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32 ABSTRACT 33 The impact of climate change on surface level ozone is examined through a multi-scale modeling 34 effort that linked global and regional climate models to drive air quality model simulations. 35 Results are quantified in terms of the Relative Response Factor (RRF_E), which estimates the 36 relative change in peak ozone concentration for a given change in pollutant emissions (the 37 subscript E is added to RRF to remind the reader that the RRF is due to emission changes only). 38 A matrix of model simulations was conducted to examine the individual and combined effects of 39 future anthropogenic emissions, biogenic emissions, and climate on the RRF_E. For each member 40 in the matrix of simulations the warmest and coolest summers were modeled for the present-day (1995-2004) and future (2045-2054) decades. A Climate Adjustment Factor (CAF_C or CAF_{CB} 41 42 when biogenic emissions are allowed to change with the future climate) was defined as the ratio 43 of the average daily maximum 8-hr ozone simulated under a future climate to that simulated 44 under the present-day climate, and a climate-adjusted RRF_{EC} was calculated (RRF_{EC} = RRF_E * CAF_C). In general, RRF_{EC} > RRF_E, which suggests additional emission controls will be required 45 46 to achieve the same reduction in ozone than would have been achieved in the absence of climate 47 change. Changes in biogenic emissions generally have a smaller impact on the RRF_F than does future climate change itself. The direction of the biogenic effect appears closely linked to 48 organic-nitrate chemistry and whether ozone formation is limited by volatile organic compounds 49 (VOC) or oxides of nitrogen (NO_X = NO + NO₂). Regions that are generally NO_X-limited show 50 a decrease in ozone and RRF_{EC}, while VOC-limited regions show an increase in ozone and 51 RRF_{EC}. Comparing results to a previous study using different climate assumptions and models 52 53 showed large variability in the CAF_{CB}. 54 55 56 57 58 59 60 61

IMPLICATIONS We present a methodology for adjusting the RRF to account for the influence of climate change on ozone. The findings of this work suggest that in some geographic regions, climate change has the potential to negate decreases in surface ozone concentrations that would otherwise be achieved through ozone mitigation strategies. In regions of high biogenic VOC emissions relative to anthropogenic NO_X emissions, the impact of climate change is somewhat reduced, while the opposite is true in regions of high anthropogenic NO_X emissions relative to biogenic VOC emissions. Further, different future climate realizations are shown to impact ozone in different ways.

94 INTRODUCTION 95 In recent years, the term "climate penalty" has become a commonly used phrase to describe the 96 negative impact that climate change may have on surface ozone concentrations and the 97 subsequently more stringent emissions controls that would be required to meet ozone air quality standards.^{1,2} Despite the many comprehensive modeling studies examining the potential impact 98 of climate change on ozone (e.g., Weaver et al.³ summarize work from a number of studies on 99 the continental United States), this "climate penalty" has not yet been quantified in a way 100 101 meaningful to regulators. 102 103 In the United States, state and local agencies are required to develop State Implementation Plans 104 (SIPs) detailing the policies and control measures that will be implemented to bring ozone non-105 attainment regions into attainment with the National Ambient Air Quality Standard (NAAQS). 106 As part of the SIP process, regulators use chemical transport models (CTMs), such as the Community Multi-scale Air Quality (CMAQ) model⁴ and the Comprehensive Air Quality Model 107 108 with extensions (CAMx; http://www.camx.com/), to demonstrate that proposed control measures 109 will lead to attainment of the ozone NAAOS. 110 111 In the 8-hr ozone SIP, U.S. EPA guidelines call for models to be used in a relative sense, where 112 the ratio of the future to baseline (current) simulated daily maximum 8-hr ozone is calculated 113 instead of the absolute difference between the two simulations. The future and baseline 114 simulations typically use the same meteorology, biogenic emissions, and chemical boundary 115 conditions, and so only differ in the baseline and future control strategy anthropogenic emission inventories. The ratio of the simulated control case to baseline daily maximum 8-hr ozone at any 116 117 monitor is termed a Relative Response Factor (RRF), and represents the model response to a 118 specific change in emissions. The RRF is typically calculated for individual days that meet 119 specific model performance criteria and then these daily RRFs are averaged to obtain an overall 120 monitor-specific average RRF. To estimate the ozone concentration that would be achieved by a 121 given change in anthropogenic emissions, the product of the average RRF and a site-specific 122 Design Value (DV) ozone concentration is calculated (control ozone = average RRF × DV), where the Design Value is representative of observed summertime peak 8-hr ozone. If the future 123 124 control ozone concentration is below the 8-hr ozone NAAQS, then the proposed emission

controls are sufficient to bring the monitor into attainment (see U.S. EPA⁵ for a detailed description of how to calculate the ozone RRF and monitor Design Value concentration).

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Since CTM modeling is such an integral component in demonstrating future attainment of the ozone NAAQS, the potential climate change impact on ozone should be quantified in a way that is useful to regulators; specifically, the impact of climate change should be accounted for in terms of the RRF. The goal of this paper is to quantify results from an on-going multi-scale modeling effort investigating the potential direct and indirect effects of global climate changes on U.S. air quality in a way that is meaningful to regulators. Results are presented in a manner that is consistent with the current use of models in the development of the ozone SIP.

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MODELING

Climate and Meteorology

The Weather Research and Forecasting (WRF) mesoscale meteorological model⁶ (http://www.wrf-model.org) was used to simulate both current (1995-2004) and future (2045-2054) summertime climate conditions. The WRF model is a state-of-the-science mesoscale weather prediction system suitable for a broad spectrum of applications ranging from meters to thousands of kilometers, and has been developed and used extensively for regional climate modeling (e.g., Leung et al.⁷). For this study, WRF was applied with nested 108-km and 36-km horizontal resolution domains, centered over the continental United States, with 31 vertical layers. The 108-km domain was forced with output from the ECHAM5 general circulation model^{8,9} coupled to the Max Planck Institute Ocean Model.¹⁰ For the current decade, ECHAM5 was run with historical forcing through 1999. From 2000-2004 and for the future decade, ECHAM5 was run with the Intergovernmental Panel on Climate Change (IPCC) Special Report on Emissions Scenarios (SRES) A1B scenario. 11 The A1B projection assumes a balanced progress along all resource and technological sectors, resulting in a balanced increase in greenhouse gas concentrations from 2000 to the 2050's. The ECHAM5 driven WRF simulations for the current decade have been shown to represent the ENSO (El Nino –Southern Oscillation) patterns and extreme temperature and precipitation over the Western U.S. reasonably well. 12,13 In addition to the 108-km and 36-km simulations, WRF was also run on a 220-km horizontal resolution semi-hemispheric domain, which encompasses East Asia, the Pacific Ocean, and

North America (the semi-hemispheric WRF simulations were forced by the same ECHAM5 simulations as the 108-km simulations). Results from the 220-km simulations were used to drive semi-hemispheric CTM simulations, which provide chemical boundary conditions for 36-km CTM simulations over the continental U.S. For details on the WRF model setup and model evaluation the reader is referred to Salathé et al.¹⁴

Chemical Transport Modeling and Emissions

The CMAQ model version 4.7¹⁵, with the SAPRC99¹⁶ chemical mechanism and version 5 of the aerosol module, was used to simulate the potential impact of climate change on surface ozone over the continental U.S. CMAQ simulations were conducted on two domains (Figure 1). The first, a 220-km horizontal resolution semi-hemispheric domain, captures the transport of Asian emissions to the U.S. west coast, and provides chemical boundary conditions for the 36-km horizontal resolution continental U.S. (CONUS) domain. Simulations for both domains were conducted with 18 vertical layers from the surface up to 100 mbar, with a nominal depth in the surface layer of ~40 m.

Meteorology for both the hemispheric and CONUS domains is based on the downscaled ECHAM5 simulations, where the future climate is represented by SRES A1B assumptions. The WRF meteorological fields were processed with the Meteorology-Chemistry Interface Processor (MCIP) version 3.4.1. ¹⁷ Chemical boundary conditions (CBCs) for the CONUS domain were provided by the semi-hemispheric CMAQ simulations. For all simulations, biogenic emissions were estimated using the Model of Emissions of Gases and Aerosols from Nature version 2.04 (MEGANv2.04; http://cdp.ucar.edu) ¹⁸ using meteorological output from MCIP and the default MEGANv2 land cover data. Land use and land cover (LULC) were held constant at current decade conditions for all simulations. This version of MEGAN does not account for the impact that rising atmospheric carbon dioxide (CO₂) concentrations have on biogenic isoprene emissions, which will likely lead to an overestimate of the increase in biogenic emissions expected under a warmer future climate. ¹⁹

Anthropogenic emissions of reactive gaseous species for the semi-hemispheric domain were from the POET^{20,21} and EDGAR²² global inventories; organic and black carbon emissions were

from Bond et al.²³ Current anthropogenic emissions for the CONUS domain were based on the 187 U.S. EPA National Emissions Inventory for 2002 (NEI2002; 188 189 http://www.epa.gov/ttnchie1/net/2002inventory.html). These emissions were projected to 2050 using the Emission Scenario Projection version 1.0 (ESP v1.0)²⁴ methodology, which is based on 190 the MARKet ALlocation (MARKAL) model²⁵⁻²⁷ coupled to a database developed by the U.S. 191 EPA, which represents the U.S. energy system at national and regional levels.²⁸ The future-192 decade emissions were based on a business as usual scenario, where current emissions 193 regulations are extended through 2050 ("Scenario 1" in Loughlin et al.21). The business as usual 194 195 scenario includes an approximation of the Clean Air Interstate Rule limits on electric sector 196 sulfur dioxide (SO_2) and oxides of nitrogen ($NO_X = NO + NO_2$) emissions; a requirement that all 197 new coal-fired power plants utilize low-NO_X burners, select catalytic reduction and flue gas 198 desulfurization controls; heavy duty vehicle emission limits on SO₂, NO_x, and particulate matter 199 (PM) emissions; Tier II emission limits and fleet efficiency standards for light duty vehicles; and 200 implementation of the renewable fuel standards targets of the Energy Independence and Security 201 Act of 2007. 202 203 Percent change in modeled anthropogenic and biogenic emissions for the CONUS domain are 204 shown in Figure 3. Emissions are summarized for the regions defined in Figure 2. Under the MARKAL 2050 business as usual scenario, emissions of NO_X and SO₂ are projected to decrease 205 206 in all regions. The decrease in NO_X emissions ranges from 16% in the South to 35% in the 207 Northeast, while the decrease in SO₂ emissions is greatest in the Northwest (35%) and least in 208 the Southwest (16%). Anthropogenic emissions of carbon monoxide (CO), non-methane Volatile Organic Compounds (NMVOCs), ammonia (NH₃), and PM_{2.5} (PM with aerodynamic 209 210 diameter less than 2.5 µm) are projected to increase across all regions. Increases in CO range 211 from 7% in the South to 70% in the Midwest. Emissions of NMVOCs also show the smallest 212 increase in the South (13%), with the largest increase occurring in the Central region (33%). The increase in ammonia emissions is relatively constant across all regions (33-39%), while increases 213 214 in PM_{2.5} emissions range from 2% in the Central region to 22% in the Northwest. Emissions of 215 biogenic VOCs (BVOCs) closely follow the simulated change in temperature (discussed in the 216 Simulated Climate Change section) and show an increase in all regions, except the Northwest, which experiences a slight decrease in BVOC emissions due to a projected decrease in the 217

218	temperature of that region. As mentioned above, the increase in BVOC emissions for most
219	regions is likely overestimated since we did not account for the effects of rising levels of CO2 in
220	the atmosphere. ¹⁹
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222	Simulations
223	Six sets of simulations were conducted to examine the separate and combined effects of
224	projected climate and U.S. anthropogenic emission changes on ozone and the RRF. A summary
225	of the simulations performed for this study is provided in Table 1. Simulation CD_Base
226	represents the base case in which all variables are kept at the present-day conditions. FD_US is
227	the same as CD_Base, except that U.S. anthropogenic emissions are at 2050s levels. A1B_Met
228	is the same as CD_Base, except that future-decade instead of current-decade meteorology is used
229	to drive the CMAQ simulations (future meteorology impacts atmospheric transport and chemical
230	reactions rates, but not biogenic emissions). A1B_M is the same as A1B_Met, except that future
231	meteorology is also used to drive MEGAN to derive future-decade biogenic emissions. The last
232	two sets of simulations involve the combined effects of projected climate and U.S. anthropogenic
233	emissions changes. A1B_US_Met uses future meteorology and U.S. anthropogenic emissions
234	with biogenic emissions held at current-decade levels. A1B_US_M is the same as
235	A1B_US_Met, except that biogenic emissions are based on future-decade meteorology.
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237	Each simulation was conducted for two sets of summer climatology (June, July, August),
238	representing the warmest and coldest summers (based on the mean surface temperature across
239	the U.S.) within the current (1995-2004) and future (2045-2054) decades. All simulations use
240	chemical boundary conditions based on the 220-km semi-hemispheric domain CMAQ
241	simulations with meteorology consistent with the CONUS simulations and using present-day
242	anthropogenic emissions (see MODELING section). Present-day LULC data are applied to all
243	simulations. Wildfire emissions are not included in the simulations due to the uncertainty in
244	predicting future fires. We will address the effect of changes in projected future wildfire
245	emissions on surface ozone and PM in a future paper.
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247	RESULTS AND DISCUSSION
248	Simulated Climate Change

Changes in climate can have both direct and indirect effects on ozone levels. Direct effects include enhanced photochemistry through increases in temperature and insolation, improved ventilation from increases in wind speed and planetary boundary layer (PBL) heights, removal of pollutants from the atmosphere through precipitation, and a reduction in background ozone from increased water vapor content (Jacob and Winner² and references therein). Indirect effects include changes in temperature-sensitive emissions from biogenic sources, as well as climateinduced relocation of those sources through plant species migration. 29,30 Percent change in ozone-relevant meteorological parameters from the 36-km WRF simulations are shown in Figure 4. Results are averaged over the seven regions defined in Figure 2. Changes in meteorological parameters were calculated from averages of the warmest and coldest summers in each decade, which correspond to the summers used in the CMAQ simulations. For temperature and boundary layer height, changes in the average daily maximum are shown; while for water vapor, precipitation, insolation, and wind speed, changes in the average values are shown. On average, the change in temperature (shown as percent °C) tends to increase from west to east across the U.S., with the largest temperature increase occurring in the Northeast (15%) and the only decrease in temperature occurring in the Northwest (1%). The same general west to east trend is also seen with other meteorological parameters. PBL height increases in all regions, with the smallest increase occurring in the Northwest and Southwest (3-4%) and a relatively constant increase in the other regions (10-12%). Insolation decreases slightly in the Northwest (4%), but increases in all other regions, peaking in the northeast at 8%. Water vapor content shows the largest decrease in the Southwest (7%), with only slight decreases in the Northwest and Central regions. All other regions show an increase in water vapor content, with the largest increases occurring in the Northeast (8%) and Southeast (7%). In contrast to the other meteorological parameters, wind speed and precipitation do not show a west to east trend. Changes in wind speed vary from a decrease of 2-4% in the Northwest and Southeast to an increase of 5% in the Southwest and Central regions. Precipitation is predicted to decrease in all regions and ranges from 1% in the Southeast to greater than 50% in the Southwest. The results presented in Figure 4 are generally consistent with published results from other studies simulating a future 2050 A1B climate. For example, Leung and Gustafson³¹ simulated a

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current (1995-2005) and future (2045-2055) A1B climate using the MM5 mesoscale meteorological model³² driven by the Goddard Institute of Space Studies (GISS) global climate model. 33 The work of Leung and Gustafson 31 has been widely used in modeling studies examining the impact of climate change on air quality. 34-36 Although their work is based on the same A1B scenario as the results presented here, differences do arise because of the use of different global and mesoscale models and the choice of current and future years to simulate (e.g., some years may be warmer or colder than others). The most notable differences occur in the Northwest, where Leung and Gustafson³¹ show an increase in both temperature and precipitation, while our work shows a decrease in both parameters. These differences may be attributed to the number of years simulated; our WRF simulation results also show an increase in temperature if ten years of simulations are included in each of the 2000 and 2050 decades (results not shown). Additional differences can be seen from Zhang et al. 35, who use a two-year subset of meteorology from Leung and Gustafson.³¹ The differences seen in Zhang et al.³⁵ include an increase in precipitation in the Northwest, increased wind speed in the Southeast, and a decrease in PBL height in both the Northwest and Southwest, all of which are in contrast to the work presented here. We point out these differences to illustrate that although the work presented here is generally consistent with other similar studies, it does represent only a single future climate realization, and the use of different models, number of years simulated, and assumptions about future emissions will all result in a different climate realization.

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Ozone and Climate

Elevated ozone concentrations in polluted environments are closely linked to temperature.^{37,38} Although the exact mechanism relating temperature and elevated ozone may vary by region, it is likely due to a combination of the following: temperature-dependent chemical rate constants, the relationship between stagnation events and temperature, changes in meteorological parameters associated with elevated temperatures (e.g., insolation and water vapor), and temperature-dependent emissions (e.g., biogenic emissions).

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Figure 5 depicts the observed and modeled relationship between summertime (June, July, August) daily maximum temperature and daily maximum ozone at 72 rural sites within the Clean Air Status and Trends Network (CASTNET; http://www.epa.gov/castnet). Observations are

311 from 1998-2002 and model results are from the two summers representing the warmest and 312 coldest simulated summers from the current decade (CD Base case). Observations beyond 2002 313 are not considered because the large reduction in power plant NO_X emissions in the eastern U.S. 314 that occurred around 2002 is not reflected in the NEI2002 emission inventory. 315 316 In general, the modeled ozone and temperature fall within the range of observed values in each 317 region. However, the modeled results do not show the same day-to-day variability as seen in the 318 observations. This is not unexpected since five years of observations are used compared to two 319 modeled years, and because the model results are averaged over a 36-km grid-cell whereas the 320 observations represent measurements at a single point in space. The average ozone-temperature 321 relationship can be represented by the slope of the linear best-fit. The slopes of the modeled and 322 observed linear best-fit for each region are within approximately +/- 15% of each other, except 323 for in the Central and Southeast regions. These two regions show only minor ozone correlation 324 to temperature, suggesting that temperature is either not the main driver for peak ozone at the 325 CASTNET sites within those regions, or that temperature at these sites is less correlated to other 326 mechanisms that drive elevated ozone, such as stagnation events. The ozone-temperature 327 relationship shown in Figure 5 is generally consistent with the pre-2002 results of Bloomer et al.³⁹, but the slopes of the observed linear best-fit do not match exactly since Bloomer et al.³⁹ 328 329 include additional years (1987-2002) in their analysis, grouped sites in a slightly different 330 manner, and used all hourly data rather than the daily maximum hourly values used in this work. 331 332 Based on the ozone-temperature relationship, under a warmer future climate, ozone would be 333 expected to increase. This relationship generally holds true for the projected change in 334 temperature and ozone between the current- (CD Base) and future-climate (A1B Met) 335 simulations (Figure 6). In regions where temperature is projected to increase under a future 336 climate, ozone is also projected to increase, while in the Northwest, where future temperature is projected to decrease, ozone also decreases. The same trend is seen when biogenic emissions are 337 338 allowed to change with the future climate (A1B M case). 339 340 Although the ozone-temperature relationship is useful for developing a qualitative description of how ozone may change under a future climate, it is not sufficiently robust for use by policy 341

makers when determining the combined effects of both anthropogenic emission reductions and climate change on ozone levels. In particular, observations³⁹ and modeling studies¹ suggest that the penalty associated with climate change decreases when NO_X emissions are reduced. More recent work also suggests that the climate change penalty may be reduced at extreme high temperatures (>39°C), due to a diminishing effect of a reduced PAN (peroxyacetyl nitrate) lifetime on ozone chemistry at these temperatures.⁴⁰

Relative Response Factor (RRF)

Previous modeling studies examining the potential effects of future climate change on ozone in the U.S. typically quantify their results as a change in some peak summertime ozone metric^{34,41-45}, or examine how climate change may affect ozone-relevant meteorological phenomena such as the frequency and duration of stagnation events.^{31,46} Although these types of analyses provide some information to policy makers about how climate change may affect the success of ozone mitigation strategies, they do not address the issue in a way that is consistent with how models are used in regulatory applications. Specifically, they do not address how to account for the impact of climate change on ozone in terms of the RRF (i.e., how to adjust the RRF to reflect the climate penalty).

In other work, Liao et al.³⁶ applied the Decoupled Direct Method 3-D^{47,48} in CMAQ to quantify the sensitivity of ozone and PM_{2.5} to changes in precursor emissions (analogous to the RRF) under a high-extreme and low-extreme future 2050s A1B climate, where the extremes are based on the 0.5th and 99.5th percentiles of temperature and absolute humidity from the MM5 meteorological fields of Leung and Gustafson.³¹ Liao et al.³⁶ found that ozone sensitivity to a reduction in NO_X emissions was generally enhanced under the high-extreme climate case and reduced under the low-extreme case. They attributed the change in model response to changes in temperature-dependent biogenic emissions, which accompany the change in climate (i.e., increases in biogenic VOC emissions due to a warmer climate lead to a more NO_X-limited environment, making NO_X controls more effective at reducing ozone). Although the work by Liao et al.³⁶ provides useful information for how the sensitivity of modeled ozone response to emission reductions may change under a future climate, they do not directly address how to account for the influence of climate change on ozone in the context of the RRF. In the analysis

below, we present results in the context of the RRF and outline a methodology for adjusting the RRF to account for climate change effects on ozone.

For the purpose of this work, we define the non-climate adjusted RRF (RRF_E) below, with the understanding that this is not identical to the rigorous RRF calculation described in the U.S. EPA Attainment Modeling Guidance⁵, and that RRF_E would be replaced by an actual RRF if the following analysis were included in an ozone SIP.

$$RRF_{E} = \frac{1}{N_{exc}} \sum_{t=1}^{N_{exc}} \frac{[O_{3}]_{t, FD_US case}}{[O_{3}]_{t, CD_Base case}}$$
(1)

where $N_{\rm exc}$ is the number of days that exceed the 8-hr ozone NAAQS (75 ppb was used in this work) in the current emissions simulation (CD_Base case), t is the day, and [O₃] is the daily maximum 8-hr ozone for days in which the current emissions simulation (CD_Base case) exceeds the 8-hr ozone NAAQS. The choice of days to include in the RRF calculation is based on the current emissions case only. Since the CD_Base and FD_US simulations use the same meteorology, eq 1 is consistent with how the RRF is applied in SIP analysis. Typically, additional day-specific model performance criteria (such as thresholds for normalized mean error and bias) are applied to the modeled data, and only days that meet these additional criteria are used in the RRF calculation (see U.S. EPA⁵ for details). However, since the meteorology used in this work is constrained by global climate model output and does not represent a specific day or time, performance statistics are not calculated. In the remainder of this paper, the term RRF will refer to a general RRF that may or may not have been adjusted to account for climate change and changes in biogenic emissions. The term RRF_E will refer to the RRF defined in eq 1, which has not been adjusted to account for climate change. Climate adjusted RRFs will be defined in the next section.

Figure 7 shows RRF_E at 1135 ozone monitoring locations throughout the continental U.S.

Modeled ozone was originally analyzed at 1199 sites with continuous monitoring records from
1995-2004 based on data obtained from the U.S. EPA Air Quality System database
(http://www.epa.gov/ttn/airs/airsaqs/); however, 64 of the 1199 sites did not have a single day
where the CD Base case daily maximum 8-hr ozone was greater than 75 ppb, and those sites are

excluded from Figure 7. Values of RRF_E less than one are shown in shades of blue and imply a reduction in ozone due to the projected anthropogenic emission changes shown in Figure 2, while values of RRF_E greater than one are shown in shades of red and imply an increase in ozone. Results are summarized by region in Table 2. Nearly all sites (97%) have an RRF_E less than one, which means ozone is reduced in nearly all locations based on the future 2050s emissions. The remaining 3% of the sites that have an RRF_E > 1 are primarily located in large urban regions with high NO_X emissions that are known to exhibit an ozone disbenefit to NO_X reductions, such that ozone increases with decreasing NO_X emissions. It should be noted that even in these disbenefit regions, if NO_X emissions continue to decrease, at some point there will no longer be a disbenefit and ozone would decrease with a continued reduction in NO_X emissions.

Adjusting the RRF to Account for Climate Change

A key issue facing regulatory agencies is how to account for the potential impact of climate change on ozone within the guidelines of a SIP. One possible methodology is to adjust the RRF to account for climate change effects. This is advantageous because it builds off of the RRF analysis currently called for in the development of the ozone SIP. We do this in terms of a Climate Adjustment Factor (CAF) and define two CAFs as follows:

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$$CAF_{C} = \frac{\frac{1}{N_{all}} \sum_{t=1}^{N_{all}} [O_{3}]_{t, A1B_US_Met case}}{\frac{1}{N_{all}} \sum_{t=1}^{N_{all}} [O_{3}]_{t, FD_US case}}$$
(2)

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$$CAF_{CB} = \frac{\frac{1}{N_{all}} \sum_{t=1}^{N_{all}} [O_3]_{t, A1B_US_M case}}{\frac{1}{N_{all}} \sum_{t=1}^{N_{all}} [O_3]_{t, FD_US case}}$$
(3)

where, $N_{\rm all}$ is the number of simulation days, t is the day, and $[{\rm O_3}]$ is the daily maximum 8-hr ozone. CAF_C only accounts for changes in climate, while CAF_{CB} accounts for changes in both climate and biogenic emissions. Climate adjusted RRFs can then be defined as:

$$RRF_{EC} = RRF_{E} \times CAF_{C}$$
 (4)

433	$RRF_{ECB} = RRF_{E} \times CAF_{CB} $ (5)
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435	where, RRF _E is defined in eq 1, CAF _C is defined in eq 2 and CAF _{CB} is defined in eq 3.
436	Equations 4 and 5 are used to adjust RRF _E in Figure 7 and Table 2. Results for RRF _{ECB} are
437	shown in Figure 8 along with the CAF _{CB} , and summarized in Table 3 for RRF _{EC} and RRF _{ECB} .
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439	In all regions, except the Northwest and Southwest, climate change increases the regional
440	average RRF, the peak RRF, and the spatial variability (represented by the standard deviation) of
441	the RRF (i.e., $RRF_{EC} > RRF_E$). In the Southwest, the peak RRF and the spatial variability of the
442	RRF both increase under future climate conditions (RRF _{EC} > RRF _E), while the average RRF is
443	unchanged (RRF $_{EC} \approx RRF_E$). In the South, Midwest, and Northeast, the increase in the average
444	RRF due to climate change is sufficient to more than offset the decrease in ozone achieved by
445	the change in anthropogenic emissions (i.e., $RRF_E < 1 \le RRF_{EC}$). In other regions, the increase
446	in RRF due to climate change does not completely offset the decrease in ozone achieved by the
447	projected anthropogenic emission changes, but it does reduce the effect those changes have on
448	ozone (i.e., $RRF_E \le RRF_{EC} \le 1$). In all regions but the Northwest, the number of sites having an
449	$RRF_{EC} > 1$ greatly increases under the future climate, with nearly half (45%) of all sites having
450	an RRF $_{EC}$ > 1 (Table 3), compared to only 3% when climate change is not accounted for (RRF $_{E}$;
451	Table 2). The increase in the RRF under the future climate is consistent with other studies that
452	attribute the increase in ozone to enhanced PAN decomposition at higher temperatures and to the
453	association of higher temperatures with stagnation events. ^{2,37,49}
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455	The Northwest, which is predicted in these simulations to cool under the future climate, is the
456	only region that shows a decrease in RRF (RRF $_{EC}$ < RRF $_{E}$). However, this is an artifact of the
457	choice of summers used in this work. As previously stated, the coolest and warmest summers
458	from each decade were chosen based on the mean surface temperature across the continental
459	U.S., which does not necessarily reflect the coolest and warmest summers in the Northwest. An
460	examination of the change in average temperature across all ten years in the current and future
461	decades (not shown) found that on average, the Northwest is expected to experience a slight
462	increase in temperature in the future. Consequently, if all ten summers in each decade were
463	modeled, it is likely that an increase in the RRF would also be seen in the Northwest.

464 465 Although the effect of climate change on the RRF is generally greater than the impact of 466 associated changes in biogenic emissions (Tables 2 and 3), the impact of biogenic emission 467 changes is non-trivial. In the Northwest, the future climate is predicted to cool, resulting in a decrease in biogenic emissions, which has little impact on the RRF (RRF_{EC} ≈ RRF_{ECB}). For all 468 469 other regions, accounting for changes in both climate and biogenic emissions generally results in a minimal increase in the regional average RRF, and a larger, more pronounced increase in the 470 471 regional maximum RRF compared to the climate change only case (RRF_{EC} < RRF_{ECB}). The 472 spatial variability of the RRF (represented by the standard deviation) also increases with 473 enhanced biogenic emissions. In contrast, the number of sites having an RRF > 1 decreases in 474 all regions except the Southwest, Northwest, and Central regions when biogenic emission 475 changes are included. In the Northwest, there is no change because biogenic emissions decrease 476 with decreasing temperature. In the Central region, both biogenic and anthropogenic emissions 477 are relatively low to begin with, so an increase in biogenic emissions does not lead to an increase 478 in the number of sites with an RRF > 1. In the Southwest, the number of sites with an RRF > 1479 nearly doubles when biogenic emissions are allowed to change (from 7% of sites to 13%). The 480 majority of the additional sites in the Southwest with an RRF > 1 are located in Southern California, which is known to be largely VOC-limited^{50,51}, so an increase in biogenic VOC 481 482 emissions results in an increase in ozone production. 483 484 Overall, the change in the RRF to increases in biogenic emissions (RRF_{EC} vs. RRF_{ECB}) appears 485 closely linked to VOC-nitrate chemistry and whether a region is NO_X-limited or VOC-limited. Although the SAPRC99 chemical mechanism used in this work does recycle NO_X from organic 486 487 nitrates (RNO₃), the recycling does not occur instantaneously nor is all of the NO_X recycled. As 488 a result, when biogenic emissions increase, the corresponding increase in peroxy radicals (HO₂ + 489 RO₂) leads to enhanced formation of organic nitrates (RO₂ + NO → RNO₃) and an increase in 490 simulated RNO₃ concentrations. In regions that are generally NO_X-limited (such as much of the 491 Southeast) the enhanced formation of RNO₃ associated with increases in biogenic emissions

reduces the amount of NO_X available to participate in ozone formation, resulting in a decrease in

VOC-limited and exhibit an ozone disbenefit to NO_X reductions, experience an increase in ozone

ozone. In contrast, regions such as Southern California in the Southwest, which are generally

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when biogenic VOCs increase. This is due to a combination of NO_X being removed from the system through enhanced RNO₃ formation and a reduction in the scavenging of ozone by NO $(HO_2 + NO \Rightarrow HO + NO_2)$ becomes the preferred pathway for converting NO to NO_2 over the $O_3 + NO \Rightarrow O_2 + NO_2$ pathway). This is illustrated in Figure 9, which shows average daytime NO_X as a function of average daytime VOC for the A1B_US_Met case. Data points are color coded by the ratio of average daily maximum 8-hr (ADM8-hr) ozone from the A1B_US_M case to the A1B_US_Met. Shades of red imply an increase in ADM8-hr ozone when biogenic emissions are allowed to change with climate and shades of blue represent a decrease in ADM8-hr ozone. In regions with low NO_X and high VOC concentrations ADM8-hr ozone is reduced with future biogenic emissions, while in regions of high NO_X and/or lower VOC concentrations ADM8-hr ozone increases with future biogenic emissions.

It is important to note that we have limited our analysis to sites for which we have calculated an RRF_E (i.e., sites that had at least one day where the CD_Base case simulated a DM8-hr ozone > 75 ppb). However, it is possible that climate change could push sites that are currently in attainment into non-attainment, and any regulatory analysis using the CAF approach should consider this possibility.

Alternate CAF Methodology

Although eq 4 and 5 provide a straightforward methodology for adjusting the RRF to account for potential climate change effects, the application of the CAF in eq 2 and 3 may not be a practical approach for policy makers since the future anthropogenic emission scenario would have to be known prior to the future-climate air quality simulation. Due to the time constraints involved with the development of a SIP, the future-climate air quality simulations would likely need to be completed prior to the future year emission inventory being finalized. Therefore, an alternative approach would be to calculate the CAF using current anthropogenic emissions rather than future emissions (i.e., replacing the A1B_US_Met /A1B_US_M and A1B_US simulations in eq 2 and 3 with the A1B_Met/A1B_M and CD_Base simulations, respectively). This way, the impact of climate change as quantified by the CAF can be estimated independent of future anthropogenic emission scenarios. Figure 10a compares CAF_C and CAF_{CB} calculated using future anthropogenic emissions to those calculated using current anthropogenic emissions. With regard

to ozone formation, the primary difference between the current and future anthropogenic emission inventories is reduced NO_X emissions in the future inventory. For both $CAF_C > 1$ and $CAF_C < 1$, decreasing NO_X emissions reduces the impact of climate change on ozone (CAF becomes closer to 1.0), which is consistent with the findings of Bloomer et al.³⁹ and Wu et al.¹ who found that the penalty associated with climate change is reduced as NO_X emissions decrease. For the future climate and anthropogenic emission scenario, the change in the CAF_C is generally small and using a current anthropogenic emission inventory in the CAF_C calculation gives a reasonable approximation to the future anthropogenic emission CAF_C . However, regulators need to be aware that this may slightly over estimate the climate change impact in terms of the RRF. When changes in biogenic emissions are accounted for in the CAF calculation (CAF_{CB}), the same trends are seen but become slightly more pronounced.

Climate Impacts on the RRF

The CAF approach provides a way to account for the influence of climate change on ozone (i.e., the climate penalty) in terms of the RRF. However, this approach assumes that the RRF is independent of climate change and itself does not change under a future climate. To examine the sensitivity of the RRF to a changing climate, we calculated a new RRF following eq 1, but using results from future climate cases (i.e., replacing the CD_Base case with the A1B_Met or A1B_M cases and the FD_US case with the A1B_US_Met or A1B_US_M cases, respectively) and compared the new RRF to the original RRF from eq 1 (Figure 10b). The majority of sites (90%) had an RRF that changed less than +/- 0.02 when biogenic emissions were held at present-day levels; when biogenic emissions were allowed to change with the future climate, that number dropped to 84% of sites. The largest change in RRF occurred when biogenic emissions were allowed to change (-0.31), but generally the peak changes were between +/- 0.12. The overall bias was less than -0.0006 for both cases; suggesting that while climate change can have a large impact on the RRF at select sites, for the majority of sites the impact is small.

Other Climate Scenarios

In this work, we examined the impact of a single future climate realization on the RRF; however, the use of different climate realizations can lead to very different results. To illustrate this point, we compare results from a previous modeling study conducted by the authors that also examined

the impact of climate change on U.S. air quality. Avise et al. 45 and Chen et al. 52 simulated current (1990-1999) and future (2045-2054) ozone over the continental U.S. for five summers (July only) within each decade. The five July's were chosen to reflect the range of simulated surface temperatures across the continental U.S. within each decade. The most relevant differences between their work and the work presented here is in the future climate assumptions (SRES A2 vs. SRES A1B), global climate model (Parallel Climate Model vs. ECHAM5), and regional meteorological model (MM5 vs. WRF) used to simulate the future climate. Although the SRES A2 and A1B assumptions are different, the two emission scenarios do not begin to diverge significantly until the mid twenty-first century 14, so the difference in future emissions scenario used to drive the global climate models should have a minor impact compared to the differences in the global climate and regional meteorological models used, as well as the specific years simulated. There is also a difference in current emission scenario between the two studies (NEI 1999 vs. NEI 2002), but the inventories are sufficiently similar such that this difference should have a minimal impact compared to the differences mentioned above.

Figure 11 compares the CAF, using current anthropogenic emissions, calculated from the work presented here and a similarly calculated CAF from the work of Avise et al. (CURall and futMETcurLU cases)⁴⁵ and Chen et al. (Cases 1 & 2).⁵² In both studies, biogenic emissions were allowed to change with the future climate and LULC were held constant at current decade conditions. The comparison shows large differences in the CAF calculated from the two studies and these differences occur across all regions. Detailed analysis as to why the differences in CAF occur is beyond the scope of this work. However, we show this comparison as a way of illustrating to the regulatory community the importance of considering multiple future climate realizations in any decision making process.

CONCLUSION

Results from a comprehensive multi-scale modeling study investigating the potential impact of global climate change on summertime ozone in the U.S. were analyzed. The results are presented in a manner that is consistent with how air quality models are used in the development of State Implementation Plans (SIPs). We defined a climate adjustment factor (CAF) as the ratio of the simulated average daily maximum 8-hr ozone from a simulation using future meteorology

to one using current meteorology. The CAF is used to adjust the policy-relevant Relative Response Factor (RRF) to account for the impact that a changing climate may have on the effectiveness of emission control strategies for reducing ozone (i.e., the climate penalty). Although the climate adjusted RRF shows some regional differences, the general trend is towards an increase in the RRF when it is adjusted to account for climate change effects. This trend implies additional emission controls will be required to achieve the same reduction in ozone as would have been achieved in the absence of climate change. Changes in biogenic emissions have less of an impact than climate change itself, and the impact appears closely linked to organic-nitrate chemistry and to whether a region is NO_X- or VOC-limited. In both cases, an increase in BVOC emissions enhances organic-nitrate formation, which removes NO_x from the system. In VOC-limited regions such as Southern California, which exhibit an ozone disbenefit to NO_X emission reductions, removing NO_X from the system results in an increase in ozone. In contrast, in NO_X-limited regions such as much of the Southeast, removing NO_X through enhanced organic-nitrate formation leads to a reduction in ozone. In addition, we compared our results to a previous study 45,52 and found large variability in the CAF, which illustrates the necessity for policy makers to consider multiple future climate realizations to inform their decisions.

Although we have presented our results in a manner that is consistent with how models are used for SIP purposes, there are several differences that should be mentioned. Due to the computational demands required to conduct long-term simulations over the continental U.S., it was necessary to use a 36-km horizontal grid resolution. However, most SIP modeling is done at a higher resolution (4-km or 12-km) and the spatial averaging of the emissions that occurs at coarser resolutions could impact the results. In addition, SIP modeling used in calculating the RRF typically uses some type of reanalysis data to drive the meteorological model, rather than the global climate model output that is required for investigating future climate scenarios and calculating the CAF. However, on average, global climate models compare well with reanalysis fields ^{53,54}, so the disconnect between the meteorology used in the air quality simulations for calculating the RRF and the meteorology used in the simulations for calculating the CAF should not be of critical importance (provided a sufficient number of current and future climate years are simulated). Lastly, the work presented here investigates the impact of a 2050's climate on

the RRF. Since ozone SIPs are concerned with air quality at most out to the late 2020's, the impact of climate change on the RRF is likely to be less than that presented here over a SIP relevant time frame. Recently, the idea of a policy relevant background (PRB) ozone concentration has gained attention⁵⁵; with the thinking that as the ozone NAAQS continue to decrease, it will become increasingly difficult to achieve compliance through local emissions controls alone. In future work, we will use the modeling framework presented here to examine how the PRB ozone concentration may evolve in the future due to changes in the global climate and emissions.

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Table 1. Matrix of 36-km CONUS domain CMAQ simulations. Future meteorology is based on the IPCC A1B scenario. NEI 2002 refers to the US EPA National Emissions Inventory for 2002. MARKAL 2050 refers to a future emissions inventory that is based on the NEI 2002 and projected to 2050 using the US EPA MARKAL allocation model. The same present-day chemical boundary conditions from the semi-hemispheric CMAQ simulations are used for all cases. All simulations use present-day land-use and land-cover data.

simulation name	meteorology	anthropogenic emissions	biogenic emissions	
CD_Base	current	NEI 2002	current meteorology	
FD_US	current	MARKAL 2050	current meteorology	
A1B_Met	future	NEI 2002	current meteorology	
A1B_M	future	NEI 2002	future meteorology	
A1B_US_Met	future	MARKAL 2050	current meteorology	
A1B_US_M	future	MARKAL 2050	future meteorology	

Table 2. Summary of RRF_E by region.

	# sites	RRF_{E}				% of sites
region		average	minimum	maximum	standard deviation	w/ RRF > 1. 00
Northwest	11	0.95	0.92	0.97	0.021	0
Southwest	246	0.91	0.85	1.07	0.043	5
Central	31	0.92	0.86	0.98	0.030	0
South	124	0.95	0.88	1.02	0.040	9
Midwest	287	0.95	0.91	1.00	0.019	0
Southeast	211	0.91	0.84	0.99	0.027	0
Northeast	225	0.92	0.85	1.16	0.048	3
All Regions	1135	0.93	0.84	1.16	0.040	3

Table 3. Summary of climate-adjusted RRFs (eq 4 and 5) by region.

	# sites	RRF_{EC} (RRF_{ECB})				% of sites
region		average	minimum	maximum	standard deviation	w/ RRF >
NI d		0.87	0.84	0.89	0.020	0
Northwest	11	(0.87)	(0.84)	(0.89)	(0.017)	(0)
C 41 4	246	0.91	0.82	1.09	0.059	7
Southwest	246	(0.92)	(0.83)	(1.15)	(0.070)	(13)
Ct1	21	0.97	0.88	1.07	0.066	42
Central	31	(0.98)	(0.87)	(1.10)	(0.074)	(42)
C41-	104	1.04	0.94	1.16	0.057	66
South	124	(1.05)	(0.94)	(1.22)	(0.071)	(65)
M: decease	297	1.04	0.97	1.13	0.033	89
Midwest	287	(1.05)	(0.94)	(1.18)	(0.047)	(88)
G - 4 4	211	0.99	0.85	1.07	0.039	31
Southeast	211	(0.97)	(0.85)	(1.11)	(0.040)	(22)
NT - 11 - 1	225	1.00	0.91	1.25	0.057	32
Northeast		(1.00)	(0.90)	(1.34)	(0.074)	(30)
A 11 D	1135	0.99	0.82	1.25	0.069	45
All Regions		(1.00)	(0.83)	(1.34)	(0.079)	(43)

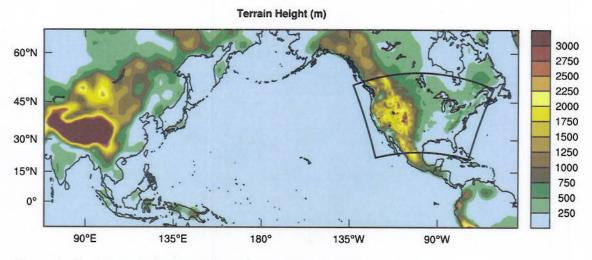


Figure 1. Semi-hemispheric and continental US (CONUS) CMAQ modeling domains.

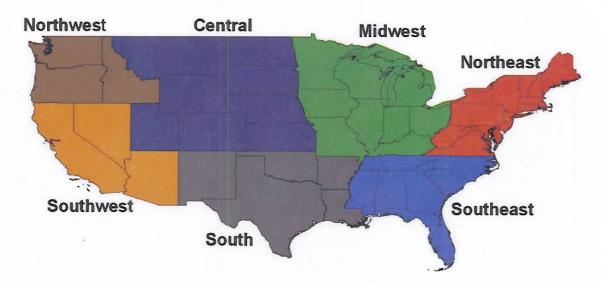


Figure 2. Definition of the regions used in summarizing results.

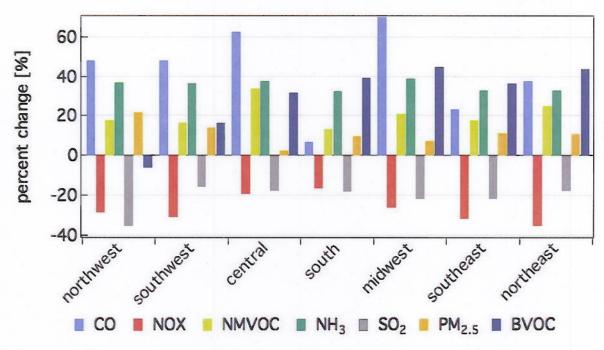


Figure 3. Percent change in continental US emissions from the present-day to the 2050's by region. BVOC represents biogenic VOC emissions that are allowed to change with the future climate.

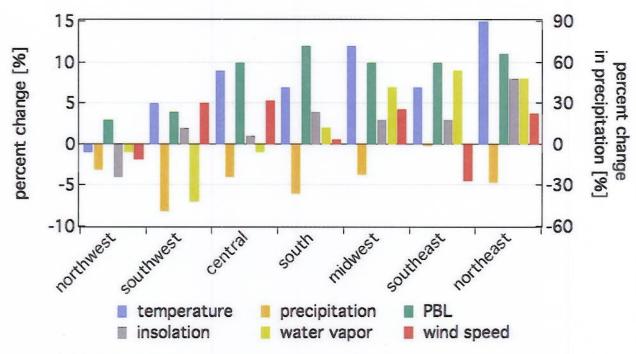


Figure 4. Simulated change in meteorological parameters due to climate change. Percent change in temperature [°C] and PBL are from average daily maximum values, while water vapor, precipitation, insolation, and wind speed are from average values.

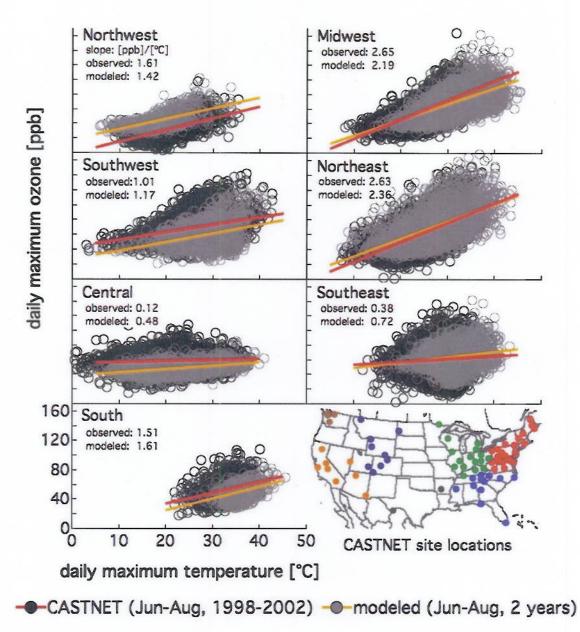


Figure 5. The observed (open dark circles) and modeled (open grey circles) relationship between summertime daily maximum hourly temperature and daily maximum hourly ozone at 72 CASTNET sites. The data have been grouped by site location based on the region definitions in Figure 3, and the site locations have been colored to show which sites correspond to each region. The observed and modeled linear best-fit lines are shown in red and orange, respectively. The slope of the linear best-fit is shown in the upper-left corner of each tile [ppb/°C].

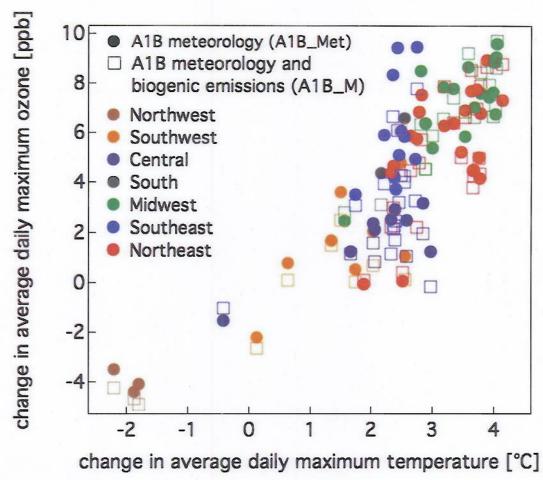


Figure 6. Simulated change in average daily maximum temperature and the corresponding change in average daily maximum 1-hr ozone at the 72 CASTNET sites when biogenic emissions are held constant (A1B_Met; solid circles) and when biogenic emissions are allowed to change in response to the future climate (A1B_M; open squares).

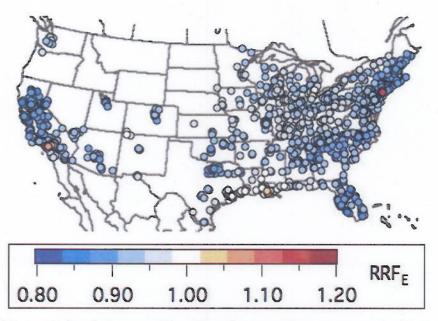


Figure 7. Spatial map of the RRF_E for the 1135 ozone monitoring locations in which the CD_Base case had at least one day where the daily maximum 8-hr ozone exceeded 75 ppb. Values less than one imply a reduction in daily maximum 8-hr ozone, while values greater than one imply an increase in daily maximum 8-hr ozone when anthropogenic emissions are reduced as shown in Figure 2.

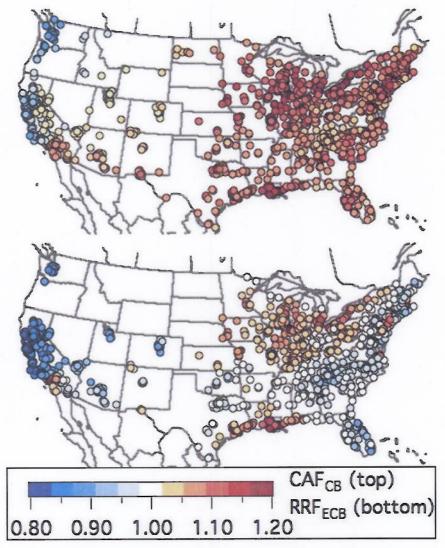


Figure 8. Climate Adjustment Factor (CAF_{CB}) for the A1B_US_M case (top), and the associated climate adjusted RRF (RRF_{ECB}; bottom). A CAF is calculated for all sites, but not all sites have an RRF.

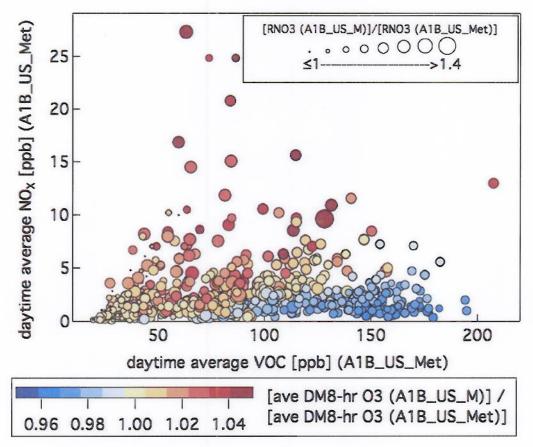


Figure 9. Simulated average daytime NO_X as a function of average daytime VOC at ozone monitor sites for the A1B_US_Met case. Data points are color-coded based on the ratio of the average daily maximum 8-hr O_3 from the A1B_US_M and A1B_US_Met cases. The size of each data point represents the ratio of average daytime organic nitrates (RNO₃) concentration in the A1B_US_M and A1B_US_Met cases.

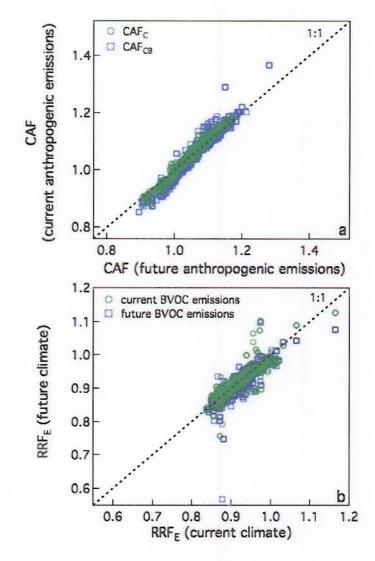


Figure 10. Top (a): comparison between CAF_C and CAF_{CB} (eq 2 and 3), when future anthropogenic emissions are used (A1B_US_M and A1B_US_Met cases), and when current anthropogenic emissions are used (A1B_M and A1B_Met cases). Bottom (b): Comparison between RRF_E from eq 1 when the RRF is calculated under the current climate (CD_Base and FD_US cases) and under the future climate (A1B_Met or A1B_M and A1B_US_Met and A1B_US_M cases, respectively).

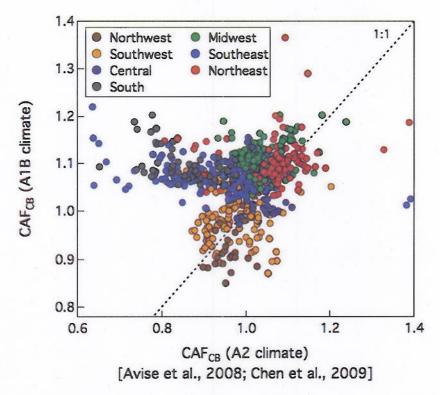


Figure 11. Comparison of the Climate Adjustment Factor (CAF_{CB}) from this study with that calculated from the work of Avise et al. (2008) and Chen et al. (2009), when current anthropogenic emissions are used and biogenic emissions are allowed to change with the future climate (i.e., Alternate CAF Methodology is used).