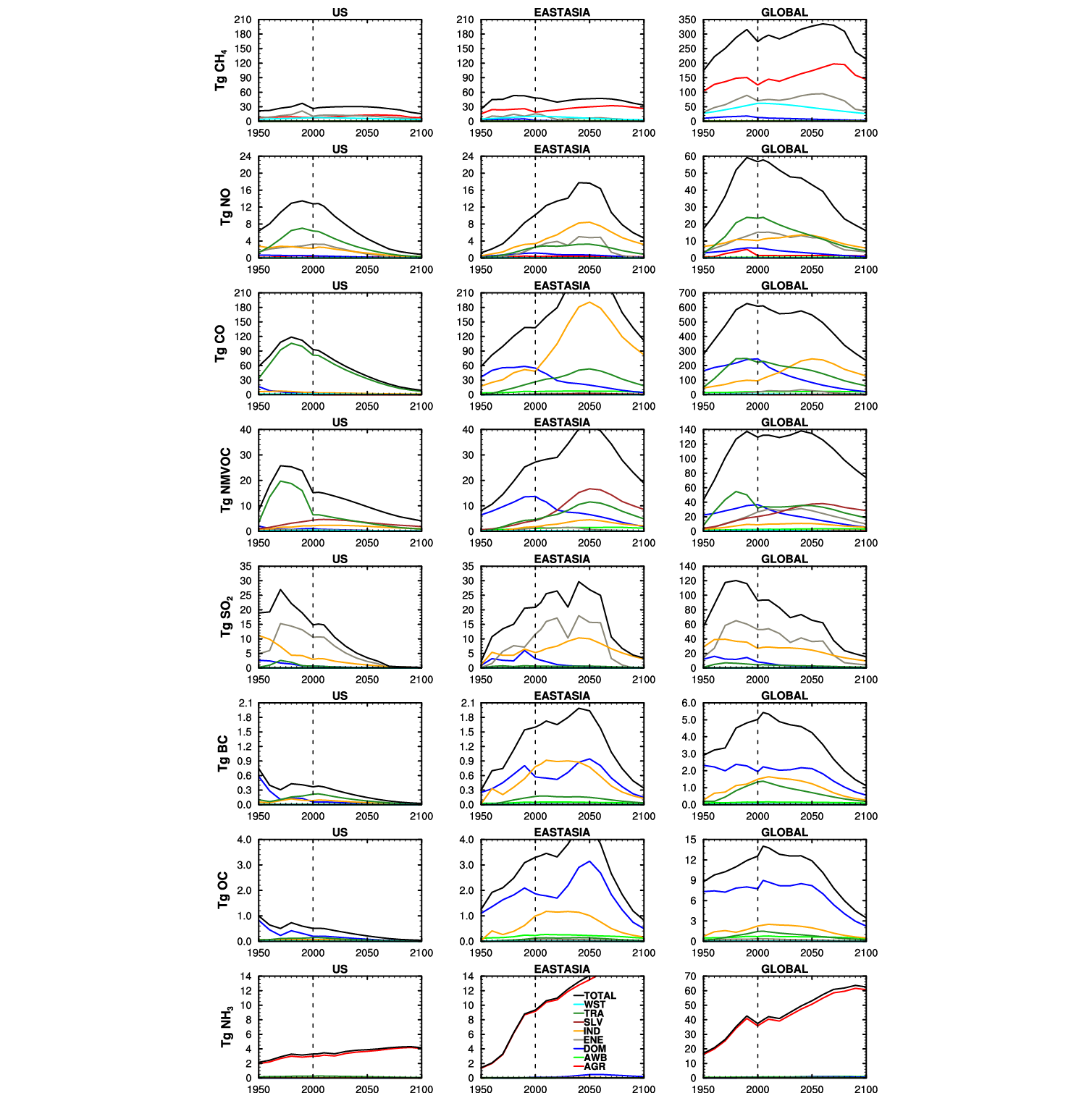
**Figure S1**. Comparison of anthropogenic emissions of a) BC, b) CO, c) NOx and d) SO2 in the United States from 1980 to 2010 across inventories (reproduced with permission from Granier et al., 2011). See Table 1 of Granier et al. (2011) for information. The MACCity inventory was derived from decadal ACCMIP emissions by linearly interpolating for each year between 1990 and 2010, with emissions after 2000 following the RCP8.5 scenario. Declining trends in pollutant emissions are generally consistent across the inventories reflecting impacts of U.S. air pollution control programs related to the Clean Air Act. Although all emissions estimates are developed as the product of activity data and emission factor, inconsistencies in these data used by emission inventory developers leads to diversity in the estimates across inventories. For example, the J&L inventory was constructed using activity data (fuel production, use, and trade) from sources that may not have considered information on emission controls within the U.S.A. High-resolution version available in separate file.

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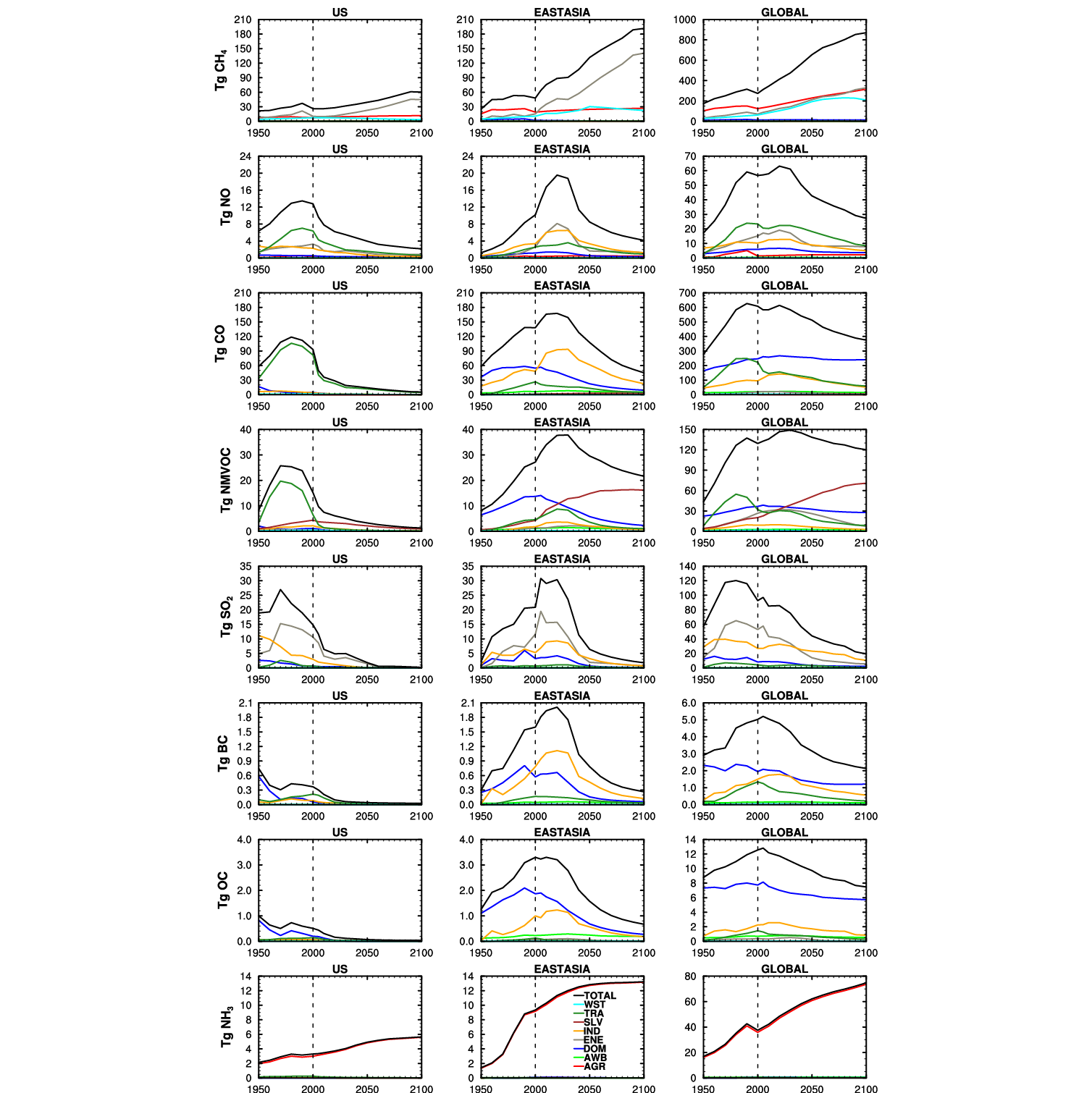
**Figure S2a.** Total and sector-based anthropogenic emissions of air pollutants and their precursors from 1950 to 2000 from Lamarque et al. (2010) and 2005-2100 following the RCP2.6 scenario for the US, East Asia, and the globe (Riahi et al., 2011). Sectors include agricultural (AGR), agricultural waste burning (AWB), residential and commercial combustion (DOM), energy production and distribution (ENE), industrial processes and combustion (IND), solvent production and use (SLV), land transport (TRA), waste treatment and disposal (WST), and sum total of all sectors (TOTAL). Global emissions do not include contribution from shipping. EASTASIA region includes China, Japan, South and North Korea. Note the different y-axis scale for GLOBAL emissions. This figure was produced from the emissions ingested by the CCMs prior to the harmonization step noted in the main text (EMISSIONS), thus resulting in small differences in the regional and/or global totals compared with those provided on <http://tntcat.iiasa.ac.at:8787/RcpDb/dsd?Action=htmlpage&page=compare>. High-resolution version available in separate file.



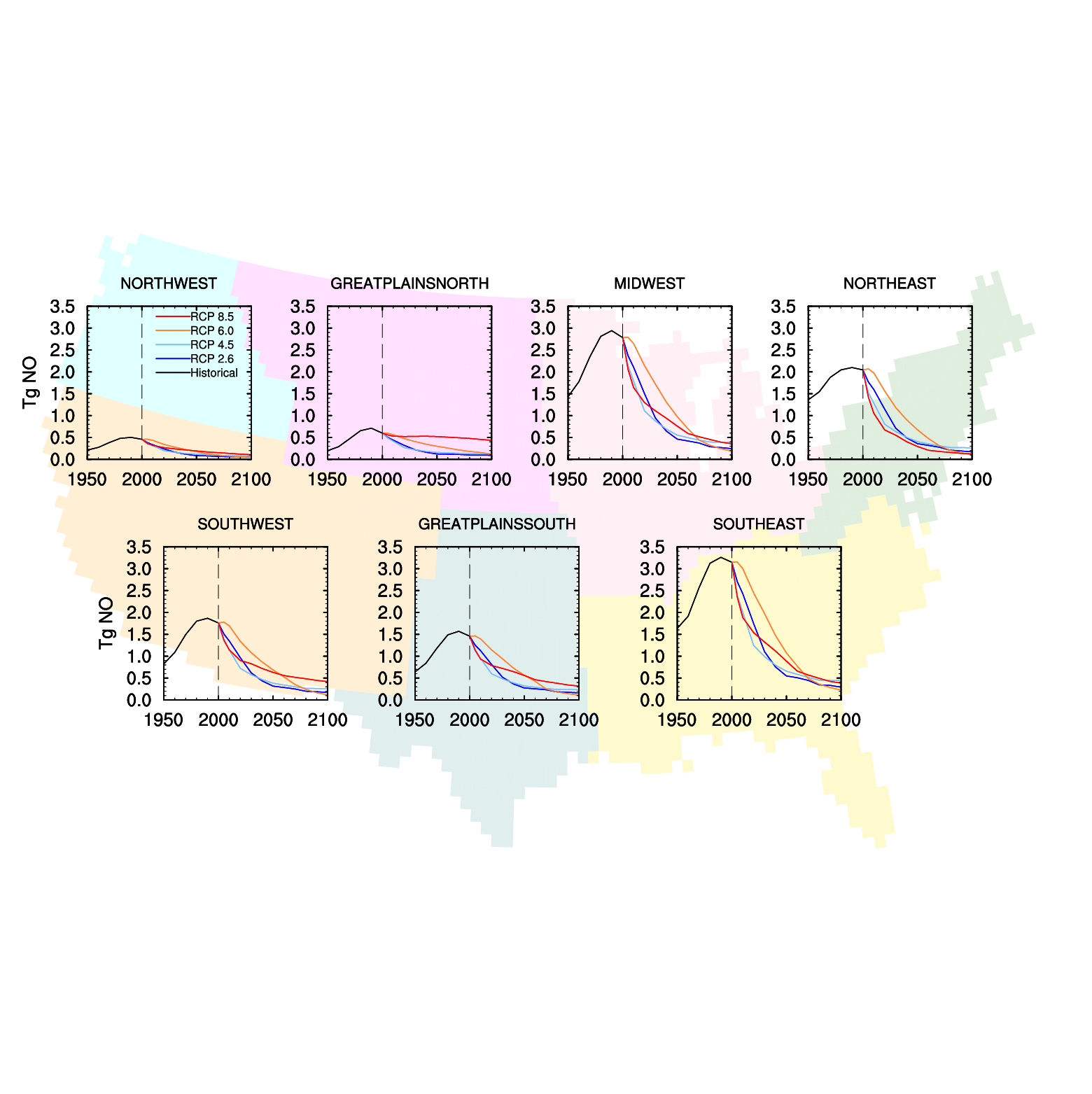
**Figure S2b**. Same as for Figure S2a but for RCP4.5. High-resolution version available in separate file.



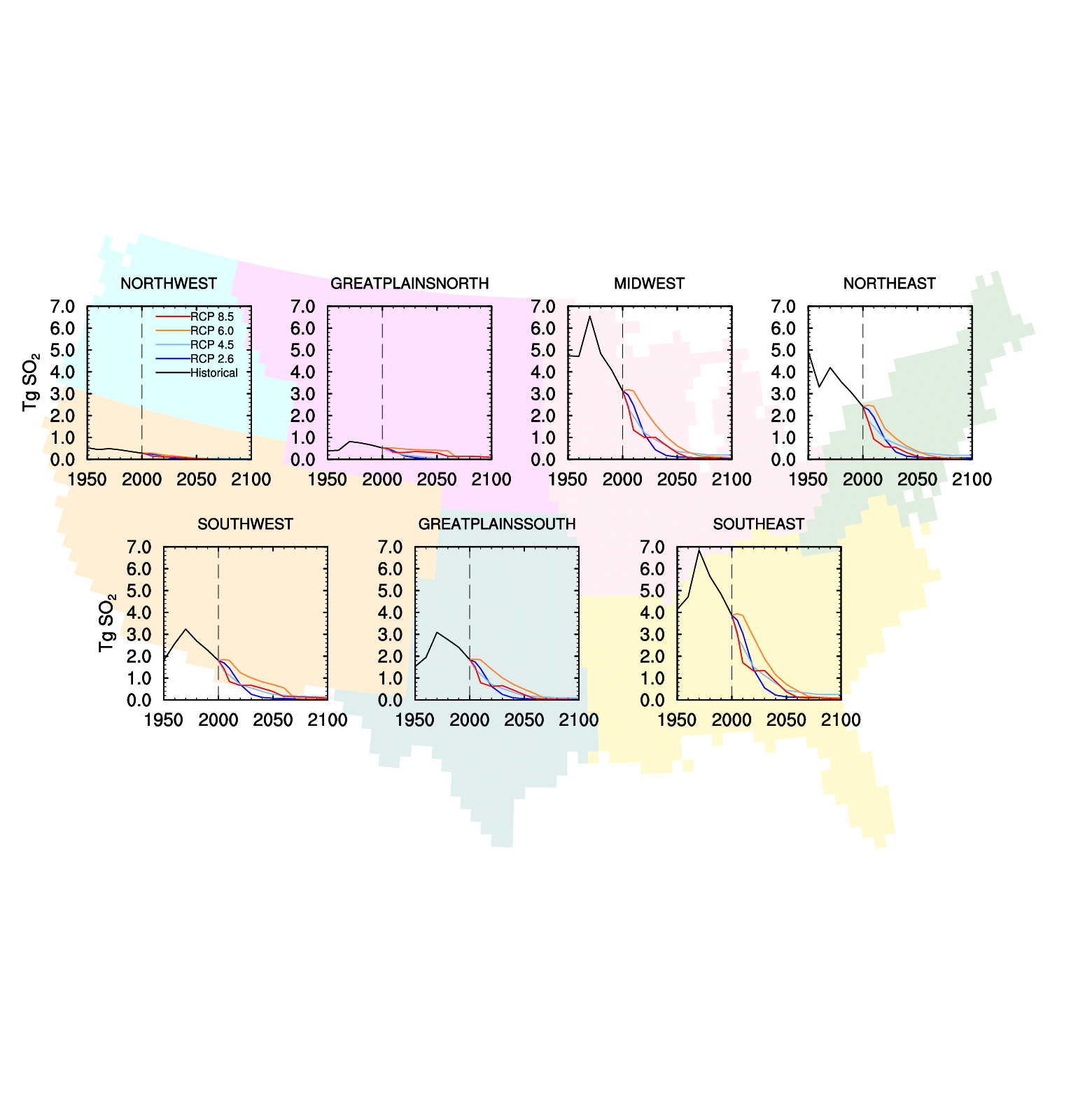
**Figure S2c**. Same as for Figure S2a but for RCP6.0. High-resolution version available in separate file.

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**Figure S2d**. Same as for Figure S2a but for RCP8.5. High-resolution version available in separate file.



**Figure S3a**. Total (anthropogenic plus biomass burning) emissions of NO from 1950-2100 following Lamarque et al. (2010) over the historical (1950 to 2000) period and van Vuuren et al. (2011) over the 2005-2100 period aggregated over seven contiguous U.S. regions defined in the Third National Climate Assessment report (Melillo et al., 2014). High-resolution version available in separate file.



**Figure S3b**. Same as for Figure S3a but for SO2. High-resolution version available in separate file.