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Considering future regional air quality impacts of the transportation sector

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ABSTRACT

Regional air pollution is strongly impacted by transportation emissions. Policy mechanisms to reduce emissions are required to reach environmental quality goals. Projecting the drivers (e.g., technical, economic, societal, regulatory) that will impact future emissions is challenging, and assessing regional air quality (AQ) is complicated by the need for detailed modeling tools and data inputs to simulate chemistry and transport of pollutants. This work assesses the contribution of emissions from transportation sources to ground-level concentrations of ozone and fine particulate matter via two methods. First, impacts are quantified for three U.S. regions including California using output from an economic optimization model to grow a base year emissions inventory to 2055. Second, impacts are considered for California using state-level projections with an updated emissions inventory and modeling suite in 2035. For both, advanced AQ models are used, showing that the impacts of light duty vehicles are moderate, reflecting shifts to more efficient and lower emitting technologies. In contrast, heavy duty vehicles, ships, and off-road equipment are associated with important ozone and $PM_{2.5}$ burdens. Emissions from petroleum fuel production and distribution activities also have notable impacts on ozone and $PM_{2.5}$. These transportation sub-sectors should be the focus of future emissions reduction policies.

1. Introduction

Policies to improve regional air quality (AQ) and reduce associated health risks represents a cornerstone of U.S. environmental quality efforts (Bachmann, 2007). Energy sectors, including the transportation sector, are responsible for the bulk of pollutant emissions driving current U.S. AQ concerns, including ground level concentrations of ozone and particulate matter less than 2.5 µm ($PM_{2.5}$) (U.S. EPA, 2005). The transportation sector encompasses the movement of persons or goods by various technology types including light-duty vehicles (LDV), medium-duty vehicles (MDV), heavy-duty vehicles (HDV), rail, ship, aircraft, and other vehicles (e.g., off-road equipment).

With petroleum fuels currently dominant, combustion processes associated with conventional transportation technologies result in significant atmospheric releases of gaseous and particulate pollutants. Emissions of criteria air pollutants from transportation comprise a large fraction of domestic totals, including carbon monoxide (CO), oxides of nitrogen (NO_x), and volatile organic compounds (VOC) (David et al., 2014). Additionally, some transportation sources emit large amounts of sulfur oxides (SO_x) and particulate matter, including $PM_{2.5}$, which carry considerable human health risk (Kleeman et al., 2000; Hasheminassab, 2013). While emissions from transportation directly impact society via induced health effects, materials degradation, aesthetics, etc., further contribution to these burdens occurs via the formation of secondary pollutant species, including ozone and secondary PM. Ozone forms in the troposphere via photochemical interactions between NO_x and VOCs in the presence of sunlight (Finlayson-Pitts and Pitts, 1997) and represents one of the most challenging pollutants to mitigate – many regions of the U.S. currently experience non-attainment for federal criteria pollutant regulatory standards for ozone (U.S. Environmental Protection Agency, 2015). Further, exposure to ozone is known to induce a range of detrimental health outcomes (Moore et al., 2008), while meeting the health-based standards have been shown to provide significant societal benefits (Hubbell et al., 2005). Similarly, $PM_{2.5}$ has been shown to increase a number of serious disease burdens and represents a foremost regional AQ concern (Pope and Dockery, 2006; Laden et al., 2000) that is also often present in concentrations above federal standards in many regions of the U.S. (U.S. Environmental Protection Agency, 2015).

In addition to direct emissions from vehicles, the production of transportation fuels yields pollutant emissions and resulting AQ impacts (Chambers et al., 2008; Rivera et al., 2011). The reliance on petroleum

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fuels requires the existence of an extensive production and distribution system in the U.S., including the regions of study in this work. Petroleum refineries convert crude oil into an assortment of products used in transportation, e.g., gasoline, aviation fuel, distillate fuels, and residual fuels (Speight, 2013). Refining products require a variety of processes (e.g., distillation, reforming, hydrocracking, coking, blending) that result in a diverse range of pollutant emissions including CO, NO_x, PM, SO_x, VOCs, and numerous air toxic compounds, e.g., benzene, toluene (McCoy et al., 2010). Furthermore, emissions associated with the production, storage, transport, and distribution of conventional petroleum fuels are known to contribute to regional AQ problems (Daum et al., 2004; Simons et al., 2015; Mac Kinnon et al., 2016; Smargiassi et al., 2014). Emissions from petrochemical facilities may be underreported and therefore AQ impacts may currently be underestimated (Jobson et al., 2004; Ryerson et al., 2003; Kleinman et al., 2002a). Thus, there is a need for more information regarding the potential effects on regional AQ from petroleum transportation fuels in the framework of policy development.

The U.S. has made significant progress in addressing such concerns through regulatory controls and technological advancements targeting reductions in emissions of gaseous and particulate pollutants (U.S. EPA, 2011b). California (CA) has developed and implemented numerous policies aimed at reducing emissions from various transportation sectors with examples including the Zero Emissions Vehicle (ZEV) Mandate (Collantes and Sperling, 2008), the Goods Movement Emission Reduction Plan (CARB, 2006a) and Sustainable Freight Action Plan (California Sustainable Freight Action Plan, 2016), and the At-Berth Regulation (California Air Resources Board, 2014). Similarly, policy examples at the Federal level include the National Program incorporating both Corporate Average Fuel Economy (CAFE) standards and GHG standards for LDV (Xie and Lin, 2017). Nonetheless, additional targeted and comprehensive pollution reduction policies and regulations for the transportation sector are required as demand increases in response to population and economic growth (Uherek et al., 2010). Transportation sub-sectors differ with regards to distributions and intensities of emissions including purpose, energy conversion technology and fuel characteristics, spatial and temporal patterns of operation, regional demands, etc. Further, future year sub-sector technological evolution and pattern changes of the major emission drivers will not be equivalent and thus some sub-sectors may grow in relative importance to regional AQ while others may lessen. For example, alternative, low-emitting technologies may be easier to develop and apply in the LDV sector than to ship or rail technologies with greater physical constraints, much longer typical service life, and different commercial market considerations.

Additionally, challenges associated with interpreting relationships between transportation source emissions and regional AQ impacts further complicate optimal policy development. The complexity of ozone (Finlayson-Pitts and Pitts, 1999) and PM (Schell et al., 2001) formation in the atmosphere requires detailed emissions dynamics, meteorology and topology information, and atmospheric modeling to simulate chemistry and transport to predict concentrations. The majority of available literature only quantifies emissions of transportation technologies in assessing potential AQ impacts (Wang et al., 2007a, 2007b, 2008; Peterson et al., 2011; Brinkman et al., 2005; Huo et al., 2009; Wang, 2002; Facanha and Horvath, 2007; Chester et al., 2010; Cooney et al., 2013). While previous studies have used atmospheric modeling to examine the AQ impacts of transportation sources (e.g., LDVs (Stephens-Romero et al., 2009; Cook et al., 2010; Brinkman et al., 2010; Thompson et al., 2011; Knipping, 2007b), HDVs (Millstein and Harley, 2010), ships (Vutukuru and Dabdub, 2008); (Song, 2010)), the transportation sub-sectors are considered individually in these efforts, which prevents comparative assessment of sub-sector impacts. Further, studies are generally conducted for one region (Stephens-Romero et al., 2009; Thompson et al., 2011), or at the national (Cook et al., 2010; Jacobson, 2008; Duvall et al., 2007) or global level (Koffi et al., 2010). An important contribution was made comparing five different transportation categories for the impacts of diesel particulate emissions in California (CA), but ozone and secondary PM were not considered (Marshall et al., 2014). The significant resources (e.g., computational, personnel) needed to appropriately model future typically limits the availability of this required information to a small number of organizations involved in planning regional AQ mitigation strategies. Furthermore, there is often a trade-off between technical detail and scope that can prevent full resolution of impacts, e.g., regional-scale with high technical resolution vs. national-scale with less technical detail (Anenberg et al., 2016).

There is a need for more insight into how each sub-sector of transportation contributes to regional AQ challenges, particularly in coming decades as policies are developed for various mitigation strategies. The current work is distinguished by quantifying the contribution to regional ground-level ozone and $PM_{2.5}$ of various transportation sources via modeling platforms allowing for comparison within three different U.S. regions, and a subsequent evaluation for one of the regions with augmented detail. For the first time, the work identifies priority targets for pollutant mitigation strategies that can assist decision makers in formulating policies. Finally, differences in the methods used to project and resolve transportation sector emissions inventories provide insight into methodological considerations for supporting AQ assessment within policy development framework.

2. Materials and methods

To assess future regional AQ impacts of transportation-related sources, emissions must be projected and spatially and temporally resolved to facilitate input into an advanced model of atmospheric chemistry and transport. For this work, two separate methods are used to provide insight into AQ impacts in different regions and to facilitate insights into methodological choices with implications for policy. The goal of these sub-sector spanning scenarios is to provide overall insights into the AQ impacts of the various transportation sub-sectors in future years. To assess the impacts of each transportation sub-sector, scenarios are constructed accounting for the removal of emissions from a given sub-sector (i.e., LDV, HDV, ships) while holding all other sectors and sub-sectors constant with the baseline. This allows the resulting impacts on AQ to quantified and resolved.

The quantity and spatial distribution of future transportation emissions will be driven by socio-economic, regulatory, technological, environmental and regulatory factors (Loughlin et al., 2011). Assessing AQ impacts in future years requires the projection of emission sources economy-wide by consistent methods. Thus, emission projections, whether representing business-as-usual (BAU) or alternative scenarios, should account for these factors to the extent practicable. First, a comprehensive accounting of regional emissions evolution under BAU conditions is needed to provide a Reference Case for comparison with control cases. Next, emissions must be grown to the target year from current levels and spatially and temporally resolved to account for direct perturbations using an emissions processing tool. Finally, a thorough assessment of AQ requires simulation of atmospheric chemistry, e.g., the photochemical formation of ozone, oxidation of VOCs, and formation of organic aerosol precursors.

2.1. U.S. regional method

An overview and comparison of the two methods is presented in Table 1. The first method is used to evaluate different regions of the U.S. (the "U.S. regional method") in 2055 including CA, an aggregate of five Northeastern U.S. states (NEUS), and Texas (TX). Regions were selected due to the presence of existing AQ challenges coupled with significant differences in regional energy demands, utilized technologies and fuels, regulatory constraints, etc.

Baseline AQ is established accounting for BAU continuation of current technological, energy, and economic trends via output from a data-intensive, energy system optimization model, the MARket ALlocation (MARKAL) model (Fishbone and Abilock, 1981; Loulou et al., 2004; Victor et al., 2018; Mahmud and Town, 2016). The approach for the U.S. regional method 2055 Reference Case follows the methodology described by (Loughlin et al., 2011) and used in (Mac Kinnon et al., 2016). Briefly, the

Table 1
Overview of the two methods used to project emissions and evaluate future AQ impacts of transportation sources.

	U.S. regional method	CA method
Base Year Inventory	2005 EPA NEI (U.S. EPA, 2005)	2012 CARB (California Air Resources Board, 2016)
Projection Year	2055	2035
Method	MARKAL Model (Lenox, 2012)	CARB CEPAM Tool (California Air Resources Board, 2016)
Emissions Processing	SMOKE version 3.5.1 (SMOKE v3.6 Users Manual, 2005)	SMOKE version 4.0 (SMOKE v4.0 User's Manual, 2016)
Air Quality Model	CMAQ version 4.7.1 (Foley, 2010)	CMAQ version 5.2 (U.S. EPA, 2017)
Chemical Mechanism	CB 05 (Yarwood et al., 2005)	SAPRC-07 (Carter, 2010)
Transportation Sources	All except aircraft	All including aircraft
Relative Strengths	• Allows for comparison of U.S. via 9 specific regions • MARKAL facilitates comparison under different energy futures	• State-of-the-science model versions and data inputs • Higher technical resolution of emission sources

Table 2
Summary of CMAQ model performance for ozone and $PM_{2.5}$.

U.S. regional method			**California method**				
	Summer			*Summer*		*Winter*	
	MNB	**MNGE**	**O3 (hourly)**	**MNB**	**MNGE**	**MNB**	**MNGE**
O_3 (hourly)	−7.6%	29.3%		−3%	25%	14%	18
	MNB	**MNGE**		**MFB**	**MFE**	**MFB**	**MFE**
$PM_{2.5}$ (24-h)	−2.8%	31.9%	**$PM_{2.5}$ (hourly)**	−16%	57%	−3%	71%

Table 3
Peak baseline concentrations of ozone and $PM_{2.5}$ predicted for the BAU case in each method.

	U.S. regional method (2055)		**CA method (2035)**	
Sub-sector	**8-h Ozone [ppb]**	**24-h $PM_{2.5}$ [μg/m^3]**	**8-h Ozone [ppb]**	**24-h $PM_{2.5}$ [μg/m^3]**
California	90	78	**Summer**	
Texas	65	19	82	67
NEUS	80	29	**Winter**	
			58	55

Table 4
Projected 2055 total daily average emissions of criteria pollutants in study regions for each transportation sub-sector.

	NO_X tons/day	VOC	SO_X	$PM_{2.5}$	NH_3
California					
LDV	103.8	107.3	1.6	24.8	52.3
HDV	285.6	70.9	0.6	4.1	2.6
Off-road	271.8	375.3	0.3	15.0	0.5
Ships	377.8	349.8	4.6	29.2	0.5
Rail	120.8	3.2	0.3	1.2	0.0
Total	**1159.9**	**906.4**	**7.4**	**74.4**	**56.0**
Texas					
LDV	172.8	142.9	5.7	12.7	33.0
HDV	316.8	71.3	2.5	0.2	2.7
Off-road	160.6	398.9	2.9	15.9	0.4
Ships	428.2	315.1	0.9	20.3	0.1
Rail	133.9	3.1	0.4	1.3	0.1
Total	**1212.2**	**931.3**	**12.4**	**50.5**	**36.3**
NEUS					
LDV	230.6	186.3	7.5	18.2	61.9
HDV	267.4	61.1	2.2	0.2	3.1
Off-road	246.8	888.0	3.8	23.8	0.7
Ships	385.0	1200.1	1.8	27.1	0.4
Rail	136.2	3.0	0.4	1.5	0.0
Total	**1265.9**	**2338.6**	**15.7**	**70.8**	**66.2**

MARKAL model is used to produce BAU emission projections from 2005 to 2055. MARKAL is an energy system optimization model that identifies the mix of technologies and fuels that meet energy demands and emission constraints at least cost. MARKAL is applied to the U.S. energy system by pairing it with the U.S. EPA 9-region MARKAL database (Lenox et al., 2012), version EPAUS9R_2010_1.3. Emissions are grown to 2055 from current levels and spatially and temporally resolved using an emissions processing tool, the Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system version 3.5.1 (Coats, 1996; Houyoux and Vukovich,

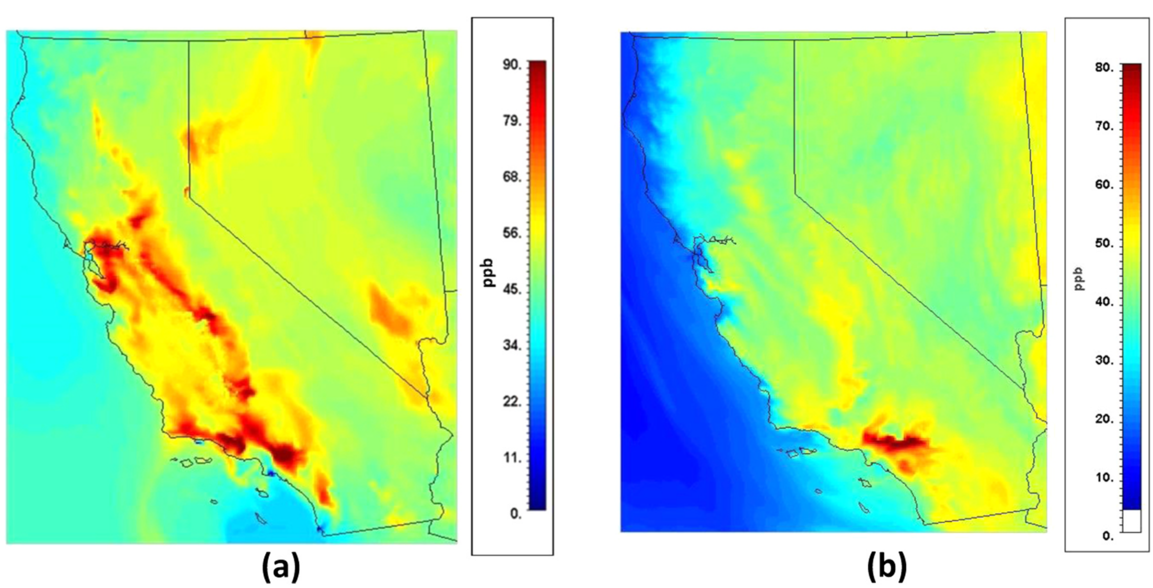

Fig. 1. Predicted peak max 8-h avg. ozone in CA for the a) U.S. regional method in 2055 and b) CA method in 2035.

Table 5
Projected 2035 total daily average emissions of criteria pollutants in CA for each transportation sub-sector.

	NO_X tons/day	VOC	SO_X	$PM_{2.5}$	NH_3
California					
LDV	38.1	89.4	1.9	18.0	22.5
HDV	178.4	36.1	1.8	8.3	5.8
Off-road	110.3	144.2	0.2	6.6	0.3
Ships	65.9	39.9	4.2	2.8	0.1
Rail	38.8	2.4	0.3	0.6	0.1
Aircraft	69.4	34.3	6.2	10.3	0.0
Total	**500.9**	**346.3**	**14.6**	**46.6**	**28.8**

1999). For the U.S. regional method, growth and control factors from MARKAL were applied to the 2005 EPA National Emissions Inventory (U.S. EPA, 2005). It should be noted that MARKAL has evolved into The Integrated MARKAL-EFOM System (TIMES) model. For example, a California specific version of TIMES (CA-TIMES) model is now available and would offer improved output relative to the 9 region MARKAL model, such as used in (Zapata et al., 2018). However, at the time of this work MARKAL was the most current version available from the U.S. EPA. Additionally, the 9 region U.S. MARKAL model is still used within the research scope used in this work, e.g., a 2018 study of decarbonizing the U.S. power sector published in 2018 (Victor et al., 2018).

For the U.S. regional method, AQ simulations were conducted using the Community Multi-scale Air Quality model (CMAQ) version 4.7.1 (Foley et al., 2010) with the CB05 chemical mechanism (Yarwood et al., 2005). CMAQ is a widely used comprehensive AQ modeling system developed by the US EPA and utilized for a range of purposes, e.g., modeling of tropospheric ozone, particulate matter, acid deposition, and visibility for regulatory applications (Byun and Schere, 2006). For the U.S. regional simulations, the model grid resolution is 4 km × 4 km for CA and 12 km × 12 km for TX and the NEUS. The simulations for the NEUS and TX were performed using a one-way nested grid modeling system that includes the eastern part of the United States, and a smaller subdomain. For both methods, meteorological input data was obtained from the Advanced Research Weather Research and Forecasting Model (WRF-ARW). For the U.S. regional simulations, the National Centers for Environmental Prediction (NCEP) Final Operational Global Analysis 1° × 1° grid data were used for WRF-ARW initial and boundary conditions. For the U.S. regional method, simulations are conducted for the week of July 7–13. The first six days of simulations are used to dissipate the effects of the initial conditions (Carreras-Sospedra, 2006). Results are obtained from the seventh day of simulation (July 13) and reported as maximum 8-h average (8-h) ozone and maximum 24-h average (24-hr) $PM_{2.5}$.

2.2. California Specific Method

The second method is used to evaluate CA with enhanced technical resolution within the emissions inventory and updated and improved modeling and data inputs (the "CA method") (SMOKE v4.0 User's Manual, 2016). Baseline AQ is established in the year 2035 by projecting emission changes associated with expected technological, energy, and economic trends via the California Air Resources Board's (CARB) CEPAM: 2016 SIP - Standard Emission Tool (California Air Resources Board, 2016). CEPAM projections are based off CARB aggregate methods for projecting emissions from on-road, area, and point sources and do not represent the output of a single model or method. Rather, the emission projections represent expected and needed reductions in pollutant emissions under the CA State Implementation Plan. The CA method utilized SMOKE version 4.0, an updated version of SMOKE which has been designed to better incorporate technology-specific coding utilizing in CARB emission accounting methods (SMOKE v4.0 User's Manual, 2016). The CA method factors from CEPAM were applied to the 2012 CARB emissions inventory (CARB, 2017).

Table 7
2055 peak reductions in maximum 8-hr average ozone and 24-hr average $PM_{2.5}$ observed in study regions for transportation sub-sector emissions removal.

	California		Texas		NEUS	
Sub-sector	**Δ 8-h Ozone [ppb]**	**Δ 24-h $PM_{2.5}$ [μg/m³]**	**Δ 8-h Ozone [ppb]**	**Δ 24-h $PM_{2.5}$ [μg/m³]**	**Δ 8-h Ozone [ppb]**	**Δ 24-h $PM_{2.5}$ [μg/m³]**
LDV	−2.79	−2.98	−1.20	−0.23	−2.24	−0.40
HDV	−9.28	−1.90	−2.42	−0.46	−2.59	−0.45
Off-road	−8.42	−2.34	−1.78	−0.68	−6.10	−1.26
Ships	−15.26	−32.44	−3.24	−4.59	−4.47	−1.50
Rail	−3.43	−0.633	−0.50	−0.07	−1.35	−0.33
PFI	−1.55	−14.49	−4.59	−3.71	−0.36	−1.84

For the CA method an updated modeling suite is utilized including CMAQ version 5.2 (CMAQ v5.2 (Pye, 2017)) at 4 km × 4 km resolution with the SAPRC-07 chemical mechanism (Carter, 2010). Biogenic emissions are developed using the Model of Emissions of Gases and Aerosols from Nature, version 2.1 (MEGAN v2.1) (Guenther, A., et al., 2012) and boundary conditions are generated using the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). Two simulation periods are conducted to capture the effect of seasonal variation in meteorology and emissions on ozone and $PM_{2.5}$ concentrations including a summer episode (July 8–21) and winter episode (January 1–14). July is selected for both methods as this period encompasses conditions typically associated with high tropospheric ozone formation, including high temperatures, an abundance of sunlight, lack of natural scavengers, and the presence of inversion layers (Carreras-Sospedra, 2006). The January period is associated with high levels of $PM_{2.5}$ in some regions of CA, including the Central Valley (Ying and Kleeman, 2009). Ground-level concentrations are obtained from the final day of simulation (July 21 and January 14) and used to determine baseline and changes in maximum 8-hr values for ozone and 24-h values for $PM_{2.5}$.

The CA method represents an improved method technically relative to the U.S. regional method, however many aspects cannot be applied to other regions of the U.S. For example, the CARB projected emissions are only available for CA. Therefore, while not directly comparable the general trends can be evaluated in terms of sub-sector impacts. Thus, a comparison between the two methods can provide insights into how regulators may interpret the results of AQ assessments generated from different methods or different years.

Table 6
Relative contribution of emissions for each subsector in CA the U.S. regional (NEI) and CA specific (ARB) methods.

	NO_X [% of total]		**VOC [% of total]**		**SO_X [% of total]**		**$PM_{2.5}$ [% of total]**		**NH_3 [% of total]**	
	NEI	*ARB*	*NEI*	*ARB*	*NEI*	*ARB*	*NEI*	*ARB*	*NEI*	*ARB*
LDV	8.9	8.8	11.8	28.7	21.6	22.6	33.3	49.6	93.4	78.1
HDV	24.6	41.3	7.8	11.6	8.1	21.4	5.5	22.9	4.6	20.1
Off-road	23.4	25.6	41.4	46.2	4.1	2.4	20.2	18.2	0.9	1.0
Ships	32.6	15.3	38.6	12.8	62.2	50.0	39.2	7.7	0.9	0.3
Rail	10.4	9.0	0.4	0.8	4.1	3.6	1.6	1.7	0.0	0.3

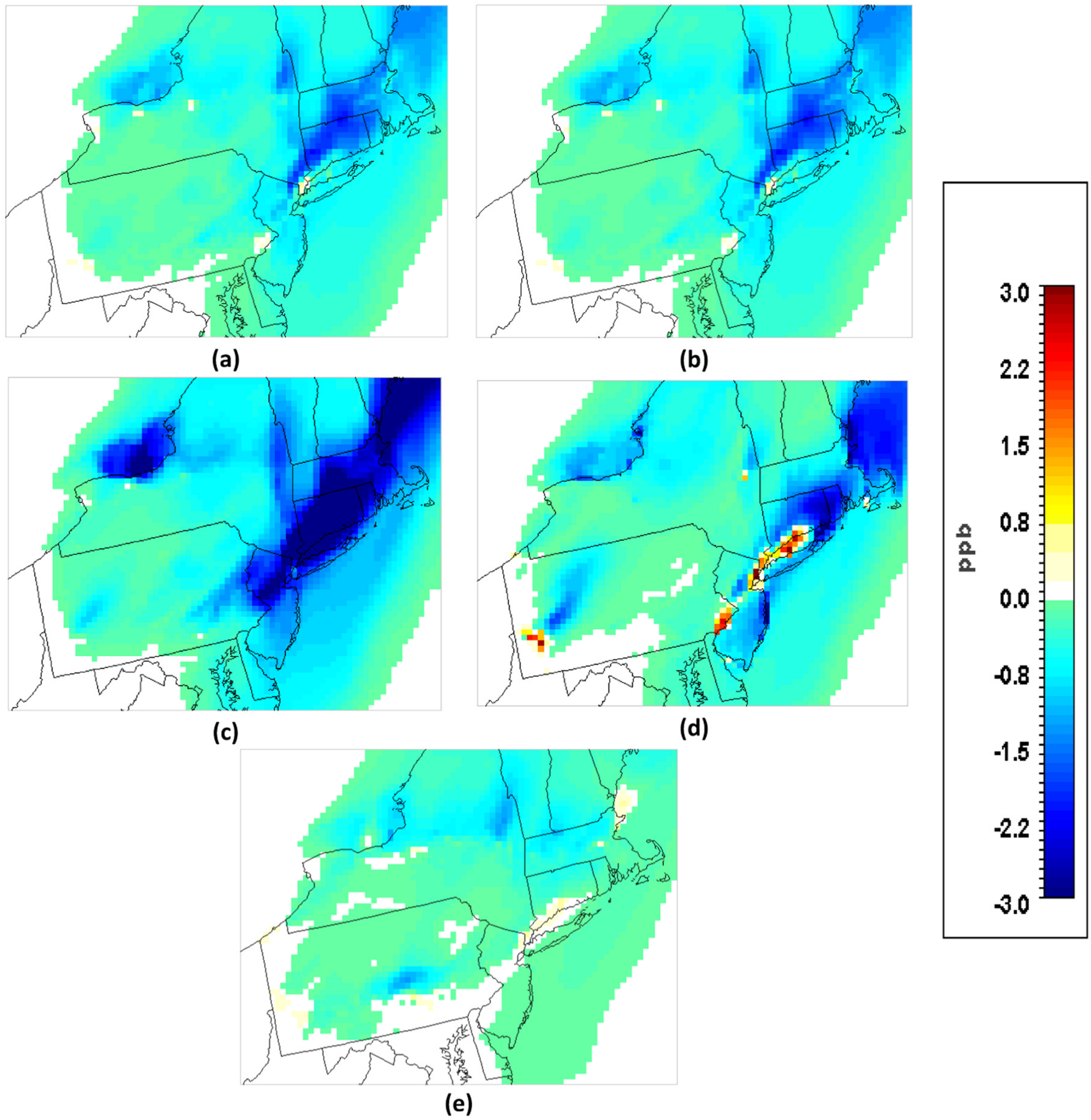

Fig. 2. Predicted differences in peak 8-hr average ozone in NEUS between the baseline and scenarios involving the removal of emissions from (a) LDV, (b) HDV, (c) Off-road, (d) Ships, and (e) Rail in 2055.

2.3. Model performance and reference case

Model performance evaluation was conducted for both the U.S. and CA methods using measurement data from the CARB AQ monitoring network and U.S. EPA Air Quality System. For the U.S. regional method, hourly measurements for ozone and daily average for $PM_{2.5}$ were used to calculate Mean Normalized Bias (MNB) and Mean Normalized Gross Error (MNGE), recommended for model evaluation (U.S. EPA, 2007). Additionally, for the CA method the mean fractional bias (MFB) and mean fractional error (MFE) is applied for hourly $PM_{2.5}$ evaluation as suggested in Reference (Boylan and Russell, 2006). Model performance for both methods is within acceptable tolerance levels (Table 2).

Metrics associated with the prediction of ozone and $PM_{2.5}$ concentrations for the Reference Case for each method are presented in Table 3. The U.S. regional method predicts peak concentrations of 80 ppb and 29 $\mu g/m^3$ for the NEUS and 65 ppb and 19 $\mu g/m^3$ for the TX domain for a summer episode. For CA, the U.S. regional method predicts a peak max 8-hr ozone value of 90 ppb and peak 24-h $PM_{2.5}$ value of 78 $\mu g/m^3$. Conversely, the CA method predicts peak levels of 82 ppb and 67 $\mu g/m^3$ for the summer episode, and 58 ppb and 55 $\mu g/m^3$ for the winter episode. Comparing the values from the summer episodes for CA, the U.S. regional method predicts higher values for the BAU Case as a consequence of higher assumed emissions within the NEI relative to the more restrictive CARB. Predicted concentrations in CA also experience spatial variations due to differences in emission processing, meteorology, emission signatures, etc. Shown in Fig. 1, peak ozone values for both methods occur in the South Coast Air Basin (SoCAB) containing all or portions of the highly populated counties of Los Angeles, Orange, San Bernardino, and Riverside. Additionally, the U.S. regional method predicts high ozone in other regions that do not appear using the CA method, including the Central Valley and San Francisco Bay Area.

3. Results

The following sections presents results for the transportation sub-sectors grouped into LDV, HDV, off-road equipment, ships, rail, and aircraft. Typically, the LDV classification designates passenger cars, light-duty trucks, and motorcycles; while MDV and HDV incorporate commercial technologies (e.g., tractor trailers, school and transit buses) designated by gross vehicle weight. While LDVs are primarily utilized for personal transport, MDVs and HDVs span a range of uses with construction, agriculture,

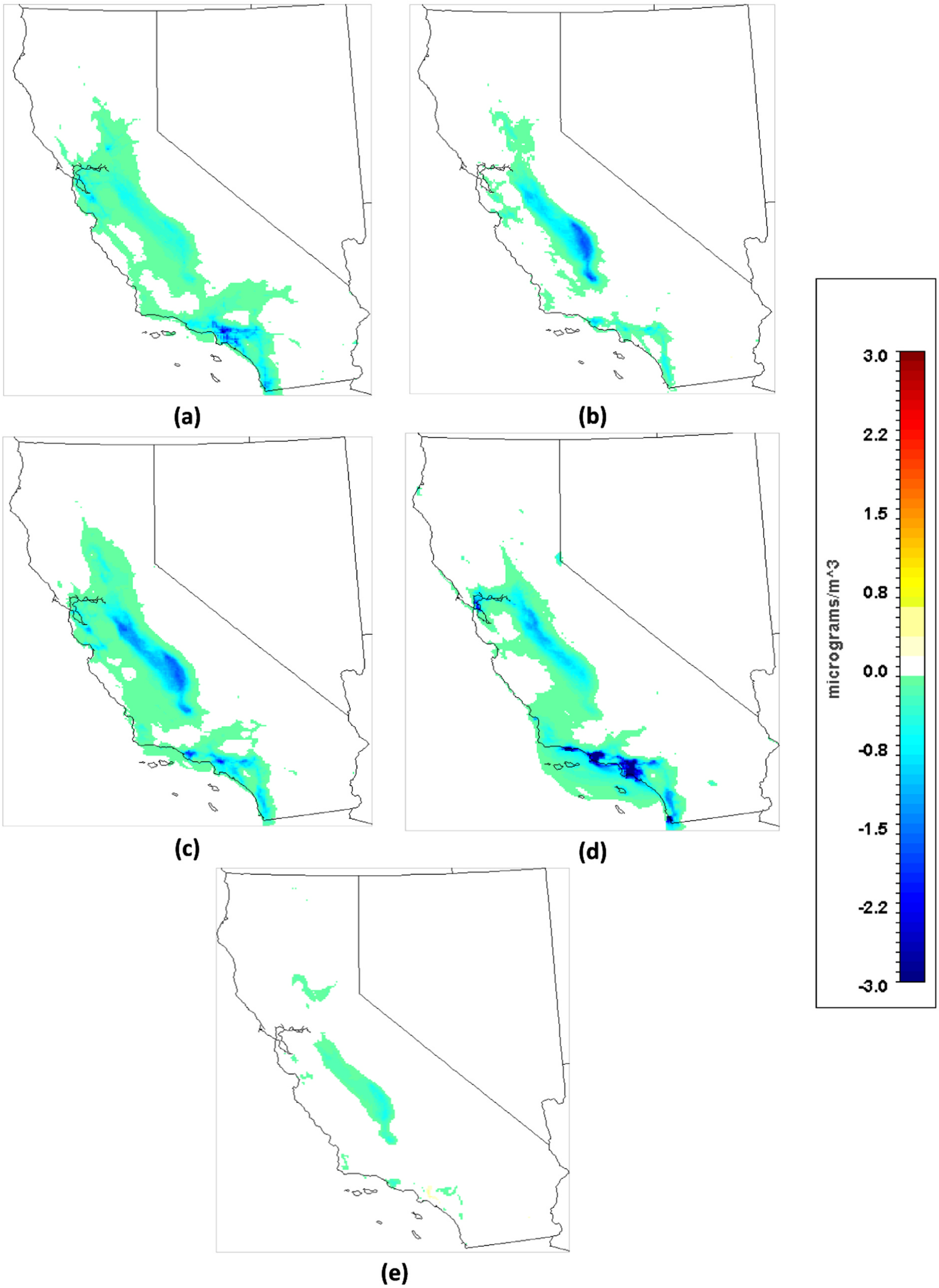

Fig. 3. Predicted differences in peak 24-hr average $PM_{2.5}$ in CA between the baseline and scenarios involving the removal of emissions from (a) LDV, (b) HDV, (c) Off-road, (d) Ships, and (e) Rail in 2055.

and transport of freight foremost (David et al., 2014). Off-road vehicles include a large and diverse spectrum of technologies including agricultural equipment (e.g., tractors, mowers, combines), airport ground equipment, construction and mining equipment (e.g., pavers, backhoes, drill rigs), industrial equipment (e.g., forklifts, terminal tractors), logging equipment, railroad maintenance vehicles, and recreational equipment (e.g., off-road

Table 8
2035 peak reductions in maximum 8-hr average ozone and 24-hr average $PM_{2.5}$ observed in CA for transportation sub-sector emissions removal.

Sub-sector	Summer episode		Winter episode	
	Δ 8-h Ozone [ppb]	Δ 24-h $PM_{2.5}$ [μg/m^3]	Δ 8-h Ozone [ppb]	Δ 24-h $PM_{2.5}$ [μg/m^3]
LDV	−2.5	−2.7	−0.4 to +0.8	−4.1
HDV	−7.2	−1.5	−2.5 to +4.0	−8.1
Off-road	−6.0	−1.1	−1.3 to +2.5	−4.9
Ships	−5.3	−4.0	−0.2 to +25.8	−3.4
Rail	−2.6	−0.5	−1.4 to +2.6	−1.6
Aircraft	−3.9	−6.0	−1.1 to +16.9	−25.1
PFI	−0.7	−2.6	−2.18 to +3.17	−1.9

motorcycles, all-terrain vehicles, golf carts). Rail transport includes both passenger and freight trains. Ships include a wide range of vessels including large ships used to transport goods or persons (e.g., container ships, tankers and cruise ships) and various other craft that range in size and purpose (e.g., tugboats, fishing vessels). Additionally, the sector includes air travel for the movement of both persons and goods associated with commercial and private aircraft.

3.1. Transportation sub-sector emissions

For the U.S. regional method, projected emissions for 2055 from each transportation sub-sector are presented in Table 4. Ships are the largest source of NO_X emissions followed by HDV. Off-road sources are the largest contributor to VOC emissions in CA and Texas, whereas in NEUS the largest contributor originates from ships. Aircraft emissions are not shown as modeling challenges prevented manipulation of aircraft emissions within SMOKE 3.5.1, but these were included for the CA method using SMOKE 4.0. These overall magnitudes help understand the potential AQ impacts of the different sectors. However, the spatial distribution of the various sub-sectors is substantially different, with implications for atmospheric pollutant formation and fate e.g., ship emissions are mostly concentrated around ports, whereas light-duty vehicles are widely spread throughout the domains, with high concentrations within populated areas. Therefore, AQ modeling is required to ultimately evaluate the contribution of each subsector to the AQ of each region.

For the CA method projected emissions for 2035 from each sub-sector are presented in Table 5. Showing the diversity of the sector, HDV are responsible for the largest share of NO_x, off-road vehicles responsible for the largest share of VOC, ships are responsible for the largest share of SO_x, and LDV are the largest emitter of $PM_{2.5}$ and NH_3. With implications for ozone formation off-road sources emit high amounts of both NO_x (second only to HDV) and VOC. Aircraft emissions represent an important source of NO_x and VOC in 2035. It is noteworthy that projected CA 2035 emissions are significantly lower than U.S. regional emissions in 2055. This is expected since CA CEPAM projections consider future strict policies to meet CA State Implementation Plan requirements, while U.S. NEI/MARKAL projections consider only near- to mid-term federal policies.

However, the methods show good agreement regarding the relative share of emissions for each sub-sector as shown in Table 6. For NO_x both methods predict very similar contributions from LDV, off-road, and rail. In particular, the off-road and rail sectors compare well in terms of total contribution between the methods. Similarly, emissions of NH_3 correlate well across all sources. Conversely, deviations are noticeable for HDV and ships, with the CA method predicting higher fractions from HDV and the U.S. regional method showing higher fractions from ships. For ships, emissions from the U.S. method are significantly higher for NO_x, VOC, $PM_{2.5}$ and NH_3, largely due to no assumed implementation of policy measures. In contrast, the CA method assumes the implementation of state policy measures including the At-Berth Regulation to reduce emissions from diesel auxiliary engines on ships at California Ports (CARB, https://www.arb.ca.gov/ports/shorepower/shorepower.htm), and the California Ocean Going Vessel Clean Fuel Regulation requiring vessels within 24 nautical miles of the California coastline to use cleaner distillate fuels in main engines, auxiliary engines, and auxiliary boilers (CARB, https://www.arb.ca.gov/ports/marinevess/ogv.htm). It should also be noted that International Marine Organization (IMO) regulations will further reduce emissions from ships, although they are not accounted for in either method.

3.2. Transportation sub-sector AQ impacts

For both methods, the ozone and $PM_{2.5}$ impacts of removing emissions from transportation sub-sectors is quantified by determining differences in ground-level concentrations between the baseline and control scenario. The results are displayed as maximum 8-h average ozone and 24-h $PM_{2.5}$ difference plots with the specified scenario minus the baseline.

3.2.1. U.S. regional method

Table 7 presents the peak reductions for the U.S. regional method in maximum 8-hr ozone and 24-h $PM_{2.5}$ that were observed in the regions for the transportation sub-sectors investigated. Note these values correspond to the change in ground-level concentrations associated with the spatial location that demonstrated the maximum value of 8-h ozone and 24-h $PM_{2.5}$. While these changes in concentration are related to and important for compliance with federal ambient AQ standards, and while they also indicate the relative importance of each sub-sector, they should be considered together with the overall spatial and temporal impacts of each case to assess impacts completely. Nonetheless, it appears that ships have the most significant impact on both maximum 8-h ozone and 24-h $PM_{2.5}$ differences in CA and 24-h $PM_{2.5}$ differences in Texas. Refineries have the most significant impact on maximum 24-hr $PM_{2.5}$ differences in NEUS and on maximum 8-h ozone differences in Texas, while off-road contributes the maximum 8-hr ozone difference in NEUS.

3.2.1.1. Northeastern U.S. Fig. 2 displays the resulting changes in 8-hr ozone occurring from the removal of the sub-sector emissions in 2055 in the NEUS. Removing emissions associated with off-road sources has high impact on ozone and $PM_{2.5}$ in 2055. HDV and LDV emissions have comparable impacts for both of the studied pollutants with areas of peak impact downwind of NYC. Removing ship emissions results in areas of significant improvement along the New Jersey and New York coastlines from ports associated with the region. Additionally, shipping activity along inland waterways yields AQ improvements extending from the Port of Pittsburgh and along the coast of Lake Ontario. Impacts of rail sources are most notable in northern New York State and central Pennsylvania, coinciding with major railway infrastructure, achieving moderate impacts on ozone and $PM_{2.5}$ relative to other transportation sources.

3.2.1.2. California. Fig. 3 displays the resulting changes in 24-hr $PM_{2.5}$ and Fig. A1 shows the impacts on 8-hr ozone occurring from the removal of sub-sector emissions in CA in 2055. Removal of LDV emissions results in changes that peak at −2.8 ppb and −3 μg/m^3 $PM_{2.5}$. Reflecting the distribution of vehicles, reductions occur over much of the state and are most pronounced downwind of urban regions, notably the SoCAB. HDV emissions have a higher impact of −9 ppb and −1.90 μg/m^3. However, while ozone impacts similar to LDV in terms of region; peak $PM_{2.5}$ impacts occur in the Central Valley. Similarly, off-road emissions have important effects on both ozone and $PM_{2.5}$ over large regions of the State including the SoCAB, the Bay Area, Central Valley, Sacramento and San Diego. The removal of ship emissions results in substantial reductions in both ground-level ozone and $PM_{2.5}$ exceeding 15 ppb and 32 μg/m^3. Areas of improvement are notable in the SoCAB and result from the heavy ship traffic at the Ports of L.A. and Long Beach (POLA and POLB). $PM_{2.5}$ impacts are large when viewed in the context of the Federal NAAQs for 24-h $PM_{2.5}$ of 35 μg/m^3. However, baseline $PM_{2.5}$ levels exceed 78 μg/m^3, with the peak value occurring adjacent to the POLB and POLA area with significant emissions from goods movement and industry activity (shown in Fig. A2).

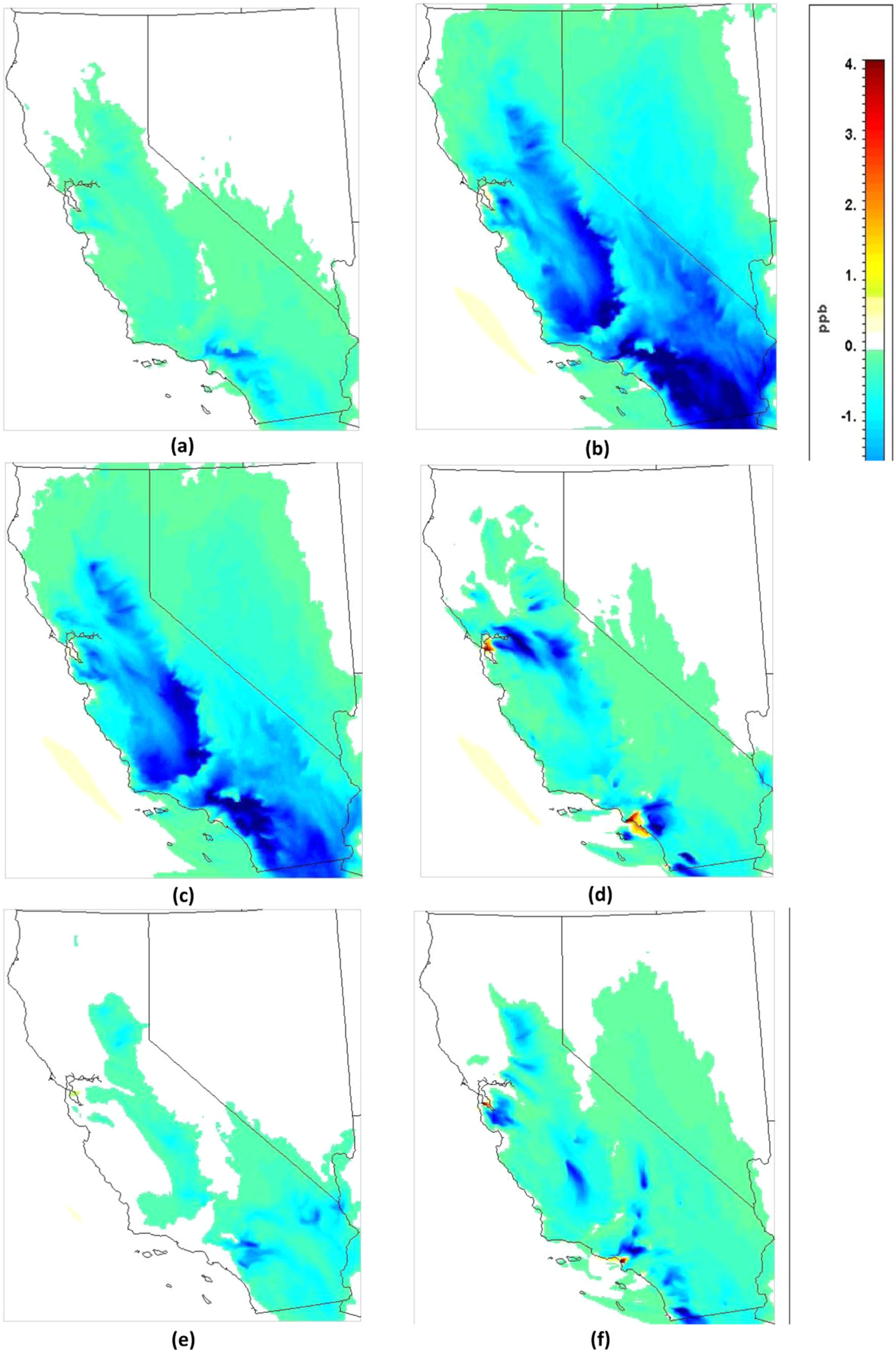

Fig. 4. Predicted differences in peak 8-hr average ozone for a summer episode in CA between the baseline and scenarios involving the removal of emissions from (a) LDV, (b) HDV, (c) Off-road, (d) Ships, (e) Rail, and (f) aircraft in 2035.

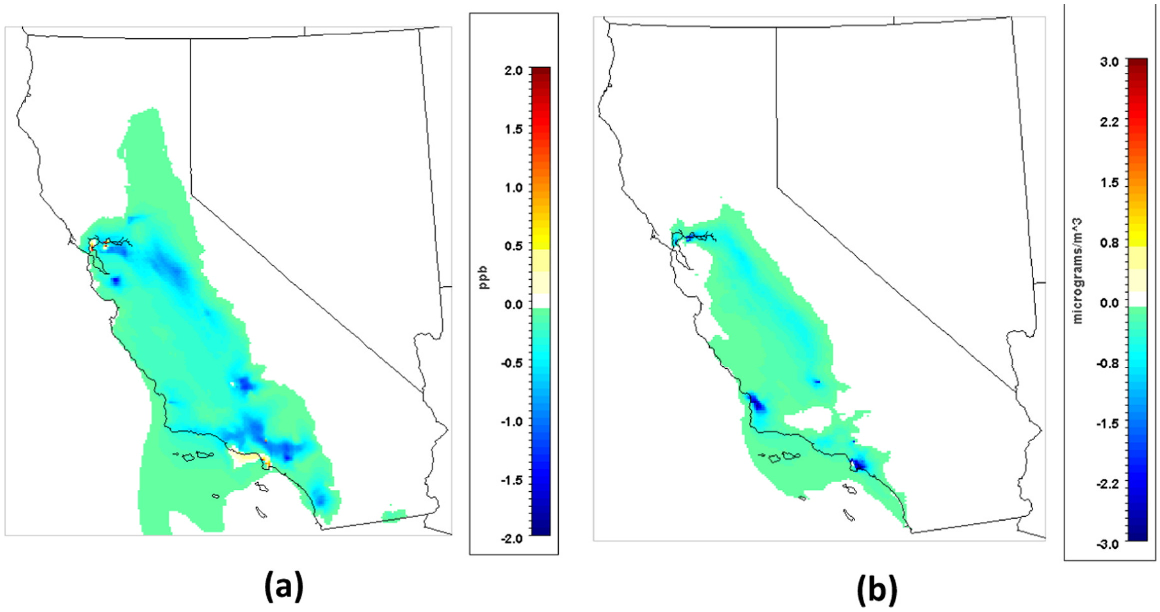

Fig. 5. Predicted differences in a) max 8-h avg. ozone and b) 24-hr avg. $PM_{2.5}$ for the PFI Case from the Base Case.

Thus, a reduction of 32 $\mu g/m^3$ is reasonable and corresponds to a reduction of approximately 59%. It should also be noted that the majority of the domain experiences background levels ranging from 1 to 30 $\mu g/m^3$. Removing rail emissions achieves moderate reductions in ozone and $PM_{2.5}$ in 2055 in rail corridors relative to other surveyed sources.

3.2.1.3. Texas. Shown in Figs. A3 and A4, removing LDV emissions in TX achieves a minor impact on ozone and $PM_{2.5}$ relative to the other transportation sub-sectors investigated, with impacts occurring primarily downwind of metropolitan areas. HDV emissions have a moderately larger impact than LDVs and off-road sources for ozone concentrations, particularly downwind of greater Dallas-Ft. Worth. Off-road sources have a slightly reduced impact on ozone relative to HDVs but a higher impact on 24-hr $PM_{2.5}$ concentrations. Similar to the other regions of study, ship emissions have prominent impacts on ambient AQ in 2055. Removal of ship-related emissions has the highest peak improvements in ozone and $PM_{2.5}$ attributable to any single transportation sub-sector. Reductions are evident starting at sites of major port locations along the Gulf Coast and extending throughout the region. Peak impacts on $PM_{2.5}$ from ship emissions are particularly notable as being an order of magnitude higher than any other transportation sub-sector. Similarly to other regions, rail-related emissions have lower impacts on AQ than other sources.

3.2.2. California Specific Method

Table 8 presents the peak reductions for the CA method in maximum 8-h ozone and 24-h $PM_{2.5}$ that were observed for the transportation sub-sectors investigated in 2035. Fig. 4 show the predicted differences in ozone for the summer episode. Eliminating HDV emissions achieves the largest peak improvement, followed by off-road sources, ships, aircraft, rail and LDV. In terms of spatial area, HDV and off-road emissions achieve the largest impact, while ships and aircraft have more localized impacts. All cases assessed demonstrated peak impacts on ozone in the SoCAB in the summer episode, which emphasizes the regional importance of source impacts, i.e., despite having the lowest impact State-wide, emission reductions from LDV and rail have enhanced importance in SoCAB. Impacts on $PM_{2.5}$ in summer (Fig. A5) differ from ozone trends in that aircraft are associated with the largest improvement magnitude, but are highly localized. Ships have the next highest peak impact. In contrast to ozone trends, LDV emissions yield a peak reduction which exceeds all sources except aircraft and ships. HDV and off-road emissions reduce $PM_{2.5}$ in SoCAB and Central Valley with similar magnitude. Shown in Fig. A6, trends for winter $PM_{2.5}$ differ from those in summer. Aircraft have the highest peak impact, with HDV, off-road, LDV, ships, and rail following. In contrast to summer, HDV have a much larger impact than LDV on winter $PM_{2.5}$. Spatially, impacts on winter $PM_{2.5}$ also differ from summer $PM_{2.5}$ as areas of peak impact are associated with the Central Valley instead of SoCAB, with the exception of LDV with pronounced impacts in SoCAB and SF Bay Area.

3.3. Petroleum fuel infrastructure

The previous cases consider the impacts of emissions directly from vehicles only. To assess the impacts of producing and distributing petroleum transportation fuels including gasoline, diesel, jet fuel, etc., scenarios were developed and evaluated for the removal of petroleum fuel infrastructure (PFI) related emissions. Adjusted sources include emissions occurring at refinery complexes, including industrial process (e.g., fuel combustion) and evaporative emissions from storage tanks, etc. Moreover, emissions from activities related to the transport and storage of vehicle fuels to dispensing locations are removed, including emissions associated with vehicle refueling activity. All additional emissions, including those directly from vehicles, are left unchanged. Thus, the PFI Case solely demonstrates the contribution to regional AQ burdens of producing, storing, transporting, and distributing petroleum transportation fuels. However, it should be noted that refineries also output non-transportation related products including various materials and chemicals.

For the U.S. regional method PFI emissions in CA yield effects on ozone and $PM_{2.5}$ that peak at -1.55 ppb and -14.49 $\mu g/m^3$, for 8-h ozone and $PM_{2.5}$ respectively (Fig. 5). Reductions occur over large areas of the state and adjacent to large refinery complexes in the SoCAB, the Central Valley, and the Bay Area. The magnitudes of $PM_{2.5}$ reductions are particularly notable with values in excess of those for direct emissions from all sub-sectors with the exception of ships. Similarly, impacts on ozone are comparable to those from removing LDV emissions and the locations of reductions in both ground-level ozone and $PM_{2.5}$ have importance as they coincide with urban regions with high populations and/or that currently struggle to achieve attainment with federal pollutant standards. The CA method predicts refinery emission ozone impacts of 0.7 ppb and 2.64 $\mu g/m^3$ for the summer episode (Fig. A8) and 1.96 $\mu g/m^3$ in the winter episode (Fig. A9). While the impacts are reduced from the U.S. regional method, reductions are nonetheless significant when compared to impacts from transportation sub-sectors including $PM_{2.5}$ in both seasons. Additionally, peak impacts occur in the SoCAB for both ozone and $PM_{2.5}$. Shown in Fig. A7, removing PFI emissions in TX yields changes that reach −4.59 ppb ozone and −3.71 μg/m^3 $PM_{2.5}$ and extend over large portions of the TX region.

4. Discussion

LDV emissions impacts are moderate in all regions for ozone relative to other sub-sectors for both the U.S. and CA methods. Similar effects in regards to future ozone impacts of LDV emissions have been reported for CA (Collet et al., 2012) and the U.S. (Collet et al., 2012; Vijayaraghavan et al., 2012; Nopmongcol et al., 2017; Collet, 2017; Collet et al., 2014). This is not surprising given the major reduction in many criteria pollutants that has already occurred in the LDV sub-sector and that are projected to occur fleet-wide. These reductions are a product of various efforts to improve LDV performance and reduce emissions and reflect the current regulatory focus and strict LDV emissions policies at both the Federal and State levels. However, the moderate impacts attributable to LDVs relative to other transportation sources should be evaluated in the context that, (1) improvements occur in populated urban regions and thus have human health implications, and (2) LDVs will continue to be an important source of domestic GHG emissions. For example, the CA method predicts that LDV emissions impact $PM_{2.5}$ notably in key regions for both winter and summer. Additionally, the considerable AQ impacts of producing and distributing motor gasoline should be considered as a necessary companion of LDV fleets. Thus, while LDVs will continue to be an important target for alternative low-emitting technologies it may be more effective to develop policy targeting mitigation on the basis of GHG reductions.

The primary and secondary pollutant impacts occurring in both methods marks off-road equipment as a required target for future mitigation strategies. The importance of off-road sector emissions to regional AQ burdens in this work supports findings from other studies (Nopmongcol, 2017; Collet, 2017). Additionally, contributions to ground-level ozone and $PM_{2.5}$ warrant policy attention in the development and deployment of cleaner technologies and fuels for ships. Aircraft emissions are associated with some of the largest magnitude impacts on both ozone and $PM_{2.5}$, although the impacts are highly localized in comparison to other sectors. Nevertheless, major airports are often located in densely populated regions and represent an important source for mitigation that should be considered. In particular, transportation activities associated with goods movement including ships, off-road, HDV, and rail, emerge as a dominant source of regional air pollution. Considering the co-location of sources sites of concentrated activity, such as ports and regional distribution centers, emerge as perhaps the most important to address. A current understanding of this exists in CA and programs and policies are in place and/or under development to reduce emissions, e.g., CA's Goods Movement Emission Reduction Plan (CARB, 2006b) and Sustainable Freight Action Plan (California Sustainable Freight Action Plan, 2016). However, expected growth in demand for global shipping in tandem with reduced emissions from other sectors will increase the importance of reducing port emissions. Further, operational and other constraints increase the difficulty of deploying alternative strategies for some technologies, e.g., fuel energy density requirements of ships and HDV, duty cycles and technical specificity of off-road equipment. Thus, the results support the urgent need for policy support for research, development, and deployment plans for low- and zero-emissions technologies and fuels in goods movement.

Reductions in ozone and $PM_{2.5}$ that occur from PFI-related sources also merit investigation for future mitigation strategies. For example, in TX reductions in ozone from PFI emissions exceed those from direct emissions from any of the individual transportation sub-sectors. Therefore, maximizing the benefits of deploying alternative transportation technologies and fuels will likely require corresponding reductions in emissions from PFI. The impacts described here are consistent with the current understanding in the literature; particularly for large capacity refining complexes. Refining facilities co-emit large quantities of NO_x and reactive VOCs (Sexton and Westberg, 1983); conditions typically associated with rapid and efficient ozone formation (Ryerson et al., 2003), and have been shown to contribute to elevated ozone and $PM_{2.5}$ levels in urban regions (Kulkarni et al., 2007; Murphy and Allen, 2005; Kleinman, 2002b). However, these impacts have never previously been assessed for multiple regions in future years until the current study. Further, refining is generally a large scale, high capacity process and many facilities are operated continuously resulting in the steady generation of large quantities of emissions for extended periods (e.g., months to years) which differs from the defined temporal emissions signatures from other sub-sectors.

Meteorological and other seasonally-dependent factors may necessitate the development of policy designed to target different sources during different annual periods. While seasonally dependent control strategies have been utilized before (examples include seasonal restrictions on wood burning (Bay Area Air Quality Management District) and variable fuel blends (U.S. Environmental Protection Agency)), meeting targets may require augmented seasonal strategies including the deployment of alternative technologies or the restriction of transportation source activity.

Differences in metrics prevent a direct comparison of the two methods used here, but insights for policy development can still be gained. It is interesting to note that many of the trends for sub-sectors remained constant across methods, e.g., the importance of off-road equipment and ships in future years to ozone and $PM_{2.5}$ concentrations and the relatively moderate impact of LDV on ozone. Contrastingly, differences include the greater impact of HDV on ozone and $PM_{2.5}$ and LDV on $PM_{2.5}$ in the CA method. The CA method is significantly updated for both model versions and data inputs and therefore represents a prediction of future AQ with improved accuracy relative to the U.S. regional method. The CA method is better for applications requiring a single BAU case, or with high accuracy requirements. However, the method is rigid in that alternative cases are not available and the method is not applicable to other regions of the U.S., which prevents comparison across regions. Conversely, the US method can be used to study the outcome of various policies and programs on future emissions easily due to the flexibility of running MARKAL with different constraints, e.g., impacts of including externalities such as a carbon tax or health damages fee (Brown et al., 2013), techno-economic variation in resources (McLeod et al., 2014), emission scenarios (Trail, 2014), and many others. The MARKAL method also feasibly allows for assessment of individual regions (Nsanzineza, 2017) or at the U.S. scale (Brown et al., 2013). Therefore, the choice of modeling tools for future AQ assessment must consider the scope and goal of the assessment.

5. Conclusion and policy implications

This work quantifies the impacts of emissions from various mobile transportation sources and petroleum fuel production infrastructure on ambient ground-level ozone and $PM_{2.5}$ concentrations in three regions of the U.S. in 2055, and provides a more in-depth study of CA in 2035 with updating modeling methods and data inputs. The transportation sector is a major contributor to ground-level ozone and $PM_{2.5}$ concentrations in the U.S.; thus all transportation sub-sectors can reduce emissions to produce notable AQ improvements for both modeling methods and horizon years. Emissions from ships and off-road sources in urban regions are associated with significant effects on ozone and $PM_{2.5}$ in all regions studied for both methods. Additionally, HDV emissions result in notable contributions to regional AQ burdens. In contrast, LDV and rail emissions have a lesser impact for both methods relative to other transportation sources. Emissions from producing, storing, and distributing petroleum-based fuels have important impacts on ambient AQ comparable to direct emissions from mobile sources. These results provide insight into emerging transportation sector sources for regional AQ improvement planning with the central recommendation to enhance the focus of future mitigation strategies upon the goods movement sectors, off-road sources, and upon petroleum fuel refining activities.

Policies designed to achieve emission reductions must be developed with a detailed understanding of potential strategies, including their evolution in terms of technological maturity and economics. For example, pursuing zero emission off-road equipment will be more straightforward for some applications (e.g., forklifts, lawn and garden) relative to others (e.g., agricultural tractors, construction and mining equipment) due to the current commercial availability of technologies, technical demands required for each application, etc. Similarly, reducing ship emissions can occur from a variety of strategies including operational changes, efficiency measures, and the deployment of alternative propulsion and auxiliary engine technologies and fuels. Policies targeting emission reductions from these sectors should be designed to integrate strategies with high technological readiness in the near-term, while supporting the development and advancement of the "ultimate" answer (such as zero emission technologies) in the long term.

Implications from this work also demonstrate the importance of considering the full life cycle of vehicle and fuel combinations when developing transportation policy within an environmental framework. This is true not only for conventional vehicles and fossil fuels, but also alternative low-emitting technologies. For example, transitions to electrified vehicles should be encouraged through policies that consider emission impacts on the electrical grid to avoid worsening of AQ due to power plant emissions. Similarly, shifts to biofuels must carefully consider the life cycle impacts relative to the conventional fuel being displaced.

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Appendix A. Considering regional air quality impacts of transportation sector emissions sources

Air quality results

U.S. regional method ozone sub-sector results

See Figs. A1–A3

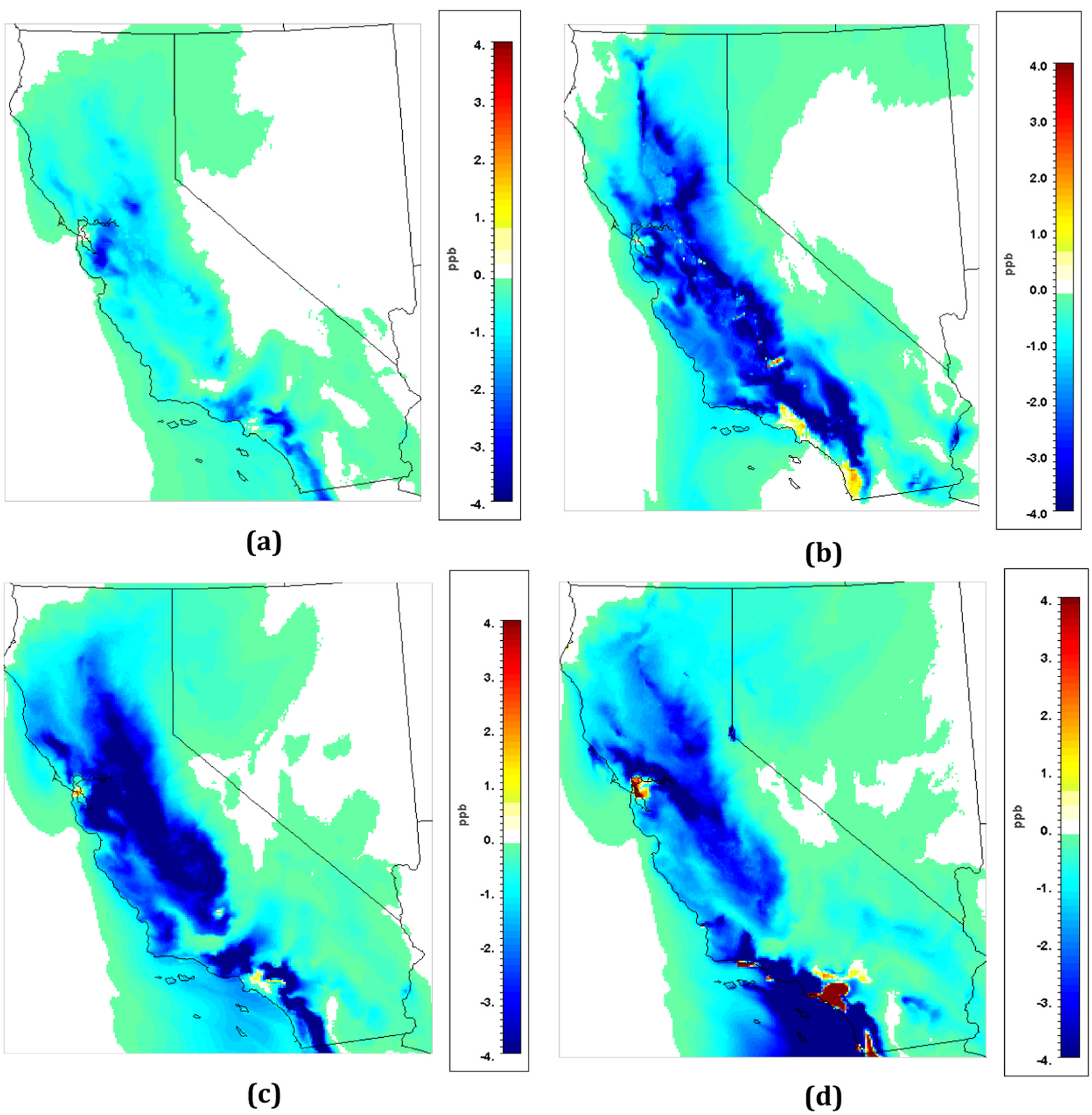

Fig. A1. Impacts in CA on maximum 8-h average ozone from (a) LDV, (b) HDV, (c) Off-road, and (d) Marine and Rail in 2055.

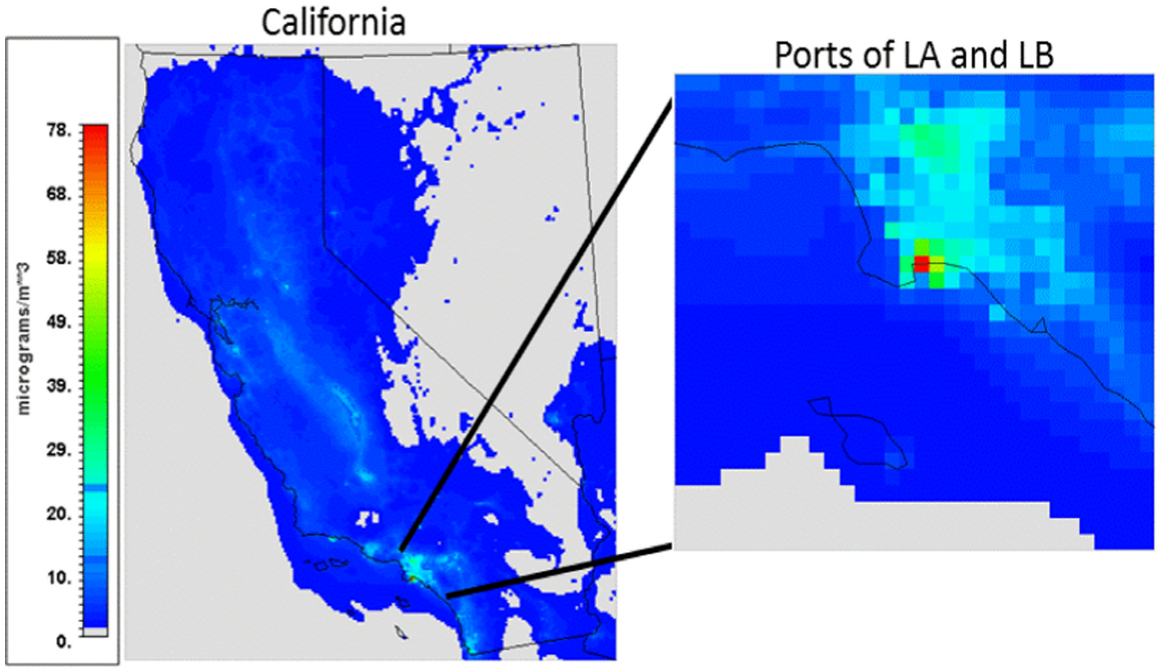

Fig. A2. 2055 Base Case 24-hr PM2.5 levels in CA with area associated with peak concentrations around the POLB and POLA shown.

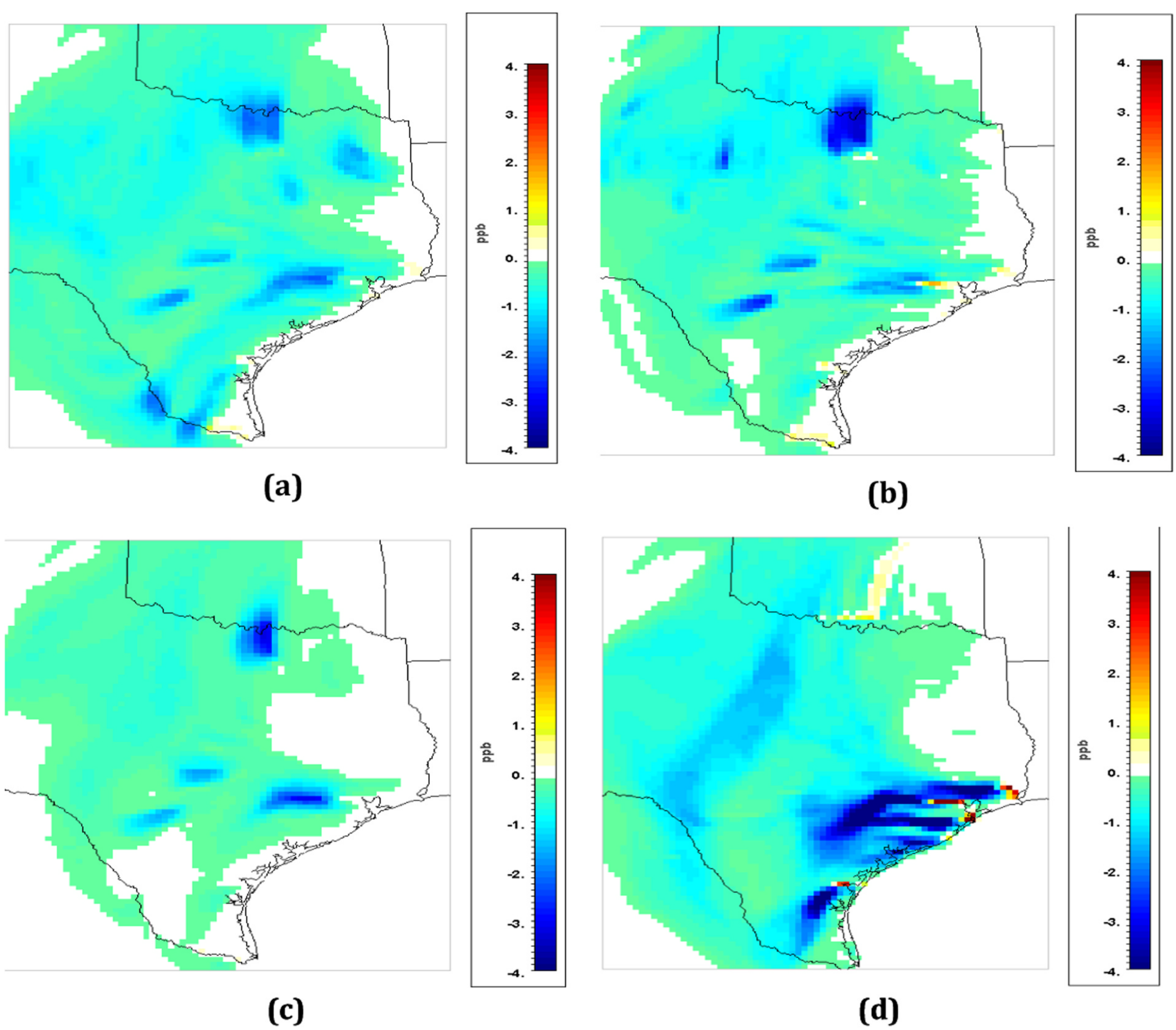

Fig. A3. Impacts in TX on maximum 8-hr average ozone in 2055 from (a) LDV, (b) HDV, (c) Off-road, and (d) Marine and Rail.

U.S. regional method $PM_{2.5}$ sub-sector results

See Fig. A4California method $PM_{2.5}$ sub-sector results. See Figs. A5 and A6

PFI AQ results

See Figs. A7–A9

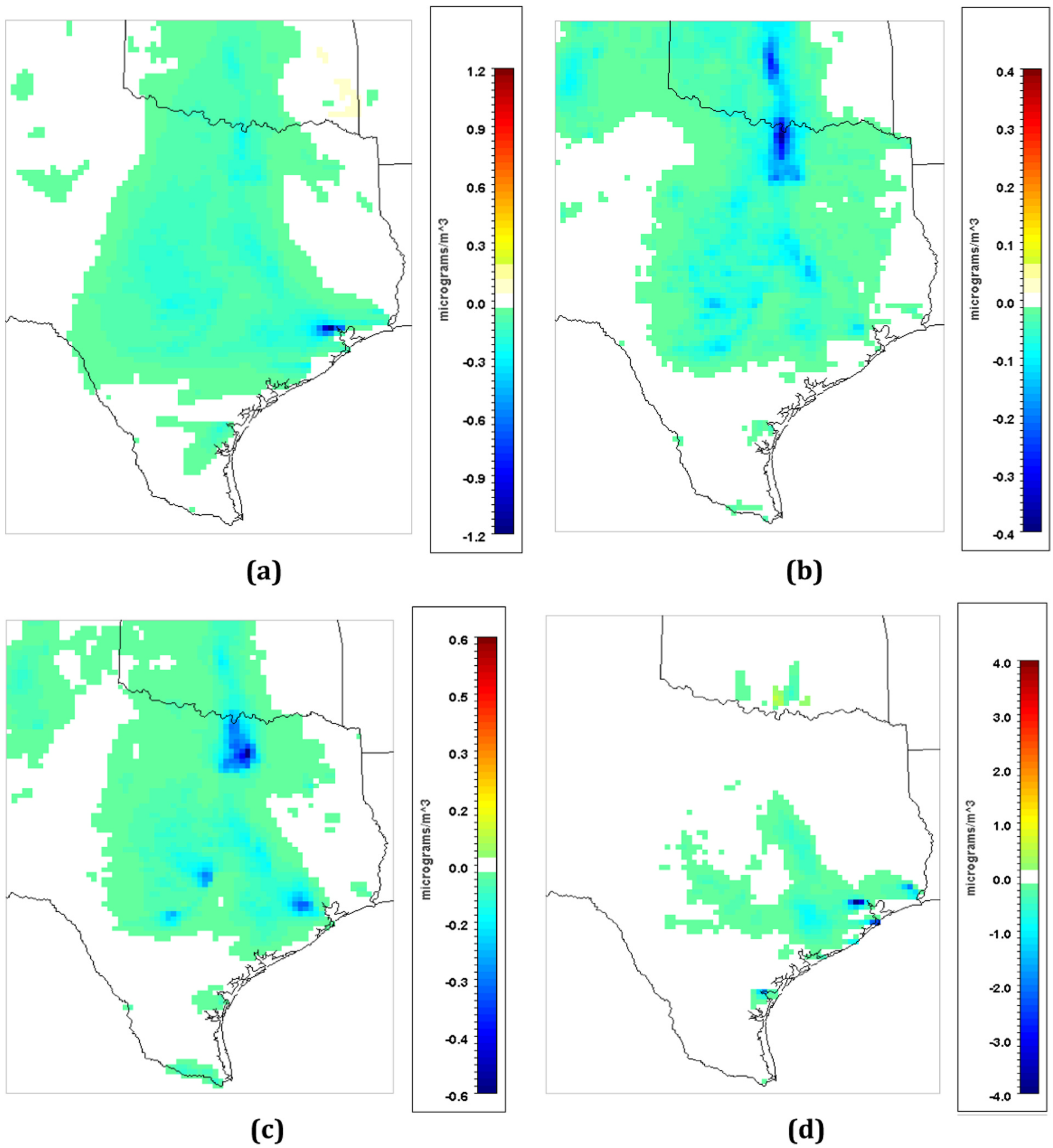

Fig. A4. Impacts in TX on 24-h PM2.5 from (a) LDV, (b) HDV, (c) Off-road, and (d) Marine and Rail.

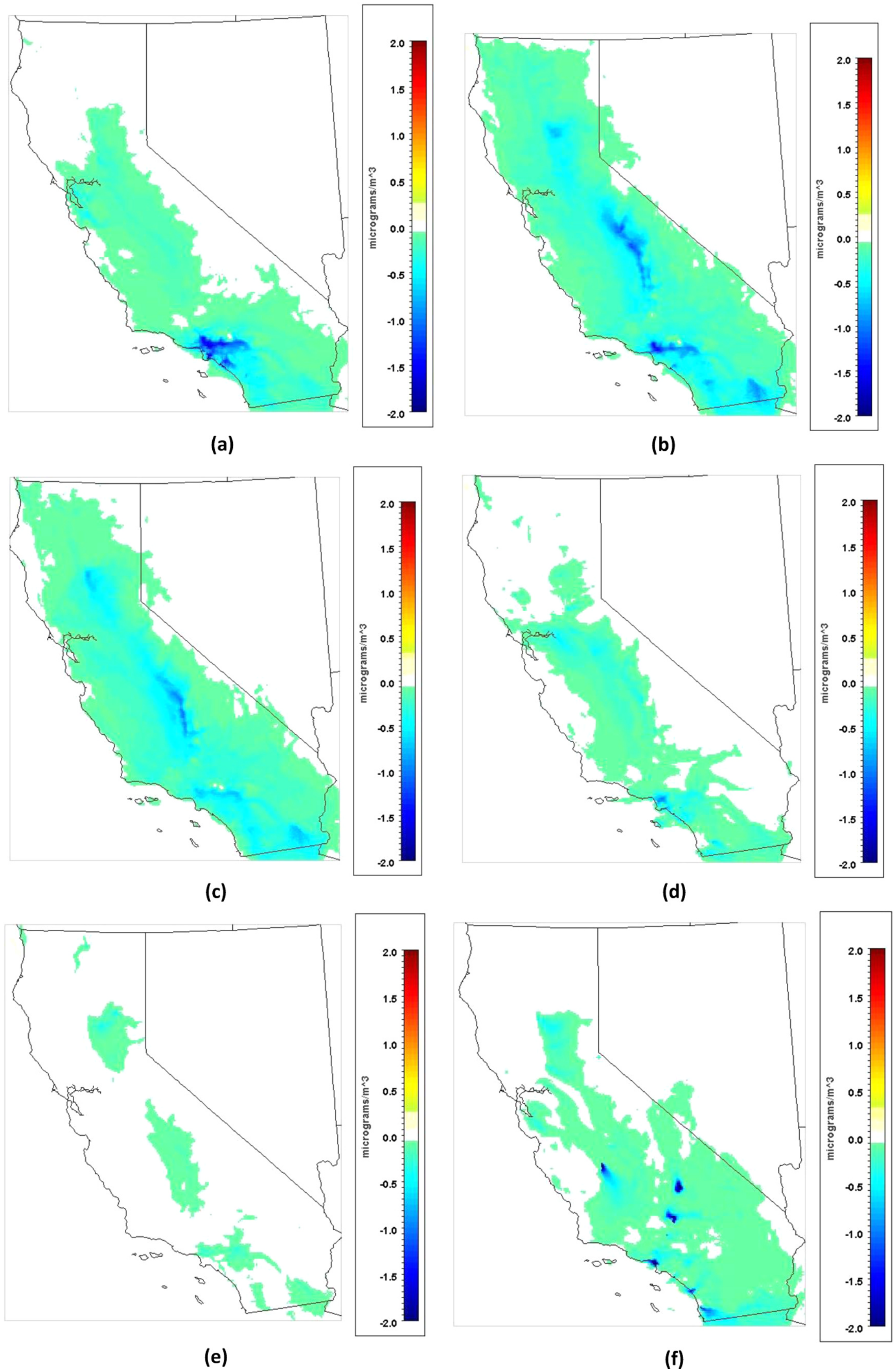

Fig. A5. Predicted differences in peak 24-h average $PM_{2.5}$ for a summer episode in CA between the baseline and scenarios involving the removal of emissions from (a) LDV, (b) HDV, (c) Off-road, (d) Ships, (e) Rail, and (f) aircraft in 2035.

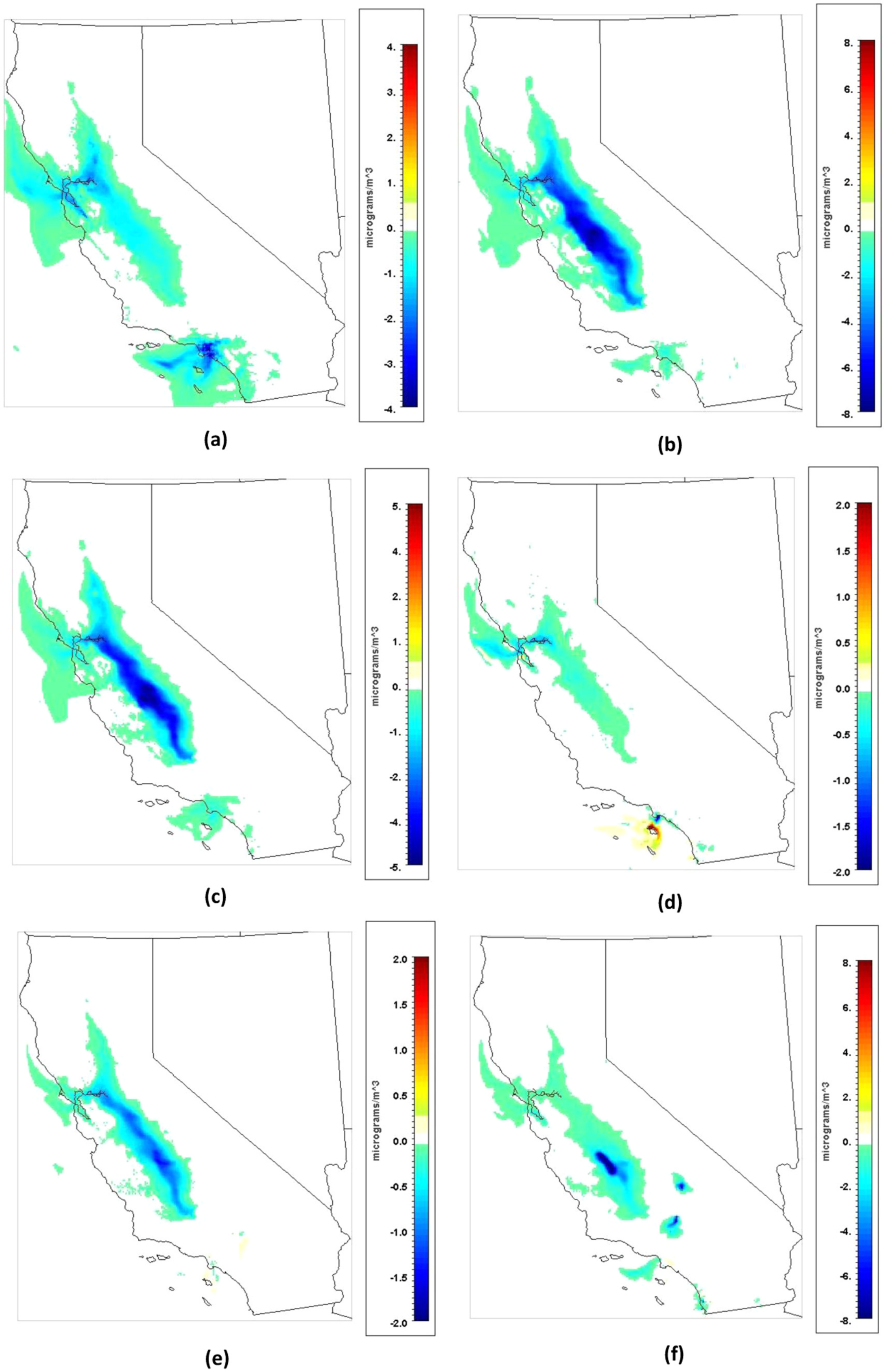

Fig. A6. Predicted differences in peak 24-hr average $PM_{2.5}$ for a winter episode in CA between the baseline and scenarios involving the removal of emissions from (a) LDV, (b) HDV, (c) Off-road, (d) Ships, (e) Rail, and (f) Aircraft in 2035.

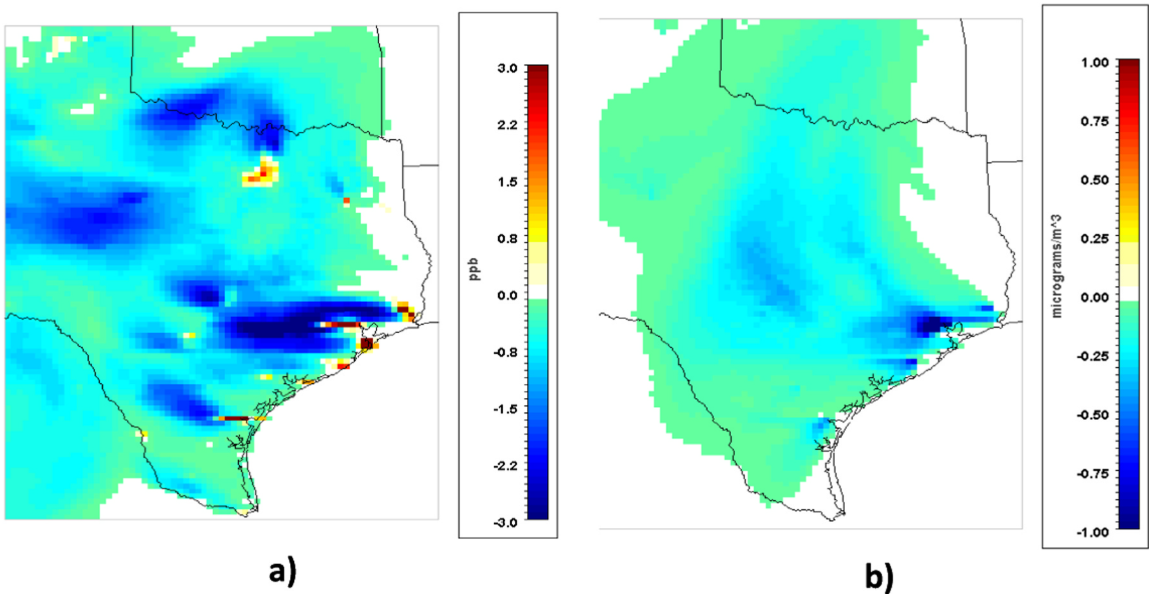

Fig. A7. Predicted differences in a) max 8-h ozone and b) 24-h $PM_{2.5}$ for the PFI Case in TX.

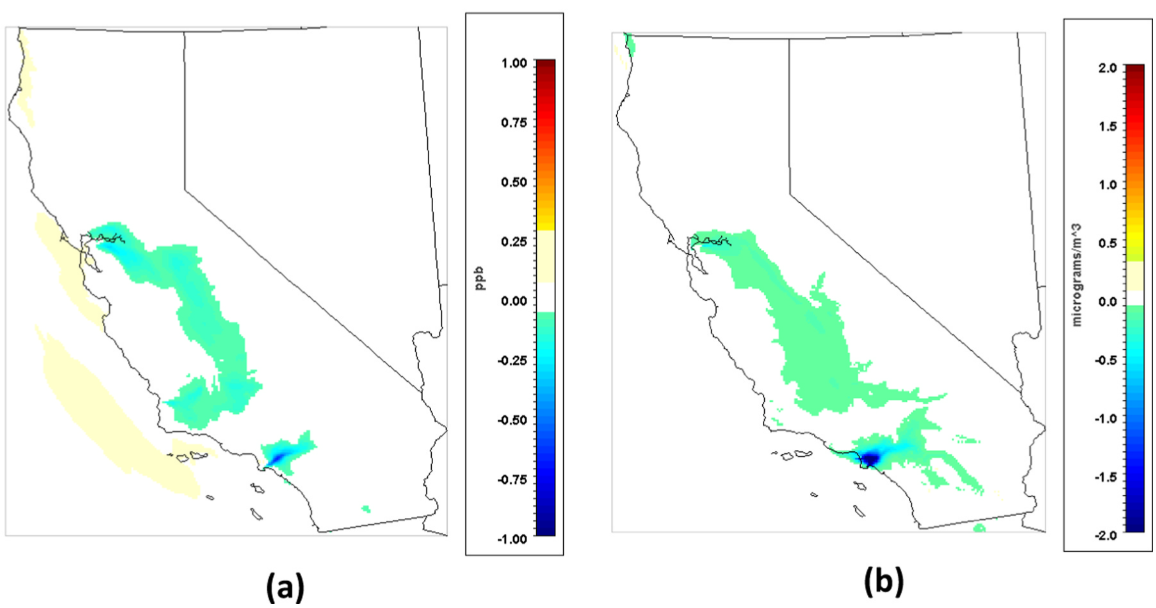

Fig. A8. Predicted differences in a) max 8-h ozone and b) 24-hr $PM_{2.5}$ for the PFI Case relative to the Base Case in CA predicted for the summer episode in 2035.

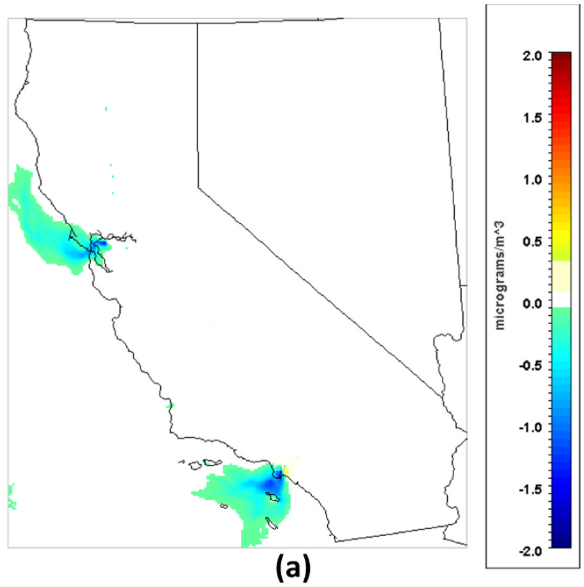

Fig. A9. Predicted differences in 24-hr $PM_{2.5}$ for the PFI Case relative to the Base Case in CA predicted for the winter episode in 2035.

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