Impacts of potential CO_2-reduction policies on air quality in the United States

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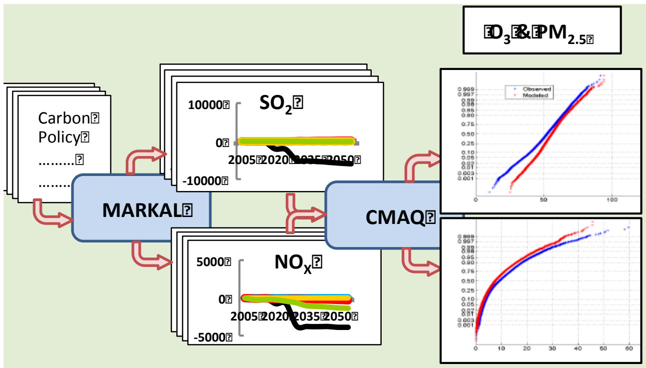
ABSTRACT



Impacts of emissions changes from four potential U.S. CO_2 emission reduction policies on 2050 air quality are analyzed using the community multi-scale air quality model (CMAQ). Future meteorology was downscaled from the Goddard Institute for Space Studies (GISS) ModelE General Circulation Model (GCM) to the regional scale using the Weather Research Forecasting (WRF) model. We use emissions growth factors from the EPAUS9r MARKAL model to project emissions inventories for two climate tax scenarios, a combined transportation and energy scenario, a biomass energy scenario, and a reference case. Implementation of a relatively aggressive carbon tax leads to improved $PM_{2.5}$ air quality compared to the reference case as incentives increase for facilities to install flue-gas desulfurization (FGD) and carbon capture and sequestration (CCS) technologies. However, less capital is available to install NO_X reduction technologies, resulting in an O_3 increase. A policy aimed at reducing CO_2 from the transportation sector and electricity production sectors leads to reduced emissions of mobile source NO_X, thus reducing O_3. Over most of the U.S., this scenario leads to reduced $PM_{2.5}$ concentrations. However, increased primary $PM_{2.5}$ emissions associated with fuel switching in the residential and industrial sectors leads to increased organic matter (OM) and $PM_{2.5}$ in some cities.

INTRODUCTION

Air pollution has been shown to adversely impact ecosystem and human health, and future global changes in climate, emissions and land use are expected to impact air pollution.[1–5] Recently, the World Health Organization (WHO) characterized air pollution as a class 1 carcinogen[6] and the Global Burden of Disease study[7,8] found that exposure to ambient particulate matter (PM) and ozone (O_3) are major contributors to premature death. For decision-makers to appropriately mitigate future air pollution, the impact of future changes in emissions, population, land-use and climate should be considered. Of particular concern is the air quality impact of climate mitigation policies. A major source of uncertainty in predicting future air quality lies in projecting future emissions of pollutant precursors. However, recent modeling advances have shown potential to capture air quality trends[9,10] and account for complex interactions between driving forces such as population growth, socio-economic development, technology change, and environmental policies.[11] In this study, we assess the impact of four potential climate mitigation policies on air quality in the U.S. in a future (2050) climate by using recent advances in climate downscaling and emissions projection approaches.

Significant work on investigating the impact of future climate change on air pollutant concentration has been realized to date.[1,5,12] A general consensus among studies is that future climate change can cause increased O_3 concentration in some regions of the U.S., though changes in PM will likely be small and variable. While these studies focus on air pollution changes due to climate change, some recent studies have addressed the impact of future changes in emissions as well.[2,12–15] Hogrefe et al.[14] used the Community Multiscale Air Quality (CMAQ) model with inputs of downscaled future climate[16] and anthropogenic emissions according to A1B projections of the Asian Pacific Integrated Model (AIM); decreased O_3 was found over most of the U.S., despite the tendency for rising temperatures to increase O_3 concentration. Hogrefe et al.[14] suggested assessing the impacts of alternative emissions scenarios on $PM_{2.5}$ concentration. Tagaris et al.[12] projected emissions to the near future (2020) using the 2020 Clean Air Interstate Rule (CAIR) emissions inventory and to the distant future (2050) using the Integrated Model to Assess the Global Environment (IMAGE). Maximum daily 8-hr average (MDA8) O_3 and aerosol concentrations decreased over most of the U.S. due to the emissions reductions. Other studies specifically address the co-benefits of reducing greenhouse gas emissions for air quality.[15,17–19] For example, McCollum et al.[15] linked the Greenhouse Gases and Air Pollution Interactions and Synergies (GAINS) model and MESSAGE (Model for Energy Supply Systems And their General Environmental impact) integrated assessment model to develop an ensemble of future global energy scenarios and study the expected impacts on human health related to air pollution. While this study found that efforts to reduce CO_2 emissions lead to improve air quality, McCollum et al.[13] stress the need for comparison among various models used to predict future emissions impacts on air pollution.

In this study, we use a chemical transport model (CTM) to simulate air pollutant concentrations and apply recent climate downscaling and emissions modeling advancements to assess a suite of detailed future emissions scenarios of a future year chosen for their potential to mitigate climate change. In particular, we use the EPA U.S. 9-region national database (EPAUS9r)[20] with the MARKet Allocation (MARKAL v1.1, November 2012) energy system model[11,21] to develop emissions scenarios and spectral nudging to downscale global climate[22,23] to the regional scale over the U.S. The benefits of using spectral nudging to downscale global climate are described in Lui et al.[22] The MARKAL energy system model selects from available technologies to provide the least-cost path that satisfies specified demands of the residential, commercial, industrial, and transportation sectors for regionally-based energy services. The flexible modelling framework allows examination of mid-to-long-term technology choices as well as specific policy options that shape the evolution of an energy system in meeting specific environmental or other goals. MARKAL serves as a useful tool to identify the likely technologies that will be used to meet greenhouse gas or criteria air pollutant-related policies and objectives. Various versions of MARKAL are used in previous studies to estimate emissions for investigating air quality changes due to the implementation of policies aimed at reducing emissions of greenhouse gas (GHG) and air pollutants in Shanghai and Beijing[24–26] and in developing countries such as Nepal[27] and Pakistan.[28] To our knowledge, this is the first study to use MARKAL to investigate the effect of CO_2 reduction strategies on air quality in the U.S.

CMAQ[29] is used to analyze the impact of emissions changes from four potential climate change mitigation policies on regional air quality in 2050 in the contiguous United States; these policies are compared to a reference case scenario, which represents a "business as usual" policy scenario. We use growth factors from MARKAL[30] to develop the 2050 emissions inventory and the Sparse Matrix Operator Kernel Emissions (SMOKE) model to provide spatial and temporal variation of emissions. Future meteorology was downscaled from the Goddard Institute for Space Studies (GISS) ModelE2 General Circulation Model (GCM) to the regional scale using the Weather Research Forecasting (WRF) model with spectral nudging.[22] Trail et al.[23] provide a detailed description of meteorology used in the present study and compare present (2006-2010) and future (2048-2052) regional climate from an air quality perspective. Trail et al.[23] also conducted an extensive evaluation of the 2006-2010 results using observations from the same period. In a previous study[31], we used the CMAQ model to compare present (2006-2010) and future (2048-2052) air pollutant concentrations and their sensitivities to emissions from different sectors for the reference case emissions scenario and found decreased O_3 and $PM_{2.5}$ concentrations over most of the U.S. In the present study, we compare air pollutant concentrations, including O_3 and $PM_{2.5}$, in the reference case for the year 2050 with two climate tax policy scenarios (CT1 and CT2), a combined transportation energy sector policy scenario (TE) and a biomass energy policy scenario (BE). We chose four of the six CO_2 emission reduction strategies considered in Rudokas et al.[30] to examine a variety of different air quality outcomes. We also analyze air pollutant concentrations and National Ambient Air Quality Standards (NAAQS) exceedances in major U.S. cities and provide a discussion of the implications of the results of this study.

METHODS

Responses of future air pollutant concentration to climate mitigation policies are simulated using a CTM with inputs of emissions from multiple policy scenarios and downscaled meteorology. Emissions inputs are prepared for the CTM using an energy system cost optimization model. Components of the modeling system are described below

Meteorology. The GISS ModelE2 provides the initial and boundary conditions to a regional climate model for the years 2006-2010 and 2048-2052.[32] The global simulation has a horizontal resolution of 2°×2.5° latitude by longitude and 40 layers, following a sigma coordinate up to 150 hPa with constant pressure layers between 150 and 0.1 hPa. Future atmospheric conditions over the 21st century which follow the scenario development process for IPCC AR5 drive the simulations. The "Representative Concentration Pathway" (RCP) 4.5[33,34] is used for this study, being a scenario of decadal global emissions of greenhouse gases, short-lived species, and land-use-land-cover which produces an anthropogenic radiative forcing at 4.5 W m^{-2} (approximately

650 ppm CO_2-equivalent) in the year 2100.[34] While the model calculates significantly different temperatures between 2000 and 2050, the temperatures are not very different between RCPs in 2050 so we apply the RCP4.5 future climate scenario to all of the future emissions scenarios in this study. The GISS simulation was originally spun up from 1850. However, the GISS model was reapplied, using the original base GISS simulation, to provide higher frequency results than were originally available. These simulations were initiated with a three year re-initialization spin-up, starting in 2003, and 2045. Instantaneous outputs of physical parameters were produced at 6-hr intervals for regional downscaling by WRF. The WRF Model[35] (version 3.4) is used to downscale GISS simulations for the years 2006-2010 and 2048-2052 with 10 day spin-up times. The present study only uses meteorological results from the years 2010 and 2050 which were average years during those five year periods but also includes periods of summer stagnation. The model domain covers the contiguous U.S. (CONUS) and portions of southern Canada and northern Mexico and is centered at 40°N and 97°W with 164×138 horizontal 36 x 36 km grids cells (Figure s1). Details of regional climate downscaling work have been reported elsewhere and the ability of GISS-WRF to reproduce the long-term yearly climatic means and the meteorological fields that strongly impact air quality are evaluated in Trail et al.[23]

Emissions. The NEI energy related emissions of SO_2, NO_X, VOC, CO, NH_3 and $PM_{2.5}$ are projected to the years 2010 and 2050 for a reference case and for four alternative emissions scenarios (2050 only) using projection factors calculated using the EPAUS9r with MARKAL.[21] MARKAL models future energy dynamics of the energy systems in the nine Census Divisions of the U.S. (Figure s1). NEI emissions are scaled by multiplying the original emissions inventory by the projection factors from MARKAL. The reference case emissions scenario assumes the implementation of the following policies: Clean Air Act Title IV (Acid Rain Program) SO_2 and

NO_X requirements, CAIR, Utility Mercury and Air Toxics Standards (MATS), aggregated state Renewable Portfolio Standards (RPS) by region, Federal Corporate Average Fuel Economy (CAFE) standards as modeled in AEO 2012, Tier 2 light duty vehicle tailpipe emission standards and heavy duty vehicle fuel and engine rules. Projections of non-energy related emissions were calculated according to the A1B emissions scenario developed by the Intergovernmental Panel on Climate Change Special Report on Emissions Scenarios (IPCC SRES)[36], though these changes in SO_2, NO_X and $PM_{2.5}$ emissions are small compared to the changes in energy related emissions from the MARKAL projections.

Four alternative emissions scenarios, including two carbon tax scenarios (CT1 and CT2), a combined transportation energy scenario (TE) and a biomass scenario (BE), with very different emission outcomes were chosen from Rudokas et al.[30] The scenarios were developed by assuming the implementation of various climate mitigation policies in addition to the policies assumed in the reference case. CT1 represents a carbon tax option with taxes beginning in 2015 at \$20 per ton CO_2 and reaching \$90 per ton in 2050, while CT2 is a more aggressive option with taxes beginning in 2020 at \$50 per ton and reaching \$1,400 per ton in 2050. The CT1 and CT2 carbon taxes are applied economy wide and in nominal dollars. The CT2 scenario is intended to represent an upper and lower end of carbon tax options with the CT1 and CT2 scenarios. Further consideration of energy efficiency improvement, either as a strategy or in response to higher taxes, is an important consideration to be further investigated. TE assumes a 70% GHG reduction from transportation sectors and an additional electricity sector emission rate limit of 880 lb/MWh for CO_2, 0.0058 lb/MWh for SO_2 and 0.14 lb/MWh for NO_X, which is similar to that of new combined cycle natural gas power plants. The purpose of the additional limit on the electricity sector is to mitigate increased emissions from electric generation due to increased use of electric vehicles. Finally, BE assumes that all available biomass will be used in the energy sector. Rather than predetermining which sectors the biomass would be directed to or how the biomass would be employed, MARKAL uses linear programing methods to select the least cost set of technologies and fuel sources to meet the prescribed level of end-use energy demand.

Hourly, gridded and speciated emissions are generated for input to CMAQ using the SMOKE V3[37] model which uses inputs from the 2005 National Emissions Inventory (NEI). The Biogenic Emissions Inventory System (BEIS) and the Biogenic Emissions Landcover Database 3.0 (BELD3) are used in SMOKE to compute hourly emissions from U.S. vegetation. Fire emissions, representing average emissions of a typical year from the 2005 NEI, are constant for all simulations. Natural biogenic emissions change as a function of meteorology but are not projected like anthropogenic emissions. Lightning NO_X emissions are not included in the simulations. Kaynak et al.[38] used CMAQ to simulate ozone production due to lightning NO_X emissions. They conducted simulations with and without lightning NO_X and that MDA8 O_3 changes due to lightning NO_X were small. The resulting inventory consists of pollutants emitted from area, mobile, point, fire, ocean, biogenic, and agricultural sources.

Air Quality. Simulations of the transformation and fate of air pollutants for the four alternative emissions scenarios in the year 2050 and for the present (2010) and future (2050) year reference cases are carried out using the CMAQ 4.7.1 model.[29] Gas-phase chemistry is modeled using the SAPRC-99[39] chemical mechanism. The domain covers the entire continental US as well as portions of Canada and Mexico (5328×4032 km) (Figure s1) using a 36-km horizontal grid-spacing with thirteen vertical layers extending ~15.9 km above ground. The first layer is 18 m thick and there are 7 layers below 1 km. The modeling domain uses a Lambert Conformal Projection centered at 40ºN, 97ºW with true latitudes of 33ºN and 45ºN. Boundary conditions are adapted from an annual GEOS-Chem simulation[40] and are dynamic over the course of a year, although they are constant between the present and future year simulations in order to isolate the impact of regional climate change and changing emissions on US air quality. The top of our CMAQ domain goes well into the stratosphere and ozone from the stratosphere does penetrate into the troposphere. However, it is recognized it is difficult for models such as CMAQ to fully capture this process. Stratospheric intrusion events may be captured at the boundaries since they were adapted from GEOS-Chem. The present study focuses on relatively low elevation cities where stratospheric intrusion is not as large of a source of surface ozone. Default initial conditions of air pollutant concentrations are used here with a spin-up period of 10 days for each simulation.

In a previous study, Trail et al.[31] compare present (2006-2010) and future (2048-2052) air quality using the same methods as in the present study for the reference case emissions scenario. 2010 and 2050 air quality model results were found to be typical compared to their respective time periods. The same study also evaluated the simulated present day air quality with observations and found that simulated O_3 and $PM_{2.5}$ agreed well with observations. In particular, they found that, while simulated MDA8 is biased high, the results agree best with observations at higher MDA8 concentration in most regions. The annual mean $PM_{2.5}$ normalized mean bias (NMB) was -21% with the largest negative bias occurring during the summer. Results from Trail et al.[31] reveal that the simulated 98th percentile highest 24-hr average $PM_{2.5}$ concentrations agree well with observations in most regions."

RESULTS

The changes from the reference case of emissions rates of major air pollutants from 2010 to 2050 for six CO_2 emissions scenarios, including the four scenarios analyzed in this study, are described extensively in Rudokas et al.[30] For the first carbon tax scenario, CT1, they found a 20% reduction in SO_2 emissions from the combined industry and electricity sectors and little change in NO_X emissions in 2050 versus the 2050 reference case (Figure s2). The CT2 scenario, on the other hand, leads to a 61% decrease in SO_2 emissions from the electricity sector (from 1.746 to 0.679 Tg yr^{-1}) and a 20% decline from industry sectors (from 1.264 to 1.019 Tg yr^{-1}) (Table 1) compared to the 2050 references case. Although renewables double by 2050 in the CT2 scenario relative to the reference case, increased use of renewables does not have a noticeable impact on total emissions. Decreased SO_2 emissions result from the increased use of flue-gas desulfurization (FGD) process technologies because the flue gas must have low SO_2 content in order for the carbon capture and sequestration (CCS) technologies to be effective. However, electricity sector NO_X emissions match the reference case emissions through 2025 but increase by 20% by 2050 for the CT2 scenario. The lower reduction in electricity sector NO_X emissions under the more aggressive carbon tax scenario relative to the reference case is a result of reduced investment in NO_X controls and increased coal generation NO_X emissions in states that are not subject to the Clean Air Interstate Rule's NO_X cap. EPAUS9r projects a 35% decrease in NO_X control technology investments with the more aggressive CT2 scenario than the 2050 reference case scenario. Increased NO_X emissions from coal generation increase in regions that are not subject to the Clean Air Interstate Rule's NO_X cap occur after the year 2025. It should be noted that while NO_X emissions do increase in the electricity sector for the CT2 scenario, the assumed NO_X regulations modeled in Rudokas et al.[30] (e.g., Acid Rain Program, Clean Air Interstate Rule) are still binding.

Table 1. Annual emissions of SO_2 and NO_X (Tg yr^{-1}) from the commercial, residential, industrial, electricity and transportation sectors and total emissions simulated in MARKAL for the 2005 and 2050 reference case and the four alternative emissions scenarios.[30]

	SO_2 Emissions (Tg yr^{-1})					
Sector	**2005**	**2050**	**CT1**	**CT2**	**TE**	**BE**
Commercial	0.187	0.140	0.138	0.124	0.186	0.139
Residential	0.148	0.055	0.055	0.050	0.061	0.055
Industrial	1.248	1.264	1.197	1.019	1.424	1.250
Electricity	9.296	1.746	1.452	0.679	0.351	1.567
Transportation	1.362	0.106	0.105	0.099	0.086	0.106
Total	12.235	3.311	2.946	1.970	2.109	3.117
	NO_X Emissions (Tg yr^{-1})					
Sector	**2005**	**2050**	**CT1**	**CT2**	**TE**	**BE**
Commercial	0.170	0.219	0.207	0.172	0.206	0.221
Residential	0.366	0.335	0.337	0.290	0.340	0.333
Industrial	1.179	2.319	2.355	2.420	1.966	2.299
Electricity	3.317	1.897	1.862	2.217	0.817	1.923
Transportation	11.106	3.389	3.389	2.933	2.071	3.358
Total	16.137	8.158	8.150	8.032	5.400	8.135

SO_2 emissions in the TE scenario increase from 140 in the 2050 reference case to 186 thousand tons per year (33%) in the commercial sector but decrease in the transportation sector by 19% (from 0.106 to 0.086 Tg yr^{-1}) and in the electricity sector by 80% (from 1.746 to 0.351 Tg yr^{-1}). NO_X emissions from the industrial, electricity and transportation sectors decrease from the 2050 reference case by 15%, 57% and 39%, respectively (decreases of 0.353, 1.080 and 1.318 Tg yr^{-1}). The dramatic reduction in NO_X and SO_X from the electricity sector resulted from the assumptions made by Rudokas et al.[30] regarding the emissions rates for coal plants in the TE scenario. The MARKAL analysis assumed the emissions rates for all coal plants would conform to the standards of a new combined cycle natural gas plant after 2020. The rationale for the emissions rate assumption in the transportation scenario was to examine the implications of both a clean transportation and clean grid future because battery electric vehicles and plug-in hybrid electric vehicles are the two primary technologies deployed to meet the transportation GHG target. The emissions characteristics of the recharging infrastructure (i.e., electricity grid) will greatly affect the implications of a low carbon transportation future. The BE scenario leads to a 10% decrease in emissions of SO_2 from the reference case by 2050 and a slight decrease in NO_X emissions.

Ozone. Using the reference case emissions scenario and comparing present and future air quality, Trail et al.[31] found that the O_3 mixing ratio is expected to decrease in the future over much of the U.S. despite the tendency for climate change to increase O_3 mixing ratio. Decreased O_3 over time, according to Trail et al.,[31] is mainly attributed to decreased emission rates of ozone precursors (e.g. VOC, CO and NO_X) from mobile sources in response to the increased fraction of vehicles meeting current standards along with further decreases in NO_X from electricity generation.

A site is in non-attainment of the current NAAQS standard for O_3 if the 4th highest MDA8 O_3 mixing ratio for the year, averaged over three consecutive years, is greater than 75 ppb. Here we compare MDA8 mixing ratios of four emissions scenarios to the reference case for the year 2050. The TE scenario shows the largest MDA8 decreases from the future reference case while the CT2 scenario leads to increased MDA8 and the CT1 and BE scenarios have little impact on MDA8 O_3 concentrations (Figure s3). During the summer, seasonal average MDA8 is up to 4 ppb greater for the CT2 scenario than the reference case over most of the eastern U.S. and parts of the Mountain region. Increased MDA8 concentrations in the CT2 scenario is caused by the higher NO_X emissions as explained previously. The 4th highest MDA8 concentration for the CT2 scenario also increases from the reference case by between 2 to 6 ppb over the eastern U.S. (Figure 1). In the CT2 scenario, Atlanta, Chicago, New York, Philadelphia, Los Angeles and Phoenix all experience an increase in the number of days with MDA8 concentrations exceeding the NAAQS standard of 75 ppb (Table 2). The CT1 scenario, on the other hand, shows only small changes in 4th highest MDA8 concentrations and number of days with exceedances in the major cities.

Decreases in NO_X emissions from the electricity and transportation sectors lead to decreases of MDA8 concentration in the TE scenario over much of the U.S. Seasonal average MDA8 concentration decreases over most of the U.S. by up to 5 ppb during the spring and fall and by over 10 ppb during the summer with the largest decreases occurring over the eastern U.S. (Figure s3). The 4th highest MDA8 concentration also decreases by up to 20 ppb over most of the eastern U.S. and parts of the Pacific regions (Figure 1). The 4th highest MDA8 of the year is lower for the TE scenario than the reference case in every city analyzed with the largest decreases occurring in Atlanta and Philadelphia of 15 ppb and 10 ppb, respectively (Table 2). The 4th highest MDA8 of the year in New York exceeds the NAAQS standard of 75 ppb in the reference case but the NO_X emission reductions in the TE scenario lead to decreases in 4th highest MDA8 to below the standard. In Los Angeles, decreased NO_X emissions over time in the reference case lead to a decrease in 4th highest MDA8 concentration from 110 ppb in 2010 to 94 ppb in 2050, however the number of days exceeding the standard only decreases slightly from 45 to 41 days. In the TE scenario, further reductions in NO_X emissions lead to a large reduction in the number of days exceeding the standard (from 41 to 28 days) and decrease in the 4th highest MDA8 mixing ratio (from 94 ppb to 87 ppb). For the BE scenario, the change in O_3 is very small.

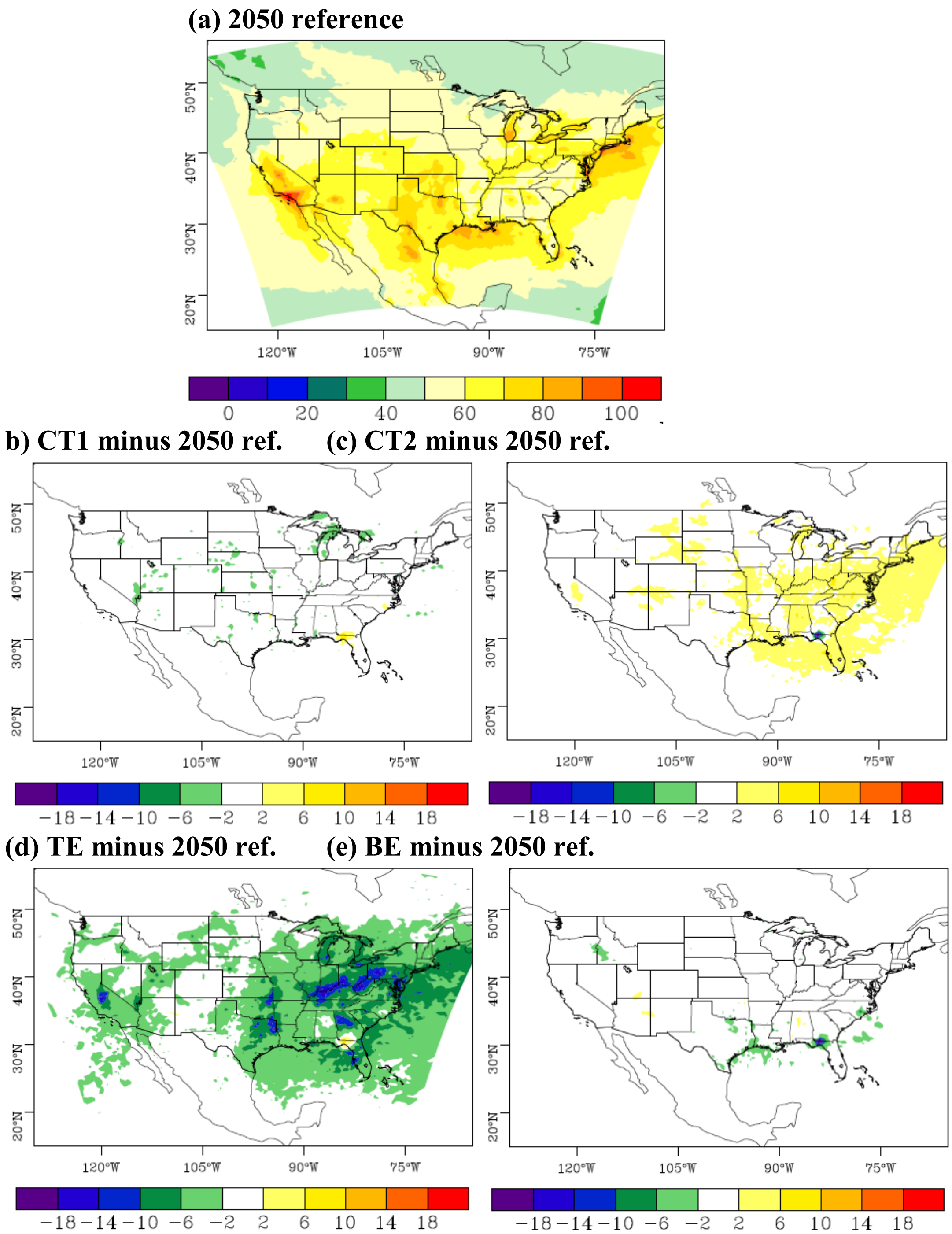


Figure 1. (a) 4th Highest MDA8 ozone of the year (ppb) for the future year (2050) reference case and the change in 4th highest MDA8 ozone for the (b) low carbon tax scenario (CT1 minus reference), (c) high carbon tax scenario (CT2 minus reference), (d) transportation and energy scenario (TE minus reference) and (e) biomass energy scenario (BE minus reference)

Table 2. 4th highest MDA8 ozone (ppb) of the year and the number of days where MDA8 exceeded 75 ppb in parentheses for the future (2050) year reference case and for the four alternative emissions scenarios

City	2050	CT1	CT2	TE	BE
Atlanta	74 (1)	73 (3)	75 (3)	59 (0)	74 (1)
Chicago	72 (1)	71 (2)	74 (2)	68 (1)	72 (1)
Los Angeles	94 (41)	94 (42)	94 (42)	87 (28)	94 (42)
New York	81 (14)	82 (12)	86 (20)	74 (3)	82 (14)
Philadelphia	74 (3)	74 (3)	78 (9)	64 (1)	74 (3)
Phoenix	89 (29)	114 (36)	89 (30)	87 (22)	86 (29)
Seattle	62 (0)	62 (0)	62 (0)	61 (0)	62 (0)

Table 3. Highest 98th % 24-hr average $PM_{2.5}$ ($\mu g\ m^{-3}$) of the year and mean annual $PM_{2.5}$ ($\mu g\ m^{-3}$) for the future (2050) year reference case and for the four alternative emissions scenarios

	2050		**CT1**		**CT2**		**TE**		**BE**	
City	**98th %**	**Mean**	**98th %**	**Mean**	**98th %**	**Mean**	**98th %**	**Mean**	**98th %**	**Mean**
Atlanta	22.4	9.1	20.3	9.8	18.6	8.2	20.3	9.2	22.2	10.3
Chicago	24.8	9.2	26.8	9.9	25.3	8.7	26.1	9.3	28.0	10.2
Los Angeles	18.4	8.6	20.2	9.3	19.2	9.0	19.2	8.9	20.2	9.5
New York	32.5	11.2	34.2	12.6	32.7	10.7	43.8	14.1	38.0	13.2
Philadelphia	28.5	9.4	29.2	10.1	28.2	8.4	32.7	9.4	31.3	10.6
Phoenix	10.4	6.5	11.2	6.9	9.8	6.0	9.5	5.7	10.8	6.8
Seattle	17.5	6.7	20.5	7.6	20.4	7.4	19.2	7.0	21.5	8.0

Particulate Matter ($PM_{2.5}$). In the CT2 scenario, the scenario with the largest reductions in $PM_{2.5}$ concentrations, reductions in SO_2 emissions from the electricity sector (~60% reduction), lead to reductions in sulfate aerosol concentrations with the highest reduction taking place during summer (Figure 2 and Table 3). Average annual sulfate aerosol concentrations in Atlanta, New York and Philadelphia are over 1 μg m^{-3} lower for the CT2 scenario compared to the reference case (Table 4). Decreased sulfate aerosol accounts for lower annual average $PM_{2.5}$ concentrations in Atlanta (from 9.1 to 8.2 μg m^{-3}) and Philadelphia (from 9.4 to 8.4 μg m^{-3}) and a decrease in the 98 percentile 24-hr $PM_{2.5}$ by 3.8 μg m^{-3}. The 98% highest $PM_{2.5}$ concentration increases slightly in Chicago, Los Angeles and Seattle (Table 3). The CT1 scenario, on the other hand, sees slight increases in annual average and peak 24-hr $PM_{2.5}$ concentration and increased organic matter (OM) aerosol in every city analyzed. As the demand for natural gas increases in the CT1 and CT2 scenarios, the residential and industrial sectors tend to use more cost effective fuels the result in higher emissions of primary $PM_{2.5}$, including OM and BC (black carbon), and the increased $PM_{2.5}$ concentrations in the CT1 scenario. (Rudokas et al. supplementary material[30]). However, in the CT2 scenario, the increased primary PM emissions are not enough to overcome reduced sulfate aerosol concentration.

In the TE scenario, the scenario with the second largest $PM_{2.5}$ decreases, decreased emissions of NO_X from the mobile sectors and electricity sectors account for lower seasonal average $PM_{2.5}$ concentrations during the wintertime, since NO_X is converted to nitrate aerosol and is a major component of $PM_{2.5}$ during winter (Figure 2 and Table 4). The largest $PM_{2.5}$ decreases, up to 4 μg m^{-3} occurring over the eastern U.S. in the summer, result from lower SO_2 emission rates from the electricity and transportation sectors in the eastern regions. Lower sulfate aerosol concentrations account for decreased summertime $PM_{2.5}$ concentration in particular since sulfate is typically most abundant during summer. Although $PM_{2.5}$ tends to decrease in most eastern U.S. regions, annual average $PM_{2.5}$ and the 98th percent 24-hr $PM_{2.5}$ increases in most urban areas, exceptions being Atlanta and Phoenix (Table 3). In particular, the 98th percent 24-hr $PM_{2.5}$ in New York increases from 32.5 to 43.8 μg m^{-3}. The increased urban $PM_{2.5}$ in New York corresponds to increased urban OM concentration, which more than doubles in annual average concentration, from 2.3 to 5.0 μg m^{-3} (Table 4), and increased urban elemental carbon (EC) concentrations. While light-duty vehicles shift from gasoline to electric in the TE scenario, residential fuel use switches result in increased emissions of primary $PM_{2.5}$, organic carbon (OC), and EC (see Rudokas et al. supplementary material[30]). Increased emissions of primary OM and $PM_{2.5}$ aerosol also leads to increased annual $PM_{2.5}$ concentrations of up to 2 μg m^{-3} over Portland, Dallas, Houston, Austin, Minneapolis and San Francisco (Figure 2).

In the BE scenario, increases in annual $PM_{2.5}$ concentration are seen over the eastern U.S. of 1 - 2 μg m^{-3} relative to the reference case (Figure 2). Sulfate aerosol is the main component of $PM_{2.5}$ that increases in urban areas in the BE scenario due to increased SO_2 emissions (Table 4). Urban areas tend to see the largest increases in $PM_{2.5}$ with New York seeing an increase in 98th percent 24-hr average $PM_{2.5}$ from 32.5 to 38.0 μg m^{-3} (Table 3).

In addition to the decreases between the present and future, the CT2 and TE scenarios lead to further decreases in annual average PM in the eastern U.S. of up to 2 μg m^{-3} less than the 2050 reference case (Figure 2). The BE scenario tends to increase PM concentration by up to 2 μg m^{-3} over much of the U.S. during the entire year, especially in the eastern regions while the CT1 scenario shows only small changes over the U.S.

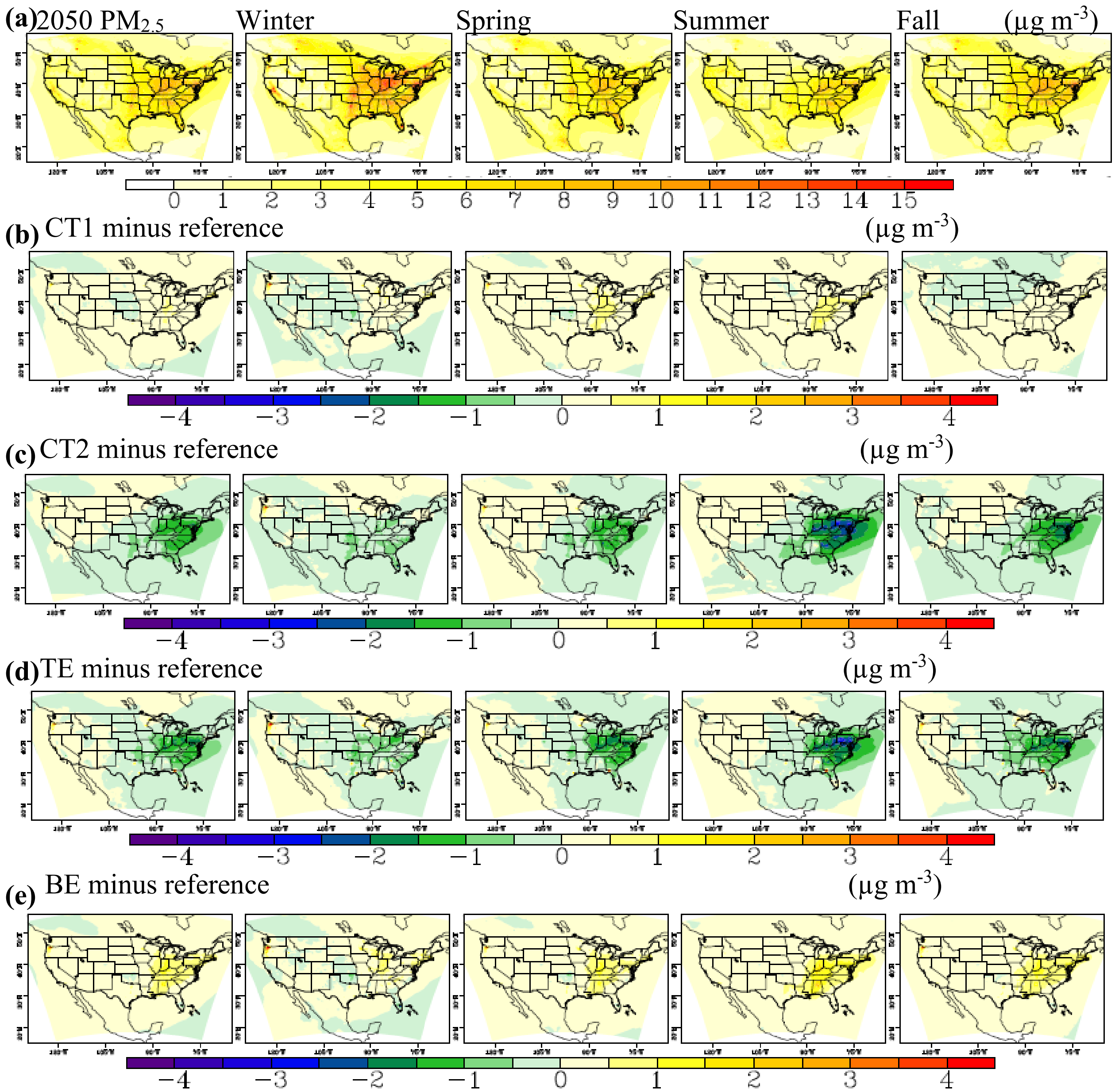


Figure 2. (a) Annual and seasonal average $PM_{2.5}$ concentrations ($\mu g\ m^{-3}$) for the future year (2050) reference case and the change in $PM_{2.5}$ concentration for the (b) low carbon tax scenario (CT1 minus reference), (c) high carbon tax scenario (CT2 minus reference), (d) transportation and energy scenario (TE minus reference) and (e) biomass energy scenario (BE minus reference)

Table 4. Annual average concentrations of sulfate, nitrate and organic matter $PM_{2.5}$ component species in major U.S. cities for the future year reference case and for each alternative emissions scenario

	Mean Sulfate (μg/m^{-3})				
City	**2050**	**CT1**	**CT2**	**TE**	**BE**
Atlanta	2.6	2.7	1.5	1.9	3.0
Chicago	1.8	1.9	1.2	1.5	2.1
Los Angeles	1.4	1.4	1.4	1.9	1.5
New York	2.6	2.7	1.8	2.2	3.0
Philadelphia	2.5	2.5	1.4	1.8	2.9
Phoenix	0.8	0.8	0.7	0.8	0.8
Seattle	0.8	0.8	0.8	0.9	0.9

	Mean Nitrate (μg/m^{-3})				
City	**2050**	**CT1**	**CT2**	**TE**	**BE**
Atlanta	0.6	0.6	0.6	0.4	0.6
Chicago	1.6	1.6	1.7	1.1	1.5
Los Angeles	0.3	0.4	0.3	0.3	0.3
New York	1.0	1.0	1.0	0.9	1.0
Philadelphia	1.1	1.1	1.1	0.9	1.0
Phoenix	0.2	0.2	0.2	0.2	0.2
Seattle	0.3	0.3	0.3	0.3	0.3

	Mean Organic Matter (μg/m^{-3})				
City	**2050**	**CT1**	**CT2**	**TE**	**BE**
Atlanta	2.2	2.5	2.5	3.1	2.6
Chicago	1.5	1.9	1.7	2.3	1.9
Los Angeles	3.0	3.4	3.3	2.6	3.5
New York	2.5	3.3	2.7	4.9	3.4
Philadelphia	1.8	2.1	1.9	2.7	2.2
Phoenix	2.4	2.7	2.1	1.9	2.6
Seattle	2.8	3.3	3.2	2.8	3.5

DISCUSSION

In simulating the effect of CO_2 emission reduction policies on air quality in the U.S., we find two potential policies (CT1 and BE) which can lead to worse air quality, in the form of increased $PM_{2.5}$ concentrations, compared to the 2050 reference case and two policies which lead to improvements compared to the 2050 reference case (CT2 and TE). The implementation of relatively aggressive carbon taxes can lead to improvements in $PM_{2.5}$ air quality compared to the 2050 reference case due to the increased incentives to install FGD process technologies a CCS technologies. However, there is an air quality trade-off because NO_X emissions increase in states not subject to the Clean Air Interstate Rule's NO_X cap and O_3 increases as a result. The relatively less aggressive carbon taxes, on the other hand, leads to worse air quality, in the form of increased $PM_{2.5}$ concentrations because there is less incentive to install FGD and CCS technologies.

The policy aimed at reducing CO_2 from the transportation sector as well as electricity production sectors leads to reduced emissions of mobile source NO_X, thus reducing O_3 levels. Over most of the U.S., this scenario leads to reduced $PM_{2.5}$ concentrations as well. However, increased primary $PM_{2.5}$, OC and EC emissions associated with fuel switching leads to increased annual ambient $PM_{2.5}$ in many major U.S. cities. While the TE scenario is only one realization of emissions, which is subject to the limitations of the models, the results stress the impact of fuel switching in the energy market on air quality and the differences in air quality responses at different spatial scales (i.e. regional vs. urban).

The use of the EPA9r and MARKAL in conjunction with a chemical transport model is shown here to be a useful tool in assessing a range of alternative policy-based emissions scenarios that can be used to provide information to policy makers as well as to address the uncertainties associated with estimating future emissions. As noted in Rudokas et al.[30], the MARKAL database used in the present study was developed using conservative assumptions regarding the potential for increasing end-use energy efficiency to meet carbon emission reduction goals. Inclusion of a wider range of energy efficiency options would especially impact the CT2 scenario, where the model could have chosen efficiency options that would be less expensive than investments in carbon capture and sequestration. An important area of future work is the inclusion of a comprehensive range of energy efficiency options in the MARKAL database. Another future work could include further investigation into the overall health and economic impacts of the emissions scenarios used in the present study.

The results here show that CO_2 emissions reductions strategies will play an important role in impacting air quality over the U.S. The results also show that CO_2 emission reduction policies can have mixed positive and negative impacts on air quality.

ASSOCIATED CONTENT

Supporting Information.

Additional figures referenced in this manuscript are given in the Supporting Information. This material is available free of charge via the Internet at http://pubs.acs.org.

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