Estimating Criteria Pollutant Emissions Using the California Regional Multisector Air Quality Emissions (CA-REMARQUE) Model v1.0

Christina B. Zapata[1], Chris Yang[2], Sonia Yeh[2], Joan Ogden[2], Michael J. Kleeman[1]

[1] Department of Civil and Environmental Engineering, University of California – Davis, Davis, California, USA
[2] Institute of Transportation Studies, University of California – Davis, Davis, California, USA

Correspondence to: Michael J. Kleeman (mjkleeman@ucdavis.edu)

Abstract. The California REgional Multisector AiR QUality Emissions (CA-REMARQUE) model is developed to predict changes to criteria pollutant emissions inventories in California in response to sophisticated emissions control programs implemented to achieve deep Green House Gas (GHG) emissions reductions. Two scenarios for the year 2050 act as the starting point for calculations: a Business as Usual (BAU) scenario and an 80% GHG reduction (GHG-Step) scenario. Each of these scenarios was developed with an energy economic model to optimize costs across the entire California economy and so they include changes in activity, fuels, and technology across economic sectors. Separate algorithms are developed to estimate emissions of criteria pollutants (or their precursors) that are consistent with the future GHG scenarios for the following economic sectors: (i) on-road, (ii) rail and off-road, (iii) marine and aviation, (iv) residential and commercial, (v) electricity generation, and (vi) biorefineries. Properly accounting for new technologies involving electrification, bio-fuels, and hydrogen plays a central role in these calculations. Critically, criteria pollutant emissions do not decrease uniformly across all sectors of the economy. Emissions of certain criteria pollutants (or their precursors) increase in some sectors as part of the overall optimization within each of the scenarios. This produces non-uniform changes to criteria pollutant emissions in close proximity to heavily populated regions when viewed at 4km spatial resolution with implications for exposure to air pollution for those populations. As a further complication, changing fuels and technology also modify the composition of reactive organic gas emissions and the size and composition of particulate matter emissions. This is apparent most notably through a comparison of emissions reductions for different size fractions of primary particulate matter. Primary $PM_{2.5}$ emissions decrease by 4% in the GHG-Step scenario versus the BAU scenario while corresponding primary $PM_{0.1}$ emissions decrease by a factor of 36%. Ultrafine particles ($PM_{0.1}$) are an emerging pollutant of concern expected to impact public health in future scenarios. The complexity of this situation illustrates the need for realistic treatment of criteria pollutant emissions inventories linked to GHG emissions policies designed for fully developed countries and states with strict existing environmental regulations.

1 Introduction

Many countries around the world are debating cost-effective candidate strategies to mitigate threats to long-term prosperity including climate change and threats to public health. These specific issues are at least partially linked through regional air quality. Realistic mitigation plans for Green House Gas (GHG) emissions (CO_2, CH_4, N_2O, etc) usually include measures encouraging reduced energy consumption or changes to energy sources leading to reduced GHG emissions. These measures also impact emissions of criteria pollutants or their precursors (PM, NOx, SOx, VOCs, NH_3, etc) that influence regional air quality. Air quality influences public health through impacts on mortality (primarily related to $PM_{2.5}$) and morbidity (primarily related to $PM_{2.5}$ and O_3).

Many previous attempts to characterize the impact of climate policies on criteria pollutant emissions, air quality, and public health have often emphasized countries where potential health savings are largest. These previous studies have also usually performed calculations for large geographic areas without resolving details at regional scales appropriate for California (Bollen et al., 2009; Garcia-Menendez et al., 2015; Rafaj et al., 2012; Shindell et al., 2012; van Aardenne et al., 2010; West et al., 2013). These studies represent California with only a small number of grid cells or they uses simplistic representations of California's energy economy.

More recent studies addressing interactions between climate policies, emissions, and air quality in the US (Keshavarzmohammadian et al., 2017; Loughlin et al., 2011; Ran et al., 2015; Rudokas et al., 2015; Trail et al., 2015; Zhang et al., 2016) have allocated future emissions using enhanced population surrogates (Ran et al., 2015) and federal climate policies (Trail et al., 2015). The current study builds on this previous work to explicitly account for California's ambitious climate regulations broken out by detailed sectors including realistic siting of biofuel facilities. The current study also considers the effects of regenerative braking, and exhaust particulate size and speciation changes from the heavy use of alternative and renewable fuels across multiple economic sectors. These enhancements support the desired level of detailed analysis for the intersection of air, climate, and energy choices in California.

The purpose of this paper is to describe the California REgional Multisector AiR QUality Emissions (CA-REMARQUE) model that can translate complex GHG mitigation scenarios to criteria pollutant emissions inventories with sufficient detail to support fine-scale air quality models and public health analysis. Here we emphasizes solutions that optimize state-wide total GHG emissions across the entire California economy, with potential tradeoffs between different source types to achieve this objective. The complex optimization problem requires an energy economic model and so we focus on scenarios predicted by the CA-TIMES energy economic model as the starting point for the analysis. The detailed algorithms within the CA-REMARQUE model are then developed to translate predicted changes in GHG emissions associated with source activity, fuels, and technology to criteria pollutant emissions that are spatially-resolved (4 km) for each sector of the California economy. Changing emissions profiles caused by fuel substitutions are also accounted for. Final results are compared to an expert-analysis method developed for a previous global analysis to illustrate why the complex methods described in this study are needed when analysing developed regions like California that have major diversified economies and a long history of environmental regulations.

2 Methodology

Energy scenarios are translated to criteria pollutant emissions inventories by the CA-REMARQUE model in a multi-step process with unique algorithms developed for each major sector of the economy that emits air pollution precursors. All calculations start with energy scenarios developed by the energy economic model CA-TIMES. The details needed to produce criteria pollutant emissions inventories are discussed in the following sections.

2.1 CA-TIMES Energy Model and Energy Scenarios

CA-TIMES (McCollum et al., 2012; Yang et al., 2014; Yang et al., 2015) is a bottom-up energy-economic model originally based on the MARKAL TIMES model (Loulou et al., 2016). CA-TIMES is a cost-minimization optimization model that balances energy supply and demand system-wide from all economic sectors of the energy economy. Demand sectors include transportation, industrial, residential, commercial, and agricultural. Fuel and electricity supply includes electric, biofuel, hydrogen production plants and biofuel and petroleum refineries. Demand was assumed fixed for the scenarios considered (Yang et al., 2014; Yang et al., 2015). CA-TIMES allows imports from out of state, such as oil, natural gas, and electricity. Renewables and Biomass are handled separately and modelled explicitly as located in or out of state and imports are determined on a cost basis. CA-TIMES contains capital and operation costs for each technology, diverse fuel and energy carriers, and calculates GHG emissions for CO_2, CH_4, and N_2O.

The case studies considered in the present study focus on two CA-TIMES scenarios in 2050: (i) a Business as Usual (BAU) scenario that achieves the goals outlined in California Assembly Bill 32 (AB32), the Global Warming Solutions Act of 2006 and (ii) a climate friendly GHG-Step scenario that achieves an 80% reduction (relative to 1990 levels) in GHG emissions by 2050. Statewide GHG emissions under each scenario are summarized in Figure 1. In the GHG-Step scenario a “step” GHG emissions constraint is applied in which a constant 2020 cap is held until 2050, and then an 80% reduction is applied from 2050 onward. This allows the model freedom to adopt strategies that lower GHG emissions prior to 2049 if those strategies minimize costs. This 2050 GHG constraint causes aggressive change over the period 2040-49 but does not shock to the energy system in 2050 because the CA-TIMES model has perfect foresight and optimally minimizes the energy system cost (with a 4% discount factor) over the entire period from 2010 to 2050 making investment decisions to meet targets. Also, CA-TIMES investments in low-GHG technologies start slowly and grow to reach the required market share to meet the targets since technologies have finite lifetimes and cannot take over respective markets instantaneously. The criteria pollutant emissions between 2010 and 2049 were not analysed in the current study but a summary of CA-TIMES results for intermediate years is provided by (Yang et al., 2015). Both BAU and GHG-Step scenarios include current and sunset GHG regulations in California (Corporate Average Fuel Economy (CAFE) Standards (California Air Resources Board, 2005, 2009b, 2010b), Zero Emission Vehicle (ZEV) Mandate (California Air Resources Board, 2012b, c, d, e, f), Low Carbon Fuel Standard (LCFS) (California Air Resources Board, 2009c, 2011c), Cap-and-Trade Program (California Air Resources Board, 2011d, 2017) and federal and state incentives (tax credits and subsidies). CA-TIMES predicts total annual energy consumption in California for the year 2050 to be 8,763 PJ in the BAU scenario and 7,679 PJ in the GHG-Step scenario (reference value for 2010 is approximately 7,500 PJ) (Yang et al., 2015).

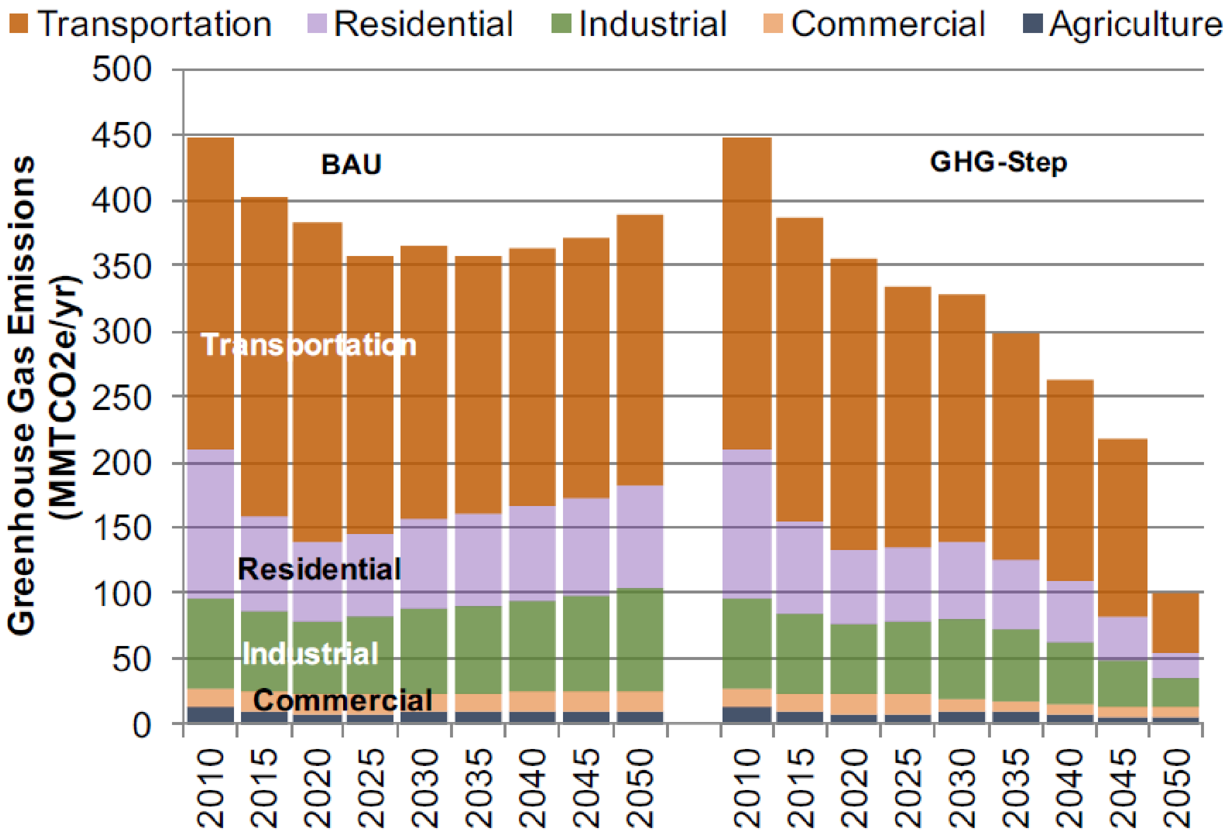


Figure 1: Greenhouse gas emissions in California under the BAU and GHG-Step scenarios.

The methods to estimate criteria emissions for different sources developed in the current paper take advantage of the best available information describing future energy and emissions as a function of location. The quality of this information varied considerably for each major source category and so the details of the methodology also varied. Figure 2 illustrates an overview of the general procedure. The changes in energy consumption and GHG emissions produced by CA-TIMES for each energy sector in the year 2050 were translated to changes in criteria pollutant emissions by accounting for changing energy activity levels or fuel switching. Literature searches were conducted to identify any previous studies describing spatial locations of future emissions within California. Altered emissions for the year 2050 were then projected from a 2010 emissions inventory with 4 km spatial resolution provided by the California Air Resources Board (CARB). Additional details for each major source type are discussed below.

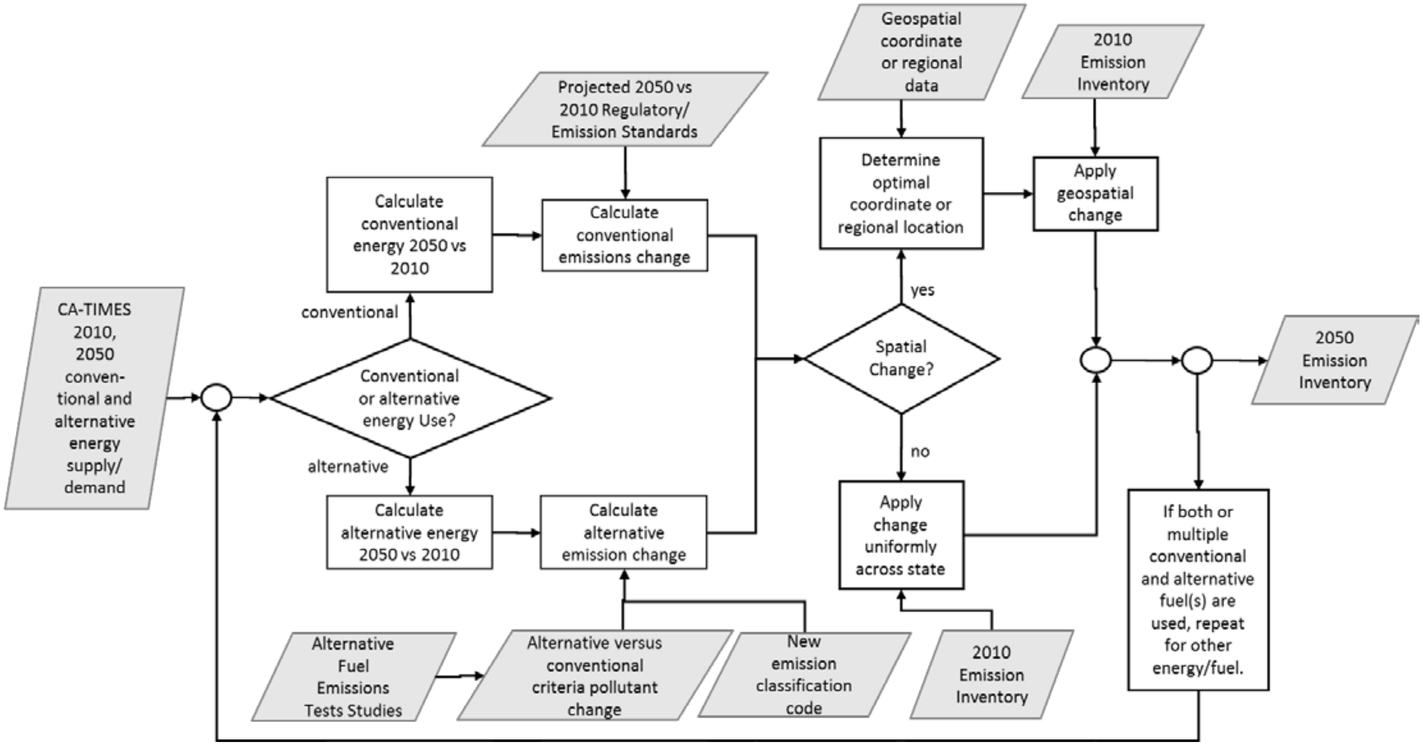


Figure 2: Process diagram of emission inventory generation for each sector or mode.

2.2 CA-REMARQUE On-road Mobile Algorithms

On-road mobile sources include passenger cars, light duty trucks (LDT), medium duty trucks (MDT), heavy duty trucks (HDT), buses, motorcycles, and motor homes. On-road emissions were generated in a multi-step process summarized in Fig. 3. In the first step, 2010-2035 emission projection trends from the EMission FACtor (EMFAC) 2011 model (California Air Resources Board, 2011a) were used to extrapolate further to 2050. In the second step, an intermediate 4km vehicular emissions inventory was generated by combining EMFAC 2050 projections with 2010 4km emission inventory as a spatial surrogate. In the third step, the 2050 fossil fuel vehicular emission rates that were projected from EMFAC as well as new emission rates gathered from alternative fuel emission literature were used to scale the 4km intermediate mobile emission inventory based on the vehicle miles travelled (VMT), trips, and vehicle class and (conventional and alternative) fuel consumption output produced for each CA-TIMES scenario.

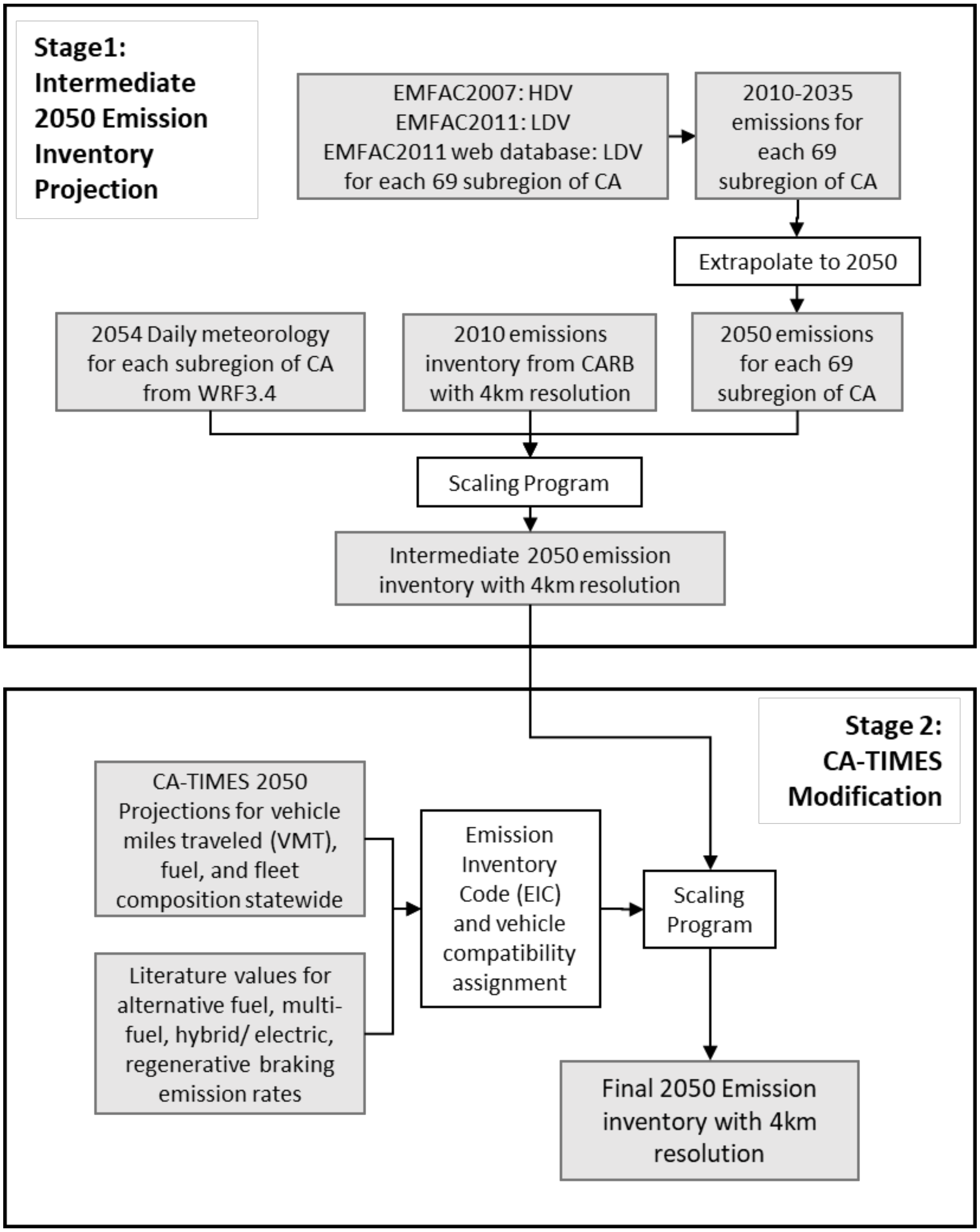


Figure 3: Simplified sequence of algorithms, calculations, and inputs used in developing the CA-TIMES alternative fuel on-road mobile emissions inventory per scenario. EIC is emission inventory code.

2.2.1 EMFAC Emissions and Activity Projections

Criteria pollutant emissions for on-road mobile sources in future years were forecast using the EMFAC 2011 model developed by the California Air Resources Board (CARB) (California Air Resources Board, 2011a). EMFAC 2011 accounts for annual VMT trends and vehicle fleet composition turnover using Department of Motor Vehicle (DMV) data. EMFAC incorporates the latest on-road mobile policies including the Low Emission Vehicle emission standards, Low Carbon Fuel Standard (LCFS), Pavley Clean Car Standard, and the Truck and Bus ruling (California Air Resources Board, 2011). EMFAC 2011 predicts past, present, and future year (up to 2035 or 2040) emissions including anticipated future emissions standards and regulations specific to California. EMFAC predicts emissions and energy activity (VMT, trips, vehicles, gallons fuels) for 69 Geographical Area Indexes (GAIs) which represent the intersection of air basins and counties (listed in Table S1).

In the current study, EMFAC was run for each calendar year from 2020–2035 to infer the emissions trends that could then be extrapolated to 2050. A simple linear regression model was used to represent VMT over the period 2020-2035, while a logarithmic regression model was fit to pollutant emissions for each vehicle type over the same time period. Future studies will use EMFAC 2014 which directly predicts emissions in 2050 making this step unnecessary.

2.2.2 Spatial Allocation of Mobile Source Emissions in an Intermediate 2050 Inventory

An existing on-road mobile emissions inventory for the year 2010 with 4 km spatial resolution served as the starting point for the projection of an intermediate emissions inventory in 2050. Scaling factors to account for VMT growth and adoption of existing policies were first calculated as the ratios between EMFAC emissions from 2010 and (extrapolated) 2050 within each of the 69 GAI regions. Separate scaling factors were developed for each pollutant emitted from different vehicle classes and control technologies as represented by unique emission inventory codes (EICs). The combined intermediate emissions (em) scaling factor $SF_{act + met}$ defined in equation (3) reflects independent changes in activity (act) (Eq. 1) and meteorology (met) (Eq. 2). Future 2054 temperature and relative humidity generated at 4km resolution with WRF3.2 (Zhang et al., 2014) were averaged to GAI regions used by EMFAC to produce hour-specific reactive organic gas (ROG) emission rates that vary from the annual average emission rates. Activity is either defined as vehicle miles travelled (VMT) or vehicle trips, depending on the emission process. For example, activity equals VMT for tailpipe emission rates (e.g. grams NO $mile^{-1}$) or tire and brake wear emissions (grams PM $mile^{-1}$). Otherwise, activity equals the number of vehicles within each type/fuel/aftertreatment category such as for evaporative emissions of non-methane hydrocarbons (grams NMHC $vehicle^{-1}$) from the fuel system (non-tailpipe emissions). Emission rates are highly dependent on the emission process (evaporative, exhaust, tire or brake wear), fuel (gasoline or diesel) and the aftertreatment device (catalytic or non-catalytic).

Emissions within each 4km grid cell of the 2010 inventory are multiplied by the 2050 to 2010 scaling factor $SF_{act+met}$ to estimate the "intermediate" 2050 emissions that will be further modified according to various additional policy choices represented in CA-TIMES.

$$SF_{act} = \frac{em(act_{2050}, met_{2010})}{em(act_{2010}, met_{2010})} \quad (1)$$

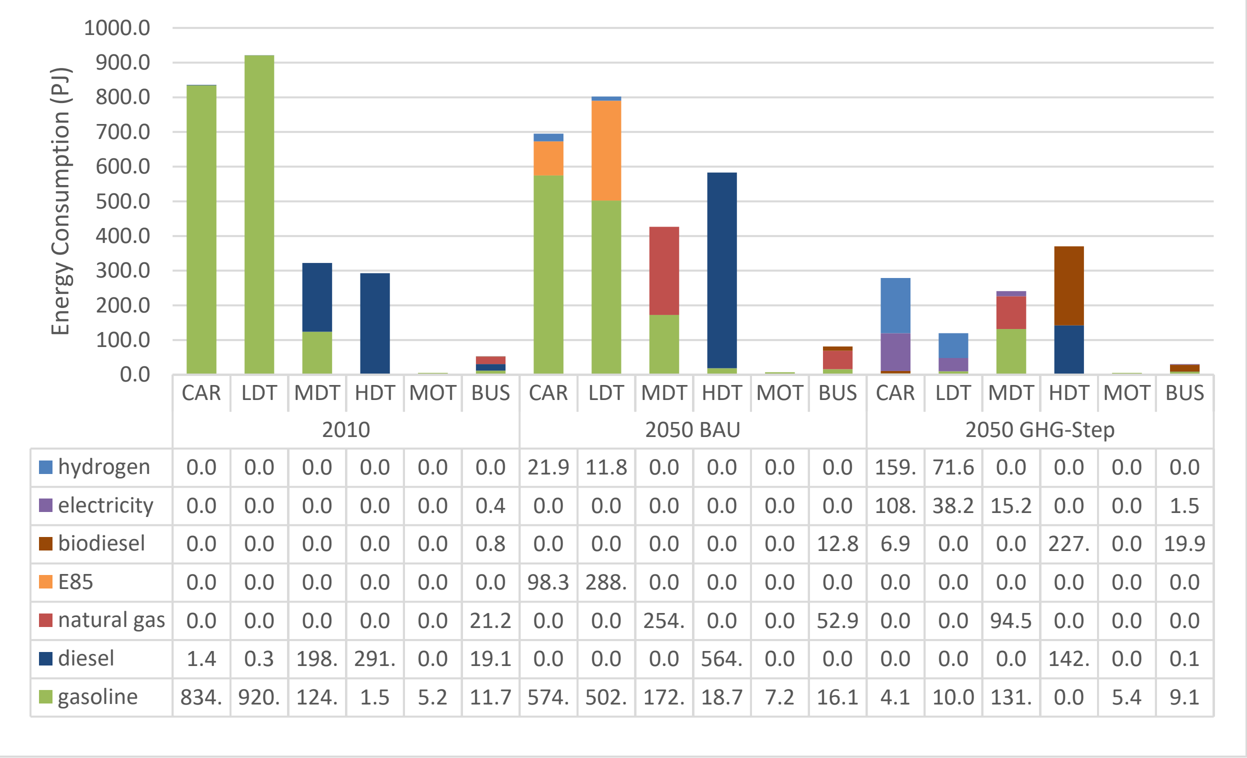
$$SF_{met} = \frac{em(act_{2010}, met_{2050})}{em(act_{2010}, met_{2010})} \quad (2)$$

$$SF_{act+met} = SF_{act} \cdot SF_{met} \tag{3}$$

2.2.3 CA-TIMES Modification of Intermediate 2050 On-Road Mobile Emissions

State-wide CA-TIMES scaling factors were applied uniformly at all locations to the 2050 intermediate emissions inventory described in the previous section to produce the final 2050 emissions inventory. EMFAC accounts for population growth and emissions changes that are required by existing air quality rules and regulations through 2050. CA-TIMES accounts for additional changes that will be required to comply with state GHG targets but which have not yet been placed into emissions rules and regulations. The final inventory retains the spatial and temporal features inherent in the intermediate emissions inventory but incorporates updated information about new fuels, technologies, and emissions rates based on state-wide predictions from CA-TIMES (Fig. 4).

EMFAC vehicles classes expressed as EIC codes were mapped to compatible vehicle classes used by CA-TIMES as described in Table S2. Spark ignition (gasoline) vehicles in CA-TIMES were further classified as catalyst-equipped or non-catalyst-equipped to match EMFAC categories. EMFAC resolves non-catalyst-equipped and catalyst-equipped gasoline vehicles into several sub-categories (light-heavy duty truck (LHDT) and heavy-heavy duty truck (HHDT) (see Table S2 for complete description of vehicle classes) while CA-TIMES does not include this level of resolution.



	2010						2050 BAU						2050 GHG-Step					
	CAR	LDT	MDT	HDT	MOT	BUS	CAR	LDT	MDT	HDT	MOT	BUS	CAR	LDT	MDT	HDT	MOT	BUS
🟦 hydrogen	0.0	0.0	0.0	0.0	0.0	0.0	21.9	11.8	0.0	0.0	0.0	0.0	159.	71.6	0.0	0.0	0.0	0.0
🟪 electricity	0.0	0.0	0.0	0.0	0.0	0.4	0.0	0.0	0.0	0.0	0.0	0.0	108.	38.2	15.2	0.0	0.0	1.5
🟫 biodiesel	0.0	0.0	0.0	0.0	0.0	0.8	0.0	0.0	0.0	0.0	0.0	12.8	6.9	0.0	0.0	227.	0.0	19.9
🟧 E85	0.0	0.0	0.0	0.0	0.0	0.0	98.3	288.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
🟥 natural gas	0.0	0.0	0.0	0.0	0.0	21.2	0.0	0.0	254.	0.0	0.0	52.9	0.0	0.0	94.5	0.0	0.0	0.0
🟦 diesel	1.4	0.3	198.	291.	0.0	19.1	0.0	0.0	0.0	564.	0.0	0.0	0.0	0.0	0.0	142.	0.0	0.1
🟩 gasoline	834.	920.	124.	1.5	5.2	11.7	574.	502.	172.	18.7	7.2	16.1	4.1	10.0	131.	0.0	5.4	9.1

Figure 4: CA-TIMES energy consumption by vehicle weight class, fuel, and scenario for on-road sources. Vehicle categories include car, light duty truck (LDT), medium duty truck (MDT), heavy duty truck (HDT), motocycles (MOT), and bus.

The use of new fuels in the on-road fleet required special consideration during preparation of the 2050 emissions inventory. As a starting point, emission rates from EICs representing conventionally-fueled vehicles were calculated from 2050 EMFAC output by dividing each pollutant emission by the respective vehicle activity indicator (either VMT, vehicle number, or fuel consumption) to serve as a baseline for CA-TIMES scenario adjustments. Next, the 181 combinations of alternative fuels and electric hybrid, dedicated or single/multi-fueled applications and vehicles weight classes were mapped to EMFAC by vehicle class and reference fuel (see Table S2 and S3). CA-TIMES predicts the amount of alternative fuel consumed, not the VMT associated with that alternative fuel. The VMT associated with each alternative fuel was therefore estimated as the VMT associated with the conventional fuel divided by the energy content of the consumed conventional fuel (E_v) multiplied by the energy content of the alternative fuel ($E_{v,f}$) output by CA-TIMES. This calculation assumes that vehicle weight and aerodynamics do not change significantly as alternative fuels are adopted. Finally, the emissions rate for each alternative fuel was estimated based on a literature review of emissions factors for conventional versus alternative fueled vehicles. Reference emission rates ($er_{v,ref}$) and "alternative to conventional" scaling factors ($er_{v,f}$ / $er_{v,ref}$) for the vehicle fuels of interest are listed in Table 1.

Table 1: Emission rate changes for alternative fuels in on-road vehicles. Alternative fuels include 85% ethanol 15% gasoline mixture (E85), biodiesel (B100), and compressed natural gas. Conventional fuels include gasoline, diesel, or ultra low sulfur diesel (USLD). After treatment devices include three way catalyst (TWC), diesel oxidation catalyst (DOC), diesel particle filter (DPF), exhaust gas recirculation (EGR), and selective catalytic reduction (SCR).

Alternative Fuel	Reference Conventional Fuel	After-treatment	Pollutant	Alt/ Conv Ratio	Conv % Change	Data Source
E85	Gasoline	same (TWC)	CO	1.00	0.0%	Graham et al. (2008)
			NOx	0.55	-45%	Graham et al. (2008)
			SOx	1.00	0.0%	Assumed
			ROG	1.00	0.0%	Graham et al. (2008)
			PM	0.25	-75%	Hays et al. (2013)
B100	Diesel or ULSD	DOC+ DPF+ EGR+ SCR	CO	0.03	-97%	Alleman et al. (2004), Alleman et al. (2005), Hasegawa et al. (2007)
			NOx	0.85	-15%	Alleman et al. (2004), Alleman et al. (2005), Tsujimura et al. (2007)
			SOx	1.00	0.0%	Assumed
			ROG	0.03	-97%	Alleman et al. (2004), Alleman et al. (2005), Hasegawa et al. (2007)
			PM	0.03	-97%	Alleman et al. (2004), Alleman et al. (2005), Hasegawa et al. (2007), Rounce et al. (2012)
CNG	Diesel or ULSD	TWC	CO	0.67	-33%	Cooper et al. (2012)
			NOx	0.19	-81%	Cooper et al. (2012)
			SOx	1.00	0.0%	Assumed
			ROG	0.34	-66%	Cooper et al. (2012)
			PM	0.08	-92%	Cooper et al. (2012)

Equation (4) illustrates how the total emissions (em_v) were calculated for a given vehicle class (subscript v) by summing the product of the emission rate and VMT for each fuel (subscript f) for the number of different fuels (n) consumed by that vehicle as defined by each CA-TIMES scenario.

$$\mathrm{em_v} = \sum_{\mathrm{f}}^{\mathrm{n}} \underbrace{\mathrm{er_{v,ref}} \cdot \frac{\mathrm{er_{v,f}}}{\mathrm{er_{v,ref}}}}_{\text{Alternative fuel/energy emission rate}} \cdot \underbrace{\mathrm{act_v} \cdot \frac{\mathrm{E_{v,f}}}{\mathrm{E_v}}}_{\text{Proportion of activity by fuel/ energy for vehicle}} \qquad (4)$$

where

v = vehicle type by weight

f = unconventional or alternative fuel type from f1, f2, f3…n

ref = reference (conventional) fuel, typically gasoline or diesel.

em_v = emissions for a give vehicle type per pollutant. Where pollutant is ROGs, CO, NOx, PM10, SOx [tons pollutant].

$er_{v,ref}$ = pollutant emission rate for a vehicle using the reference (conventional) fuel based from EMFAC [tons pollutant VMT^{-1} or tons pollutant $vehicle^{-1}$]

$er_{v,f}$ = pollutant emission rate for a vehicle using an alternative fuel based from EMFAC [tons pollutant VMT^{-1} or tons pollutant $vehicle^{-1}$]

act_v = total vehicular activity (not divided by fuel) [VMT or vehicles]

$e_{v,f}$ = energy consumption for a given fuel by vehicle given by CA-TIMES scenario [PJ]

e_v = total energy consumed for vehicle for all fuels by CA-TIMES scenario [PJ]

Alternative fuels considered by CA-TIMES include 95% volume blend methanol (M95), 85% volume blend ethanol (E85), compressed natural gas (CNG), liquid petroleum gas (LPG), biodiesel, compressed or liquid hydrogen, and electric drivetrains. Electric vehicles (EVs) include hybrid, (HEV), plug-in hybrid (PHEV), and plug-in or battery (PEV or BEV). CA-TIMES often predicted the use of multiple technologies and fuels within the same vehicle weight class (see Table S4 through Table S12 for complete lists). For example, in the case of a hybrid diesel electric vehicle which runs on 3 energy sources, diesel, biodiesel, and electricity, (e.g. a biodiesel PHEV MDT), 3 sets of emission rates (1 for each fuel) were estimated to replace the single emissions rate for the traditional CI engine for this vehicle class (diesel MDT).

Only approximately 10% of the possible vehicle type/fuel/engine combinations considered by CA-TIMES (see Table S4 to Table S12) were actually used in the 2050 BAU and GHG-Step scenarios as the model optimized for low cost and low-carbon solutions. The main alternative liquid or gaseous fuels projected by CA-TIMES were E85, biodiesel, and CNG. CA-TIMES predicted that E85 would displace gasoline while biodiesel and CNG would displace diesel based on the dominant fuel consumed for the same vehicle weight class counterpart. This fuel substitution alters emissions rates for criteria pollutants as shown in Table 1. For battery electric or fuel cell vehicles, the conventional fuel displaced was based on the dominant fuel for that vehicle class, e.g. gasoline for LDVs.

2.2.4 On-Road Mobile PM and Gas Speciation and Size Profile Changes

Tailpipe exhaust, fuel tank evaporative, and brake wear emissions were adjusted when the vehicle fuel or technology was changed. This requires new source profiles to be defined for E85, biodiesel, and CNG fueled vehicles to describe their emissions of speciated volatile organic compounds (VOCs) and size & composition-resolved particulate matter. New emissions inventory codes (EICs) were created (summarized in Table S13) and associated with new VOC and PM emissions profiles (summarized in Tables S14 – S16) for this purpose.

Multiple measurements are available in the literature for the composition of exhaust from ethanol-fueled vehicles. In the present study, the average VOC profiles measured using the Federal Test Procedure (FTP), Unified Cycle (UC), and US06 high speed drive cycles were used for the hot running E85 VOC exhaust (Haskew and Liberty, 2011). The FTP phase 1 profile was applied for the cold-start E85 VOC emissions (Haskew and Liberty, 2011). E85 PM size distributions are summarized in Table S15 (Szybist et al., 2011), while PM composition information is summarized in Table S16 (Ferreira da Silva et al., 2010; Hays et al., 2013). Figure 5 illustrates the size and composition distribution of particulate matter emitted from catalyst-equipped gasoline vehicles and catalyst-equipped vehicles fueled by 85% ethanol and 15% gasoline (E85) as an example.

Figure 5: Particle emissions size and composition distribution for catalyst equipped gasoline vehicles (left panel) and catalyst equipped ethanol (E85) vehicles (right panel).

Aftertreatment devices were found to be more influential on biofuel exhaust rates (Alleman et al., 2005; Alleman et al., 2004; Frank et al., 2007; Hasegawa et al., 2007; Rounce et al., 2012; Tsujimura et al., 2007) than changes to fuel properties and feedstock origin (Durbin et al., 2007; Graboski et al., 2003). Diesel particulate filters (DPF), exhaust gas recirculation (EGR), selective catalytic reduction (SCR), and oxidation catalyst (OC) were assumed to be deployed on diesel and biodiesel powered vehicles by 2050. PM size distributions for DPF-equipped vehicles were obtained from (Rounce et al., 2012) (Table S15), and trace element, carbonaceous and inorganic ion fractions of PM distributions were obtained from (Cheung et al., 2010; Cheung et al., 2009) (see Table S16). Gas-phase VOC emissions profiles for biodiesel were not updated from fossil diesel profiles in the current study, but this change will be considered in future work.

The CNG VOC profile and PM size distribution was constructed based on (Gautam, 2011) (Tables S14 and S15). PM emissions of carbonaceous compounds, metals, and ions were measured from CNG vehicles running on the UDDS driving cycle (Yoon et al., 2014) (see Table S16). Figure 6 illustrates the size and composition distribution of particulate matter emitted from diesel vehicles, bio-diesel vehicles equipped with a diesel particle filter and exhaust gas recirculation, and catalyst-equipped CNG vehicles.

Figure 6: Particle emissions size and composition distribution for diesel vehicles (left panel), bio-diesel vehicles (center panel), and CNG catalyst equipped vehicles (right panel).

All fully electric vehicles, such as battery electric vehicles (BEVs) and H2 fuel cell vehicles, were assumed to have zero tailpipe exhaust and evaporative emission rates. Brake wear emission rates were reduced by 59% (Antanaitis, 2010) for all partial or fully electric vehicles equipped with regenerative breaking, such as hybrid, electric battery or fuel cell vehicles. Tire wear emissions were assumed to be independent of fuel or technology type.

2.3 CA-REMARQUE Aviation, Rail, and Off-Road Algorithms

Aviation sources include commercial, civil, agricultural, or military use and primarily run on jet fuel or aviation gasoline. The rail emission sources include passenger, commuter, switching and hauling trains which currently run primarily on diesel fueled generators powering an electric drivetrain. Off-road equipment includes industrial, agricultural, and construction equipment, port and rail operations, as well as lawn and garden equipment. The list of aviation, rail, and off-road emission source categorizations are based on the EICs listed in Table S17 (including new EICs created to represent sources operating on alternative fuels previously not in the CARB inventory).

2.3.1 VISION Model

Future 2050 emissions for aviation, rail, and off-road equipment were assumed to follow the 2010 versus 2050 growth projected by the CARB VISION model (California Air Resources Board, 2012a), an off-road expansion of Argonne's on-road VISION model (Argonne National Laboratory Transportation Technology R&D Center, 2012). CARB's off-road VISION model uses historical trends to project to the year 2050 while incorporating some future standards for criteria pollutant emission rates. These include the implementation of Tier 4 130-560 kW compression-ignition diesel engine emission standards for PM, CO, and NMHC+NOx (California Air Resources Board, 2010a) leading to 90% reduction in PM emissions rates and an 85% reduction in NMHC and NOx emissions rates.

Aviation, rail, and off-road 2010 emissions at 4 km resolution ($em^{2010}_{cell,I}$) were scaled to produce an "intermediate" estimate prior to CA-TIMES adjustments using Eq. (5).

$$\mathrm{em}^{2050}_{\mathrm{cell,i,intermediate}} = \underbrace{\left(\frac{\mathrm{em}_{\mathrm{i}}^{2050}}{\mathrm{em}_{\mathrm{i}}^{2010}}\right)}_{\substack{\text{State-wide}\\ \text{emission growth}\\ \text{scaling from 2010}\\ \text{to 2050}}} \cdot \mathrm{em}^{2010}_{\mathrm{cell,i}} \tag{5}$$

where

$\mathrm{em}^{2050}_{\mathrm{cell,i,intermediate}}$ = intermediate grid cell 2050 emissions for a transport source (aviation, rail, off-road) consuming a reference or conventional fuel or energy [kg hr^{-1}]

$\mathrm{em}_{\mathrm{i}}^{2050}$ = state-wide 2050 emissions of a transport source [kg hr^{-1} or tons day^{-1}]

$\mathrm{em}_{\mathrm{i}}^{2010}$ = state-wide 2010 emissions of a transport source [kg hr^{-1} or tons day^{-1}]

$\mathrm{em}^{2010}_{\mathrm{cell,i}}$ = grid cell 2010 emissions of a transport source [kg hr^{-1}]

2.3.2 CA-TIMES Modification of Intermediate 2050 Off-Road Mobile Emissions

The portion of energy consumed for each fuel (Ei,f/Σf Ei,f) as projected by CA−TIMES was applied to the intermediate 2050 emissions inventory for each transport mode (f) and source type (i) using Eq. (6). The consumption of different fuels relative to total fuel consumption for a given mode is shown in Fig. S1-S3 for rail, off−road, and aviation modes respectively. Alternative to conventional scaling factors were applied to account for adoption of alternative fuels as summarized in Table 2. Eq. (6) also includes an after treatment or control device factor (1−η) where appropriate.

$$\mathrm{SFi,f} = \underbrace{\left(\frac{\mathrm{E_{i,f}}}{\sum_{\mathrm{f}}\mathrm{E_{i,f}}}\right)}_{\substack{\text{Portion of}\\ \text{alternative}\\ \text{fuel energy}\\ \text{consumption}}} \cdot \underbrace{\left(\frac{\mathrm{em_{i,f}}^{2050}}{\mathrm{em_{i,intermediate}}^{2050}}\right)}_{\substack{\text{Alternative}\\ \text{fuel}\\ \text{emission}\\ \text{scaling}\\ \text{relative to}\\ \text{conventional}}} \cdot \underbrace{(1-\eta_{\mathrm{i}})}_{\substack{\text{Fraction of}\\ \text{pollutant not}\\ \text{removed by}\\ \text{aftertreatment}\\ \text{device}}} \tag{6}$$

where

$\mathrm{SF_{i,f}}$ = emission scaling factor for a given new/alternative or non-conventional/non-reference fuel for a transport source [dimensonless]

$\mathrm{E_{i,f}}$= new/alternative fuel/energy consumed by a transport source (e.g. biodiesel for commuter rail) [PJ]

$\sum_{\mathrm{f}}\mathrm{E_{i,f}}$= total fuel/energy consumed by a transport source (e.g. biodiesel + diesel for commuter rail) [PJ]

$em_{i,f}^{2050}$ = state-wide 2050 emissions of a transport source consuming a new/alternative fuel [kg hr^{-1} or tons day^{-1}]

$em_{i,intermediate}^{2050}$= state-wide 2050 intermediate emissions of a transport source consuming a new/alternative fuel. [kg hr^{-1} or tons day^{-1}]

η_i = efficiency of removal from a control or aftertreatment device [fraction from 0.00-1.00]

Table 2: Emission rate changes for alternative fuels in off-road vehicles.

Transport Mode	Alternative Fuel	Reference Conventional Fuel	Pollutant	Alt/ Conv Ratio	Conv % Change	Citations
Rail	Biodiesel	Diesel	CO	0.655	-34.5%	Osborne et al. (2010)
			NOx	1.13	13%	Osborne et al. (2010)
			SOx	0.0005	-99.95%	Assumed (see text)
			ROG	0.775	-22.5%	Osborne et al. (2010)
			PM	0.805	-19.5%	Osborne et al. (2010)
Off-road/ Agricultural	Biodiesel	Diesel	CO	1	0%	Durbin et al. (2007)
			NOx	1.08	8%	Durbin et al. (2007)
			SOx	1	0%	Durbin et al. (2007)
			ROG	0.39	-61%	Assumed (see text)
			PM	1.13	13%	Durbin et al. (2007)
	Compressed natural gas	Diesel	CO	0.668	-33.2%	Cooper et al. (2012)
			NOx	0.189	-81.1%	Cooper et al. (2012)
			SOx	1	0%	Assumed (see text)
			ROG	2.349	134.9%	Cooper et al. (2012)
			PM	0.0782	-92.18%	Cooper et al. (2012)
Aviation	Biomass-based kerosene jet fuel	Kerosene jet fuel	CO	1	0%	Lobo et al. (2012)
			NOx	1	0%	Lobo et al. (2012)
			SOx	0.007	-99.3%	Assumed (see text)
			ROG	0.605	-39.5%	Lobo et al. (2012)
			PM	0.38	-62%	Lobo et al. (2011)

The final emissions for each specific offroad source consuming each specific fuel in 2050 ($em_{cell,i,f}^{2050}$) are then calculated by combining the effects of the VISION and CA-TIMES updates as shown in Eq. (7).

$$em_{cell,i,f}^{2050} = SF_{i,f} \cdot em_{cell,i,intermediate}^{2050} \qquad (7)$$

Aviation biomass-based kerosene jet fuel (KJF) emissions changes are based on Fischer-Tropsh gas-to-liquid (FT GTL) biofuel aviation emissions tests (Lobo et al., 2011; Lobo et al., 2012). These studies found minor changes to CO and NO_x emissions due to the adoption of biofuels. SO_x reduction was assumed proportional to the fuel sulfur content (Lobo et al., 2012) leading to reductions of 99% as shown in Table 2.

Off-road equipment (other than trains) operating on biodiesel instead of Ultra low-sulfur diesel (ULSD) was assumed to emit HC and NO_x with scaling factors (relative to conventional diesel emissions) of 0.39 and 1.08, respectively

(Durbin et al., 2007). No significant changes in CO, SO_x and PM due to the adoption of biodiesel versus ULSD were identified in the literature and so these emissions were assumed to remain at levels estimated for conventional diesel engines. This approach inherently assumes that the sulfur content of biodiesel will not exceed the current limit of 15 ppm for ULSD. Off-road or agricultural emission changes from switching from diesel to CNG are also found to have large reductions in most pollutants except reactive organic gases (ROGs) (Cooper et al., 2012).

Military aviation emissions were held constant at 2010 levels in the current study due to an assumption of continued exemptions for military activity.

2.3.3 Off-Road Mobile PM and Gas Speciation and Size Profile Changes

PM mass size distributions for E85, biodiesel, and CNG are assumed to be similar for off-road and on-road vehicles (Table S15). The new PM mass size distribution for biomass-based KJF is shown in Table S18 (Lobo et al., 2011). Figure 7 illustrates the size and composition distribution of particulate matter emitted from conventional jet-fuel aircraft and biomass-based kerosene jet fuel aircraft. The conventional profile is based on old source profile measurements that assumed uniform distribution of particles between diameters 0.1-1.0 µm. This conventional profile will be updated with more recent literature values in future work.

Figure 7: Particle emissions size and composition distribution for jet-fueled aircraft (left panel) and biomass-based kerosene jet-fueled aircraft (right panel).

2.4 CA-REMARQUE Marine Algorithms

The marine emission source category includes all ocean going vessels (OGV), commercial harbor craft (CHC), and recreational boats (see Table S19). An intermediate OGV emissions inventory was predicted for the year 2050 based on the extrapolation of Port of Los Angeles and Port of Long Beach 2020 trends (Starcrest Consulting Group, 2009; The Port of Los Angeles and The Port of Long Beach, 2010) (see Table S20). All other OGV emissions (not listed in Table S20) in California were held constant at 2010 levels in the intermediate 2050 inventory prior to modifications from CA-TIMES.

2.4.1 CA-TIMES Modification of Intermediate 2050 Marine Emissions

The fuels used to power OGVs were modified based on predictions from the CA-TIMES scenarios. It should be noted that the CA-TIMES model reports worldwide marine energy consumption. In the current study, it was assumed that marine vessels operating near the California coast would consume the global average mix of biofuels produced by

CA-TIMES. For example, if CA-TIMES indicates that a third of the residual fuel oil (RFO) (also call heavy fuel oil) consumed globally by marine vessels would be converted to biomass-based residual fuel oil (BRFO), then a third of the RFO marine vessel emissions near California boundaries were also converted to BRFO. As indicated by Fig. S4, CA-TIMES finds other approaches besides biofuel adoption for ships are more cost-effective for meeting the GHG target in 2050. CA-TIMES determined that it will be more economical to substitute some RFO with a lighter petroleum (diesel) to decrease carbon intensity rather than using biomass-based RFO.

Alternative fuels used in marine sources will modify criteria pollutant emissions. Biomass-based alternatives for marine residual fuel oil (RFO) were estimated to be similar to the average of B100 from palm oil, animal fat, soybean oil, and sunflower oil operating at 75% load (Petzold et al., 2011). NO_x was the only regulated pollutant observed to remain constant during emissions testing. Emissions of all other pollutants decreased as summarized in Table 3.

Table 3: Emission rate changes from ships changing from conventional fuels to biofuels.

Alternative Fuel	Reference Conventional Fuel	Pollutant	Alt/ Conv Ratio	Conv % Change	Citations
biomass-based residual fuel oil (RFO)	residual fuel oil (RFO)	CO	0.697	-30.3%	(Petzold et al., 2011)
		NOx	1	0%	(Petzold et al., 2011)
		SOx	0.012	-98.8%	(Petzold et al., 2011)
		ROG	0.413	-58.7%	(Petzold et al., 2011)
		PM	0.223	-77.7%	(Petzold et al., 2011)
Biodiesel (BDL)	Diesel (DSL)	CO	0.921	-7.9%	(Jayaram et al., 2011)
		NOx	1	0%	(Jayaram et al., 2011)
		SOx	0.0003	-99.97%	Assumed (see text).
		ROG	1	0%	(Jayaram et al., 2011)
		PM	0.684	-31.6%	(Jayaram et al., 2011)

Assuming biodiesel (BDL) and biomass based residual fuel oil (BRFO) has about 1 ppm sulfur content, and that by 2010 the sulfur content regulations ensured that marine diesel oil (MDO) and RFO had 1.5 ppm and 2.5 ppm S, respectively, then the switch to biofuels would reduce SO_x emissions by 33.3% (relative to conventional MDO) and 60% (relative to conventional RFO). Additional reductions in CO, TOG, and PM were also projected based on (Jayaram et al., 2011; Petzold et al., 2011) as summarized in Table 3.

Several international and California shoreline regulations were applied to marine emissions in the year 2050 as summarized in Table S21 and Table S22. At-berth or hotelling container, passenger (cruise), and refrigeration OGVs will use shoreline power instead of auxiliary engines for 80% of their berthing hours by 2020, (California Air Resources Board, 2007). It was also assumed that MDO or marine gasoline oil (MGO) used within 24 nautical miles of the California shore will have sulfur content of <0.1% by 2050 (California Air Resources Board, 2011e). Further offshore, all marine fuels used within 100 nautical miles of North America were assumed to have sulfur content < 1% after the year 2012 (leading to reductions shown in Table 3).

2.4.2 Marine PM and Gas Speciation and Size Profile Changes

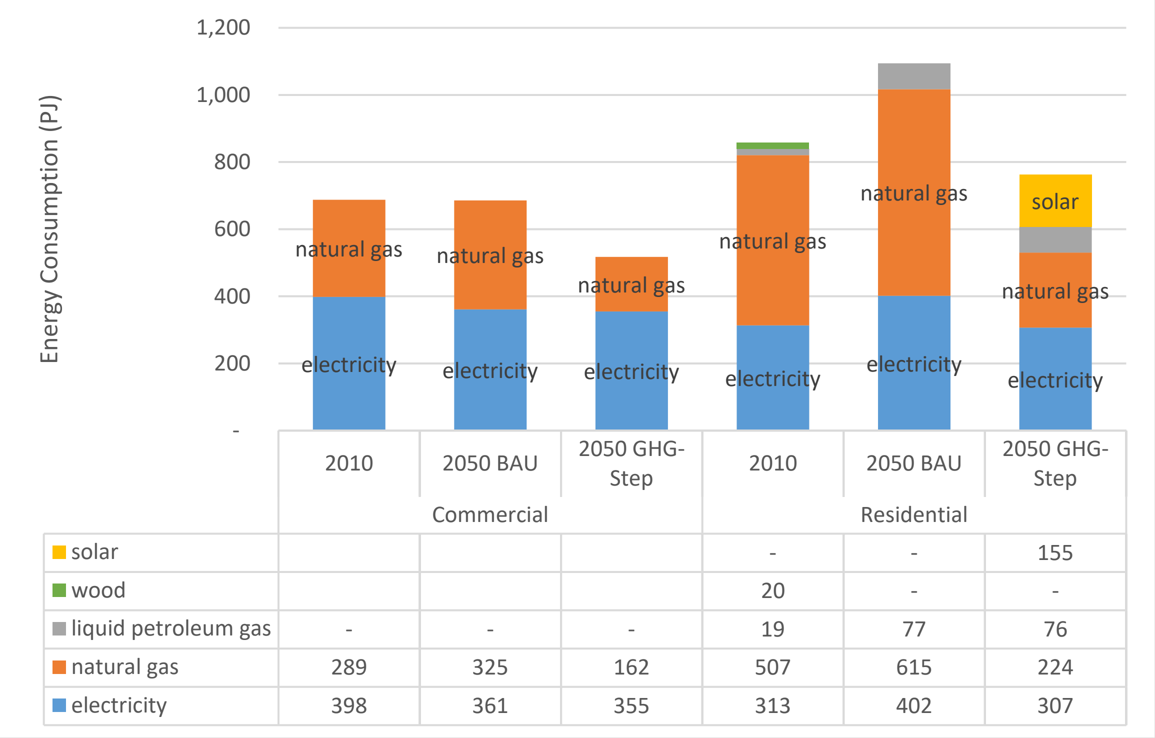
PM size distribution changes caused by the switch to alternative marine fuels were based on (Jayaram et al., 2011) (see Table S23). The size and composition distribution profiles used to represent marine emission associated with different fuels are displayed in Fig. 8.

Figure 8: Particle emissions size and composition distribution for ships powered by marine residual oil (left panel), marine bio-diesel (center panel), and biomass-based residual fuel oil (right panel).

2.5 CA-REMARQUE Residential and Commercial Algorithms

Major emissions sources within the residential and commercial sectors include natural gas combustion (space heating and water heating), biomass combustion (fireplaces and stoves), and food cooking (especially charbroiling and frying). The residential and commercial emissions associated with natural gas and food cooking were assumed to scale according to population growth projected for each county (Table S24) (State of California, 2013) to produce an intermediate emissions inventory. These intermediate residential and commercial gridded emissions were then scaled to reflect 2010 versus 2050 results from CA-TIMES (Fig. 9).

Natural gas consumption in the commercial sector reduced by half (325 PJ to 162 PJ) in the GHG-Step scenario relative to the BAU scenario in 2050. Most of commercial energy reduction is due to efficiency gains and switch from natural gas to electrification of end uses. Natural gas consumption in the residential sector also decreases (615 PJ to 507 PJ) under the GHG-Step scenario relative to the BAU scenario. Much of the energy that would have been supplied by natural gas is replaced by renewable sources such as solar (155 PJ) which was assumed to have no criteria pollutant emissions in California. Improved energy efficiency and conservation also plays a role, with residential electricity consumption decreasing (402 PJ to 313 PJ) in the GHG-Step scenario. Other combustion sources, including wood burning and distillate oil fuel consumption, were allowed to compete in CA-TIMES subject to the constraint that they could not increase above the 2010 levels in order to maintain compliance with current air quality regulations.



	Commercial			Residential		
	2010	2050 BAU	2050 GHG-Step	2010	2050 BAU	2050 GHG-Step
🟨 solar				-	-	155
🟩 wood				20	-	-
◼ liquid petroleum gas	-	-	-	19	77	76
🟧 natural gas	289	325	162	507	615	224
🟦 electricity	398	361	355	313	402	307

Figure 9: CA-TIMES energy consumption by energy resource and scenario for commercial and residential.

2.6 CA-REMARQUE Electricity Generation Algorithms

The electricity generation emissions category includes all fuel-burning and renewable power plants for industrial, residential, or commercial use. Annual generation totals for different types of California power plants were extracted from national power plant data (US Energy Information Administration Independent Statistics and Analysis, 2012; US Environmental Protection Agency, 2014). Emissions rates per unit of fuel burned were estimated for each power plant described in the basecase 2010 emissions inventory.

CA-TIMES finds that non-hydro renewable (geothermal, tidal, solar, wind, and biomass) increases from 10% (22,938 GWh) of the electricity generation mix in 2010 (144,825 GWh) to 35% and 76% (489,493GWh) in the 2050 BAU and 2050 GHG-Step scenario, respectively (see Fig. 10). However, total in-state and out-of-state electricity generation in the GHG-Step scenario is 1/3rd larger than the BAU scenario (416,219 GWh versus 643,373 GWh) to meet the increased demand from sectors such as the on-road vehicles with growing hybridization and electrification needed to meet the 2050 carbon constraint. Statewide scaling factors for electricity generation in the 2050 BAU scenario versus 2010 and the 2050 GHG-Step scenario versus 2010 are listed in Table S25.

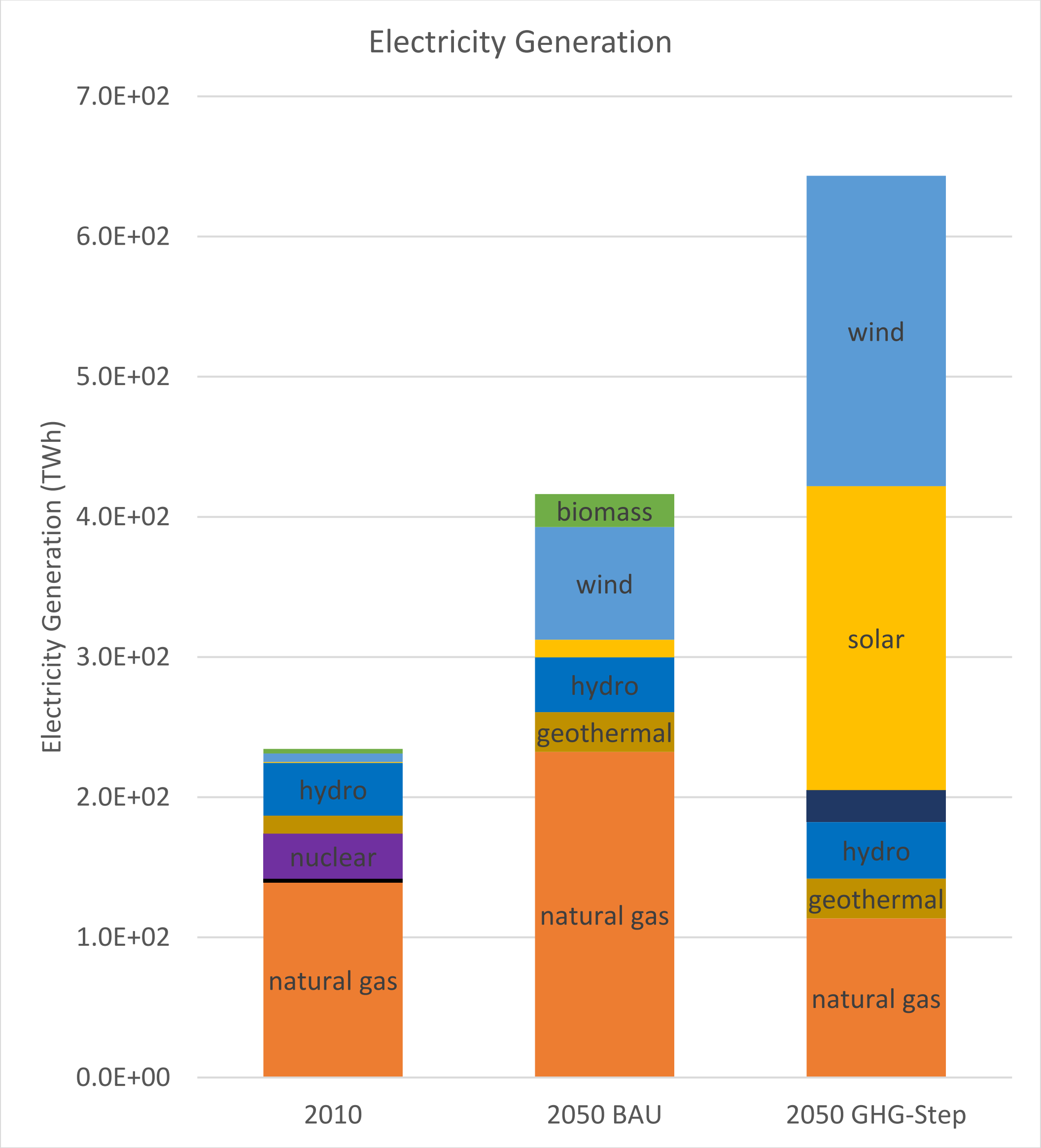


Figure 10: CA-TIMES electricity generation resource mix by scenario.

CA-TIMES calculates aggregated state-wide energy totals but energy resources (especially for renewables) are not uniformly distributed across the state. In the current study, renewable electricity production in 2050 was spatially allocated in a manner that was consistent with the energy resource potential in 12 regions (Fig. S5) as projected in 15 scenarios by the grid load distribution model SWITCH (Fripp, 2012; Johnston et al., 2013; Nelson et al., 2013). Table S26 lists the electrical generation by energy source for each SWITCH region averaged across these 15 scenarios. This profile of resource potential was then applied to the CA-TIMES predictions summarized in Table S25 yielding the 2050/2010 scaling factors for the BAU scenario (Table S27) and the GHG-Step scenario (Table S28).

The scaling factors summarized in Tables S27 and S28 assume that the out-of-state portion of electricity generation for a given fuel or energy resource in the year 2050 remained constant at 2010 levels. CA-TIMES does not provide additional information describing out-of-state generation except for a few renewables. This out-of-state portion of the electricity generation was subtracted from the CA-TIMES totals prior to scaling emissions from each power plant in California. Table S29 summarizes the out-of-state portion of electricity generation for each fuel in 2010 and assumed portions in each of the 2050 scenarios.

Additional emissions adjustments were made for new renewable fuels such as those produced by the Biomass Integrated Gasification Combined Cycle (IGCC), a process that gasifies biomass for electricity production. Much of the biomass electricity generation projected by CA-TIMES for 2050 in the BAU scenario uses biomass IGCC (see Tables S30 through S32). There are currently several coal IGCC plants in the US (U. S. Department of Energy National Energy Technology Laboratory, 2010, 2015) but no biomass IGCC plants (Lundqvist, 1993; Ståhl and Neergaard, 1998; U. S. Department of Energy National Energy Technology Laboratory, 2010). Future biomass IGCC emissions in California were estimated using several models that incorporate biomass IGCC, such as GREET, CA-GREET (Argonne National Laboratory Transportation Technology R&D Center, 2014; California Air Resources Board, 2009a, 2015), and an NREL analysis (Mann and Spath, 1997). Ultimately, biomass IGCC power plant emissions were estimated from conversion of conventional steam turbines in the 2010 ARB inventory based on emissions rates inferred from CA-GREET1.8 for 2050 (Table S33). An inter-comparison study between GREET1.8, GREET 2014, and CA-GREET2.0 showed that theCA-GREET1.8b model had the best agreement with emissions rates from approximately 30 biomass plants operating on wood residue in California.(California Air Resources Board, 2011b; US Environmental Protection Agency, 2014).

2.7 CA-REMARQUE Industrial and Agricultural Algorithms

The industrial and agricultural emissions category covers many manufacturing industries such as metal, wood, glass, textile, mining, and chemical. Food and agricultural sectors include farming livestock, crops, food production, bakeries, and breweries. Most of these industries were unchanged in the CA-TIMES energy scenarios, with the notable exception that biofuel and hydrogen fuel production replaced some traditional petroleum production, causing changes in refinery and storage emissions (shown in Figs. S6 to S8).

2.7.1 Fossil and Renewable Fuel Production

All fossil petroleum refining and storage emissions in the 2010 ARB emissions inventory were scaled according to the amount of oil production and refining that was required in California for each 2050 CA-TIMES scenario (see Fig. S6). Scaling factors were applied uniformly to all emission processes including seepage, evaporative or fugitive, and other processes. Fossil petroleum consumption generally decreased in future scenarios, but was not eliminated. As discussed in previous sections, transportation modes (e.g. marine, heavy duty trucks) still consume fossil fuel such as diesel, and the stationary sources (electricity generation, residential, and commercial) still consume natural gas. CA-TIMES determined that much of the extracted petroleum used by refineries would be imported to the state rather than extracted locally. This can be seen by the reduction of crude oil supply in California from 1510 PJ in 2010, to 426.5 PJ in the 2050 BAU scenario and 0.0PJ in the GHG-Step scenario (see Fig S6). Refining is also are projected to decline slightly between 2010 and the 2050 scenarios, with reductions of 25% in the BAU scenario and 44% in the GHG-Step scenario. This suggests that it is more cost effective or less carbon intensive to import fuel than to extract oil and gas in or around California. The total (imported and in-state) oil supply also decreases in 2050, by -26% in the BAU (3200PJ) and -44% in the GHG-Step (2400PJ) relative to 2010 (4300PJ). This reflects the adoption of electrification and alternative fuels to replacing petroleum consumption in the presence of growing energy demand in 2050.

Hydrogen (H_2) production increased in both 2050 CA-TIMES scenario results, but the increases in the GHG-Step scenario are much larger (Fig. S7). It was assumed that new hydrogen production facilities would be located at current H_2 production facilities or existing refineries. Overall 32 new natural gas steam methane reforming (SMR) H_2 facilities and 15 new biomass gasification facilities were projected to meet the demand summarized in Fig. S7. In the current study, criteria pollutant emission rates from SMR H_2 production (summarized in Table 4) were calculated from the top 3 SMR H_2 production facilities (California Air Resources Board, 2010c, 2014). Few studies have been published describing criteria pollutant emissions from biomass gasification H_2 production and so emissions rates for this production pathway were obtained from the CA-GREET model (California Air Resources Board, 2015). Direct criteria pollutant emissions from hydrogen production using electrolysis were zero since this process uses electricity to split water molecules into H_2 and oxygen (emissions from these facilities appear under electricity generation).

Table 4: Pollutant emission rate associated with hydrogen production. Unis are grams of pollutant per mmBtu of hydrogen produced.

	SMR - average of top CA H2 SMR facilities	Gasification - CA-GREET2015 Gasification versus SMR Scaling	Electrolysis
CO	4.303	0.997	0
NOx	1.701	0.34	0
SOx	0.092	0.406	0
VOC	2.33	1.118	0
PM10	0.433	0.048	0

The CA-TIMES model determined that biofuel consumption and production will be high in California in the year 2050 (Fig. S8). Biofuel refineries for different feedstock classes (wood, municipal solid waste (MSW), herbaceous, yellow grease or tallow, or corn ethanol) (see Tables S34 and S35) were located using a spatial biomass optimization model which seeks to minimize cost within resource and regulatory constraints (Tittmann et al., 2010). Biofuel refineries were prohibited in NAAQS non-attainment areas, an added constraint based on the high feedstock case described by (Parker, 2012). Production rates at in-state biorefineries were scaled to match the in-state volumes produced in CA-TIMES for each type of biofuel. Out-of-state imports and refining were assumed for crops that could not be grown at a large enough scale to meet the demand in California, such as herbaceous crops and the bulk of corn-ethanol (see Tables S34 and S35). Emissions for each biofuel refinery were estimated using CA-GREET1.8b emission rates per unit of fuel produced.

2.7.2 Biogas Capture and Use

CA-TIMES assumes that landfill gas reduces over time due to better management of organic matter in landfills, and the consumption of existing landfill stock material over many decades. All biogas in CA-TIMES is converted to biomethane through removal of CO_2 and impurities, and further blended with natural gas so that it is undistinguishable from extracted fossil natural gas.

Dairy biogas is a significant renewable energy source in CA-TIMES. California produced a fifth of the milk in the US in 2010 (California Department of Food and Agriculture, 2011) and an exponential regression using 2001–2013 CFDA data estimates the number of dairy cows in California may increase by a factor of 1.5 by the year 2050. Methane emission rates were estimated from GHG inventory Documentation (California Air Resouces Board, 2014) for each manure management practice: liquid/slurry, anaerobic lagoon, anaerobic digester, daily spread, deep pit, pasture, and solid storage. The increase in the cow population was assumed to occur uniformly across all management practices except for the systems used in biogas capture. These systems, including anaerobic digester, anaerobic lagoon, and liquid/slurry management practices, were adjusted to meet the quantities of biogas specified by each CA-TIMES scenario. The amount of waste produced by each dairy cow each year was used to estimate the annual biomethane production and energy potential of each animal. The electricity potential from biomethane is then calculated using AgSTAR conversion rates (Environmental Protection Agency, 2010; U.S. Environmental Protection Agency AgSTAR Program, 2011). The overall fugitive VOC emissions from animal waste declines in the biogas production scenarios since a large fraction of the waste is treated. Overall, fugitive dairy manure VOC emissions increased by 50% due to cow population growth in the BAU scenario, and decreased by a factor of a 33% for the GHG-Step scenario relative to 2010.

Future biomethane production sites were selected based on recommendations from the USDA's Cooperative Approaches for Implementation of Dairy Manure Digesters (U.S. Department of Agriculture Rural Development Agency, 2009). Mainly, locations were selected with nearby pipeline networks (Gilbreath et al., 2014) to transport raw biogas to a centralized clean-up facility, where it can then be compressed and sold for use by electric generation power plants or transportation fuels. This was considered a more viable option as natural gas pipeline infrastructure is easy to access, demand from electric utilities for biomethane is high to meet the renewable portfolio standard (RPS), and a centralized clean-up facility is more economical than distributed facilities.

3 Results and Discussion

3.1 On-Road Mobile Emissions

Figure 11 illustrates particulate matter emissions of tire and brake wear from on-road vehicles under the BAU and GHG-Step scenarios. The fine spatial distribution of the emissions reflects the spatial distribution of tire and brake wear emissions in the base 2010 inventory that is updated using EMFAC predictions to produce the intermediate 2050 emissions inventory. The technology changes inherent in the CA-TIMES BAU and GHG-Step scenarios are then applied uniformly across the state yielding virtually identical spatial distributions for the final 2050 BAU and

GHG-Step scenario emissions. Tire and brake wear emissions patterns illustrated in Figure 11 essentially follow predicted vehicle activity patterns in the state. Predicted emissions are highest in major urban centers and along major transportation corridors. Although increase in vehicular activity was part of this study, expansion of roadways between 2010 and 2050 were not considered in this study and may be updated in newer versions of the model.

California's environmental regulations apply uniformly across the state, which supports the assumption of uniform GHG emissions reductions for on-road vehicles. Despite the uniform regulatory landscape, some of the measures described in the CA-TIMES GHG-Step scenario rely on modified behavioral patterns and willingness or ability to adopt new technologies, which may change by region. Education levels, personal wealth, and environmental attitudes vary sharply across California. Capturing these trends in sub-regions of the state will require surveys of consumer choice and predictions of future behavior that are beyond the scope of the current manuscript.

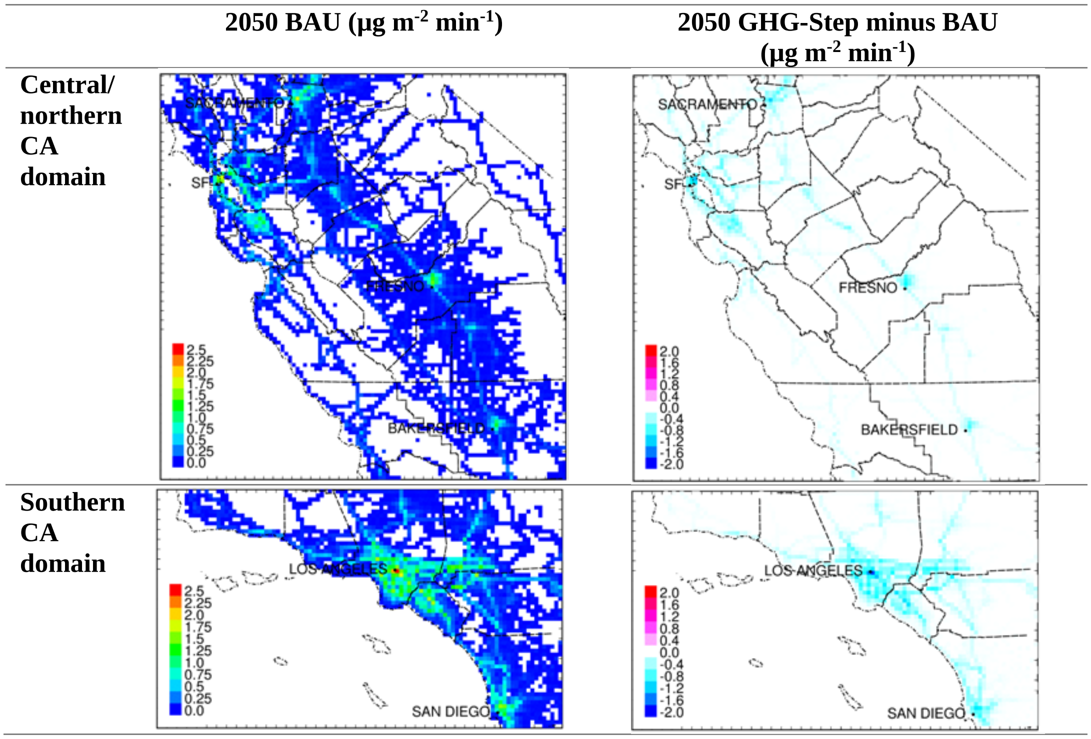


Figure 11: Particulate matter emissions from vehicle tire and break wear in the BAU scenario (left panels) and emissions change in the GHG-Step scenario (right panels). Units are μg m^{-2} min^{-1}.

Figure 12 illustrates the particulate matter emissions from tailpipe exhaust under the 2050 BAU scenario and the 2050 GHG-Step scenario. Similar to the tire and brake wear emissions, the spatial pattern for mobile sources is identical under both scenarios because the technology changes specified by the CA-TIMES model are applied uniformly over the entire state. Tailpipe particulate matter emissions once again follow patterns of vehicle activity as predicted by EMFAC. Of greater interest is the prediction that tire and brake wear emissions (Fig. 11) will exceed tailpipe emissions (Fig. 12) in both the 2050 BAU and GHG-Step scenarios due to the adoption of increasingly clean vehicle technology. Tailpipe emissions in the GHG-Step scenario are a factor of ~1.8 lower than tailpipe emissions in the BAU scenario. In contrast, tire and brake wear emissions are predicted to decrease by a factor of +3 under the GHG-Step scenario. This reflects the fact that BAU gasoline and diesel tailpipe emissions already incorporate significant emissions control technology yielding fewer opportunities for further improvement. Tire and brake wear emissions have almost no control technology in the BAU scenario, which makes the widespread adoption of electric or hybrid drivetrains using regenerative braking particularly effective at reducing emissions.

The current analysis assumes that no new major highways will be built in California and population growth is accommodated partially through increased urban density such that traffic volumes increase uniformly across the transportation network. These assumptions are simplistic but a previous study of smartgrowth in the San Joaquin Valley indicated that more detailed accounting of population growth had minimal impact on air quality (Hixson et al., 2010).

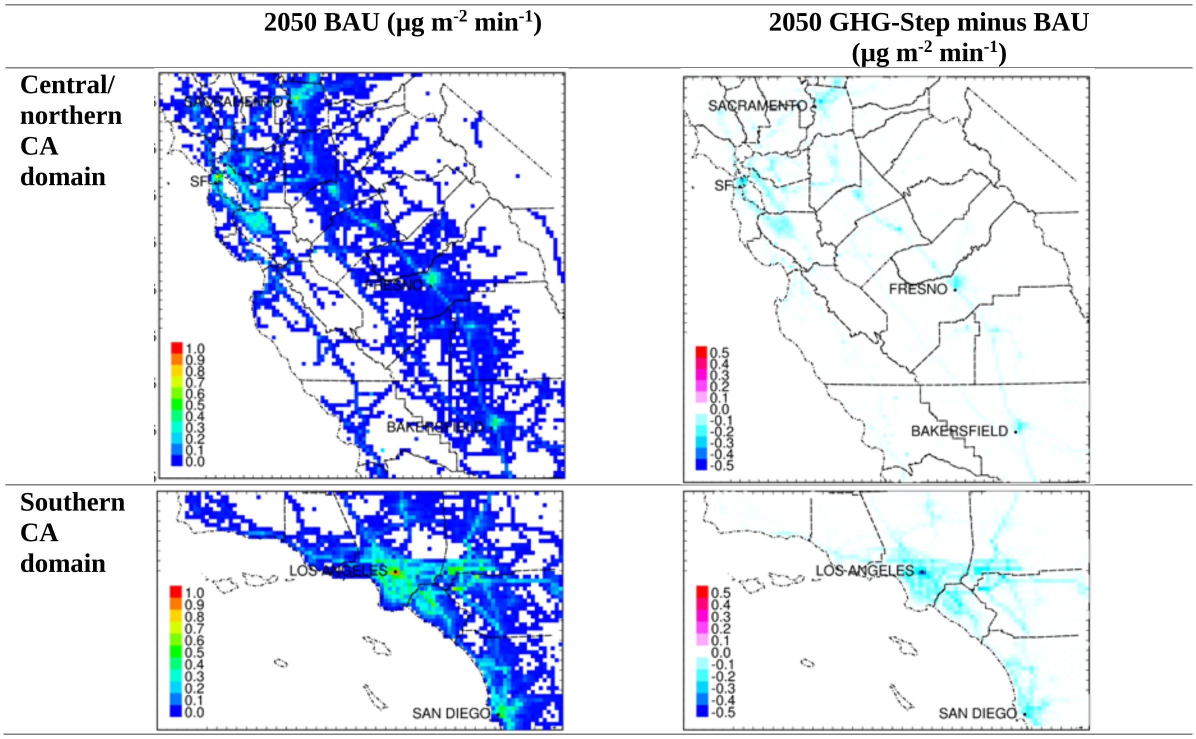


Figure 12: Particulate matter emissions of vehicle tailpipe exhaust in the BAU scenario (left panels) and emissions change in the GHG-Step scenario (right panels). Units are µg m^{-2} min^{-1}.

3.2 Rail, and Off-Road Emissions

Particulate matter emissions from off-road and rail sources are plotted in Fig. 13 for the BAU and GHG-Step scenarios examined in the current study. Maximum statewide particulate matter emissions for this source category are centered at the location of major construction projects with lower emissions rates for "routine" off-road emissions distributed more broadly according to typical activity patterns for smaller construction projects, rail, etc. The 2010 emissions inventory that acts as the basis for the 2050 projections in the current project correctly identified replacement of the east span of the Bay Bridge in the San Francisco Bay Area as the leading construction project with the highest overall emissions in the state. This ~$6.5B project spanned more than 10 years with the new bridge completed in 2013 and final decommissioning and demolition of the old eastern span scheduled for 2018.

It is difficult to predict the location of major construction projects in 2050 but it is reasonable to expect that several large projects will be active in that timeframe. Candidate projects currently under discussion include additional replacement of California's numerous highways and bridges, upgrading California's water conveyance systems to better withstand earthquakes, development of high speed rail lines, reinforcement or expansion of seawalls to protect property, etc. Each of these projects will potentially emit criteria pollutants that would affect air quality over major urban centers. In the present study, the peak emissions associated with the major construction project around the Bay Bridge were retained in the future scenario as an example of a major construction project near an urban area. Future model analysis that uses these emissions should conduct sensitivity tests to ensure that the assumed placement of this example major construction project does not influence the overall conclusions of the study.

Maximum particulate matter emissions shown in Fig. 13 decrease by a factor of approximately 1.6 4in the GHG-Step scenario relative to the BAU scenario. Adoption of biomass based fuels was also found to reduce emissions of SO_x, HC, PM, and occasionally CO from off-road and rail sources, but NO_x emissions increased for some fuel choices.

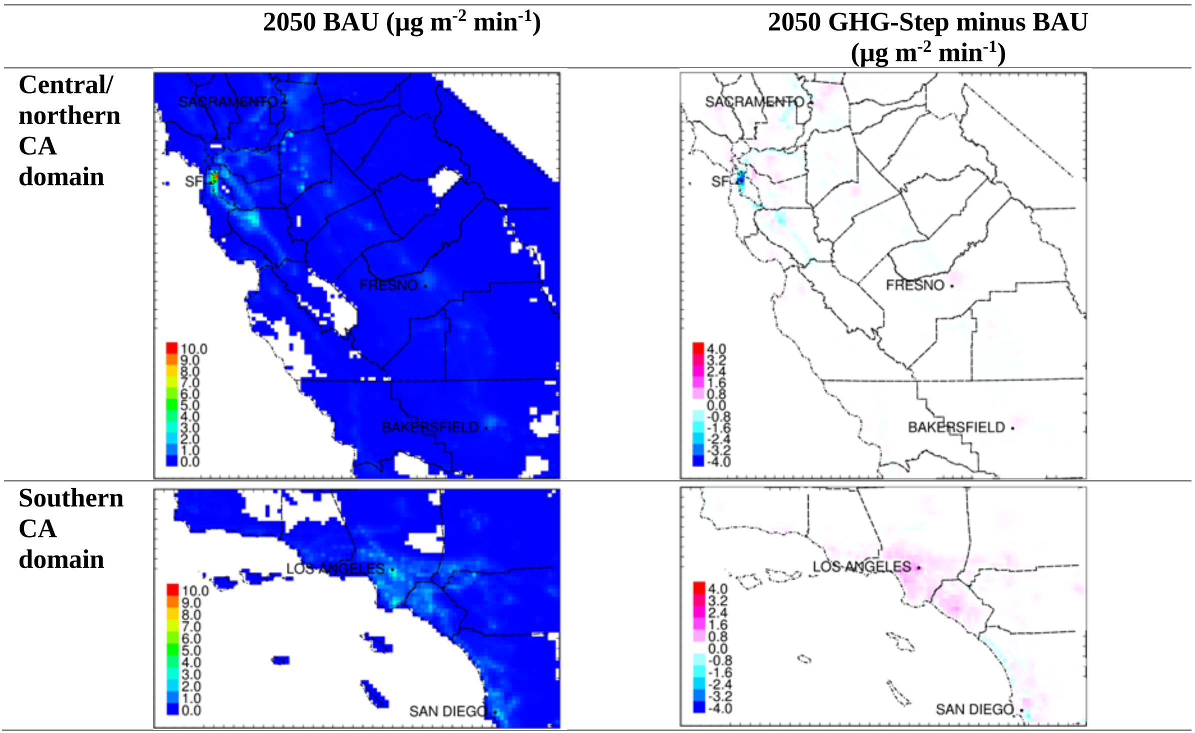


Figure 13: Particulate matter emissions from rail and other off-road sources in the BAU scenario (left panels) and emissions change in the GHG-Step scenario (right panels). Units are µg m^{-2} min^{-1}.

3.3 Marine and Aviation Emissions

Particulate matter emissions from marine and aviation sources are shown in Fig. 14 for the BAU and GHG-Step scenarios considered in the present study. The highest particulate matter emissions rates occur in off-shore shipping lanes that converge on the Port of Los Angeles, the Port of Long Beach, and the Port of Oakland. Emissions rates change with proximity to California shores due to regulations governing sulfur content of marine fuel or ship speed. Emissions patterns at inland locations reflect shipping activity on inland waterways or activity surrounding small regional airports.

Maximum particulate matter emissions rates from marine sources increase under the GHG-Step scenario as illustrated most clearly in the right panels of Fig 14. CA-TIMES determined that the available biofuel capacity could be more efficiently used to offset traditional fossil fuels for on-road transportation sources and so the GHG-Step scenario is predicted to incorporate additional fossil fuels for marine sources under the GHG-Step scenario versus the BAU scenario. The net result of the disbenefits associated with increased marine emissions versus the benefits of the decreased on-road emissions will be considered in future studies that include analysis with regional air quality models.

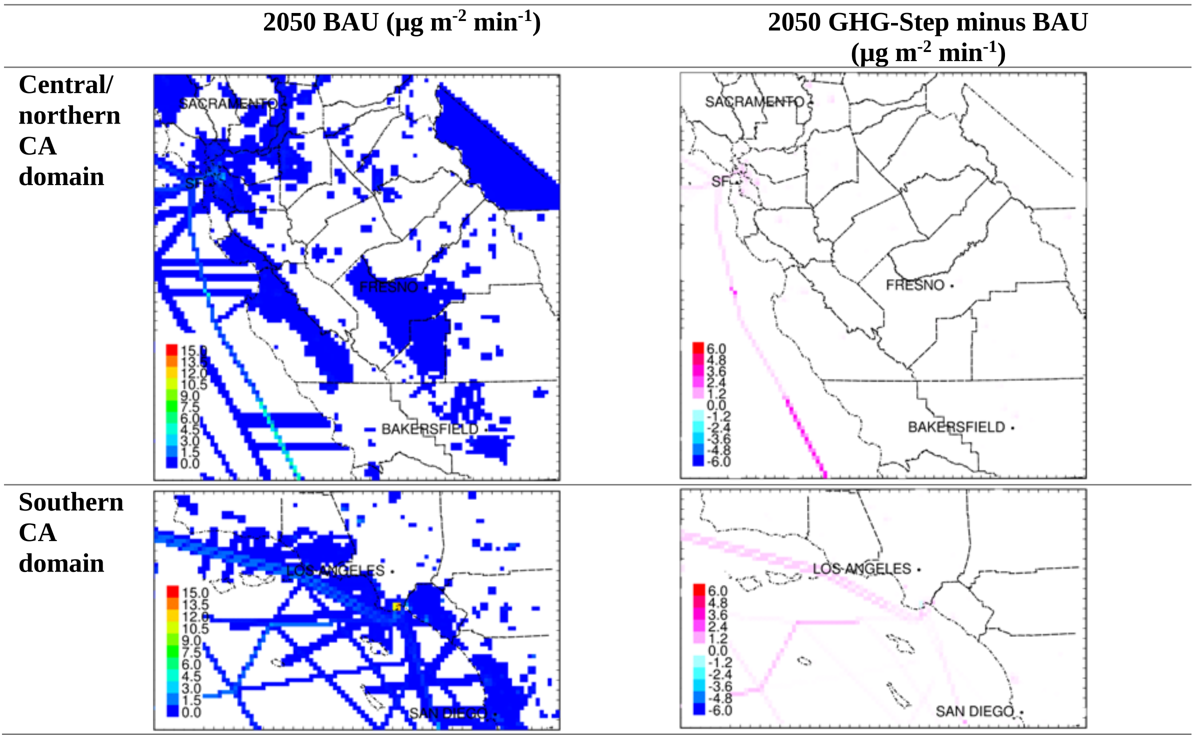


Figure 14: Particulate matter emissions from marine and aviation sources in the BAU scenario (left panels) and emissions change in the GHG-Step scenario (right panels). Units are μg m^{-2} min^{-1}.

3.3 Residential and Commercial Emissions

Fig. 15 illustrates particulate matter emissions from residential and commercial sources under the 2050 BAU and GHG-Step scenarios. The spatial patterns of emissions largely follow the estimated population projections in

California in the year 2050 as summarized in Table S24. Population growth was assumed to be identical under the BAU and GHG-Step scenarios yielding virtually identical spatial distributions for both scenarios. The adoption of new technologies and altered behavioral patterns predicted by the CA-TIMES model under the GHG-Step scenario were applied uniformly over the state without modification by income, education level, or regional differences in environmental attitudes. Predicted changes to particulate matter emissions from residential and commercial sources are modest with slight reductions of ~10% mostly attributed to energy efficiency measures. Widespread adoption of biomethane to replace natural gas is predicted in the GHG-Step scenario but this fuel change has little impact on criteria pollutant emissions.

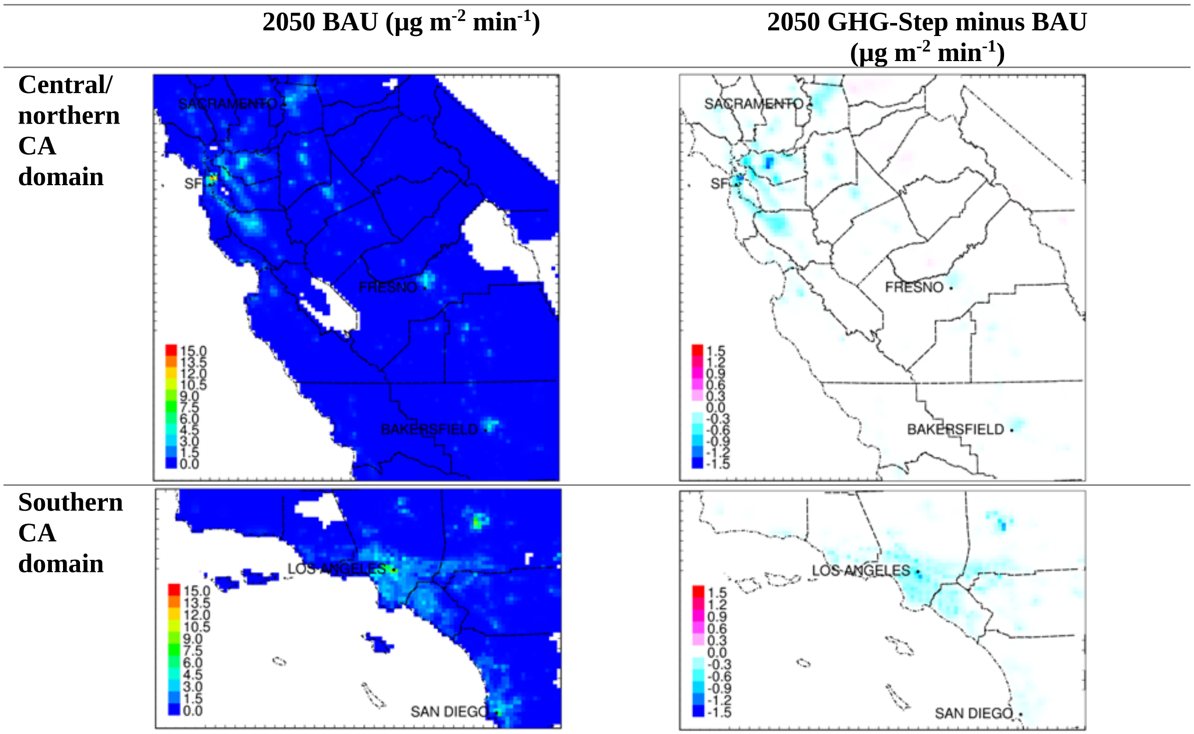


Figure 15: Particulate matter emissions from residential and commercial sources in the BAU scenario (left panels) and emissions change in the GHG-Step scenario (right panels). Units are µg m^{-2} min^{-1}.

3.4 Electricity Generation Emissions

Fig. 16 illustrates predicted emissions of particulate matter from combustion processes used to generate electricity. These emissions are represented as point sources and so only the grid cell containing an electrical generation unit are colored. The highest emissions rates for individual grid cells are associated with a small number major electrical generation stations typically powered by natural gas in the BAU scenario. The majority of the colored grid cells in Fig. 16 are associated with smaller backup generators that operate intermittently and therefore have very low emissions. These backup units are typically powered by a fossil fuel such as diesel fuel in the BAU scenario, with a shift to biofuels in the GHG-Step scenario. This fuel switch has modest impact on total emissions given the low utilization of these units.

Peak emissions rates of particulate matter in the GHG scenario decrease by a factor of ~1.7 in the GHG-Step scenario primarily due to a reduction in fossil fuel electricity generation in favor of a shift to solar and wind sources (see Fig. 10). All generating stations are assumed to continue operation at a reduced rate in the GHG-Step scenario rather than selectively decommissioning some stations. The age and efficiency of existing natural gas generating stations will likely be key factors determining how they are operated in the future scenarios. Solar and wind electricity generation does not emit criteria pollutants and so the location of these facilities is not shown in Fig 16.

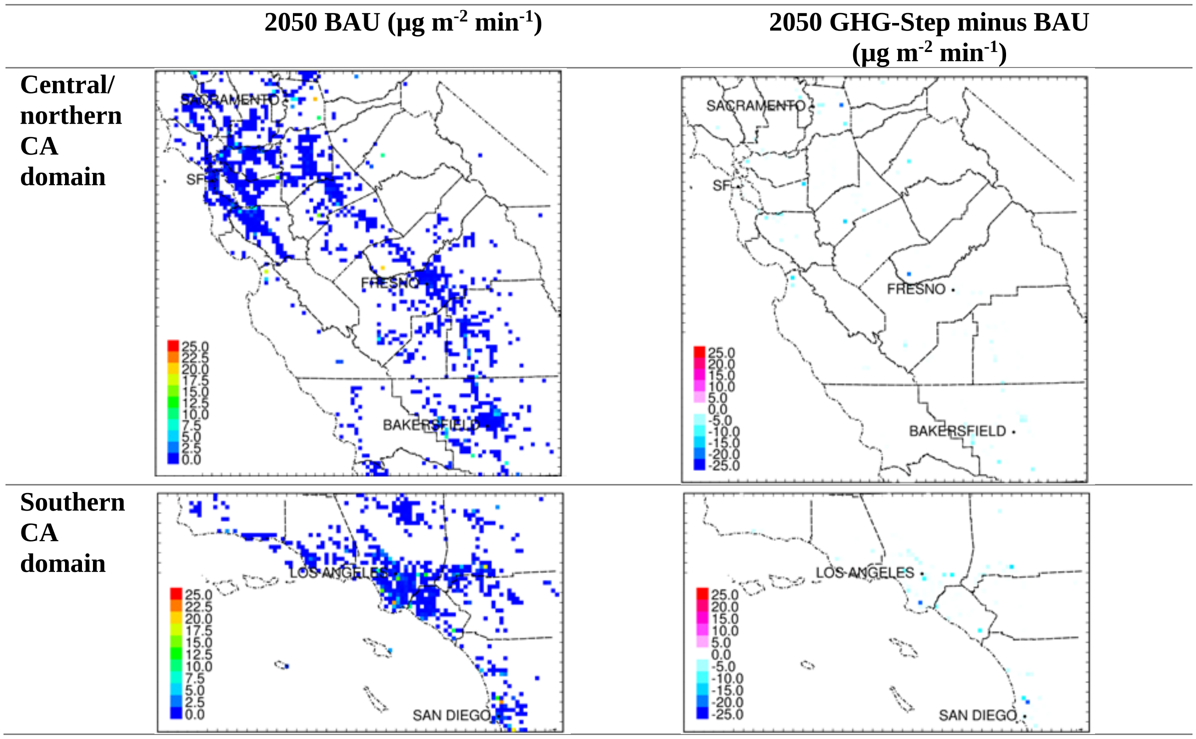


Figure 16: Particulate matter emissions from electricity generation (emission source category type 6) in the BAU scenario (left panels) and emissions change in the GHG-Step scenario (right panels). Units are μg m^{-2} min^{-1}.

3.5 Biorefinery Emissions

Figure 17 shows the locations of refineries producing biofuels (bio-refineries) in California under the BAU and GHJG-Step scenarios considered in the present study. The location of future bio-refineries was chosen to minimize transportation costs for the raw materials feeding into the refinery and the delivery of fuel to the final point of end-use. Additional zoning constraints were considered to prevent the placement of bio-refineries near schools, hospitals or other locations with sensitivity populations. More generally, a constraint was considered to restrict the placement of new bio-refineries in regions that currently violate the NAAQS. The top panels of Fig. 11 therefore do not allow the placement of bio-refineries in either the SJV or the SoCAB, while the less constrained scenarios illustrated in the lower panels of Fig. 17 do not impose this restriction. In practice, bio-refineries were generally sited near landfills, industrial, or agricultural areas within each city selected as economically optimal within the specified constraints.

The enforcement of NAAQS constrains on bio-refineries lead to a smaller number of larger refineries under both the BAU and GHG-Step scenarios. Note that overall bio-refining output is higher in the BAU scenario than in the GHG-Step scenario. Bio-fuels have lower associated GHG emissions than traditional fossil fuels but their carbon intensity is still too high to meet the GHG emissions target represented in the GHG-Step scenario. The CA-TIMES model therefore predicts that a portion of the energy supplied by biofuels in the BAU scenario will be supplied instead by wind and solar in the GHG-Step scenario.

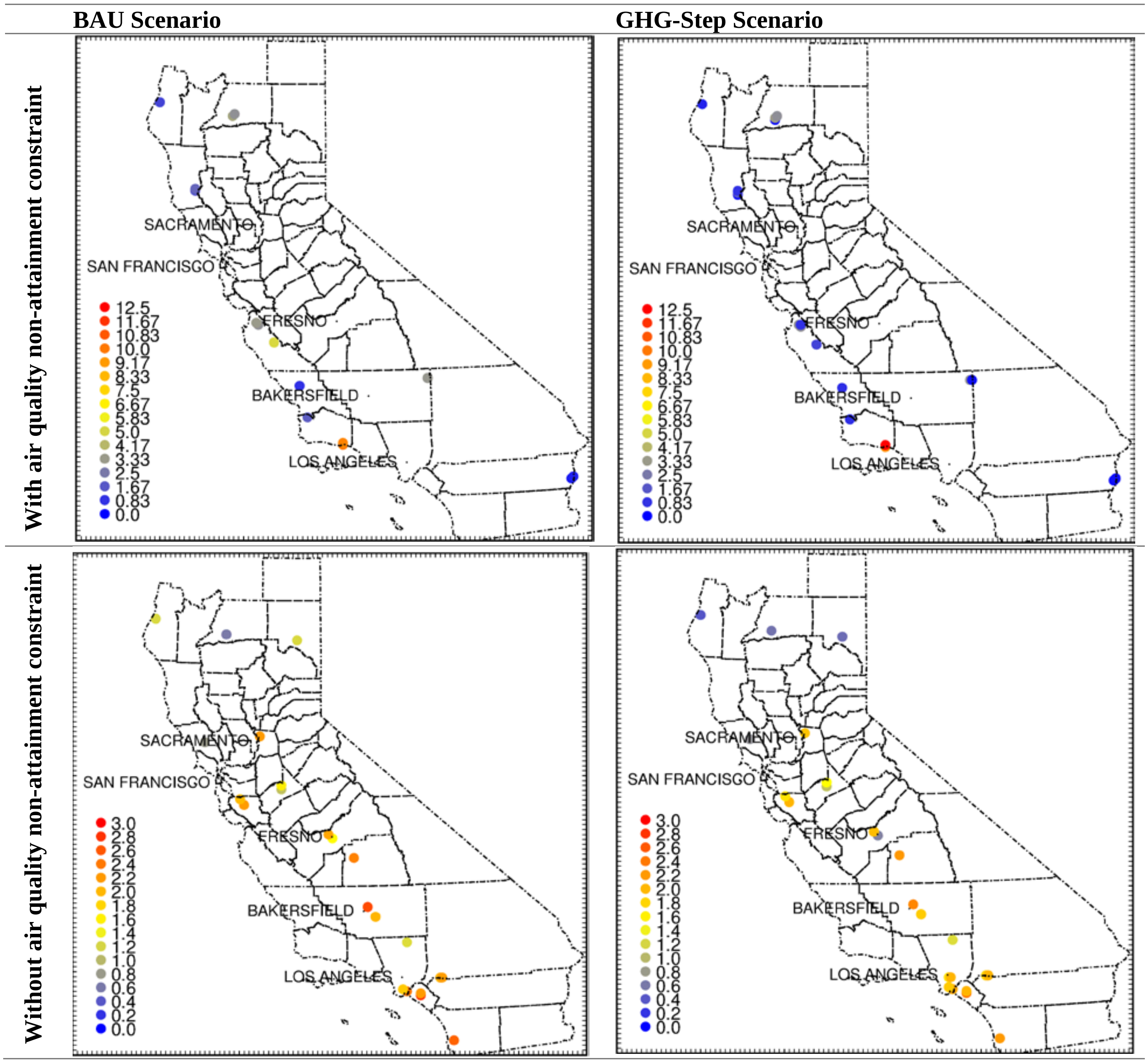


Figure 17: Biorefinery locations under the BAU scenario (left column) and the GHG-Step scenario (right column). Legend shows $PM_{2.5}$ mass emission rates per facility in µg m^{-2} min^{-1}. Top panels represent the constrained case where biorefineries cannot be located in air basins out of compliance with National Ambient Air Quality Standards (NAAQS). Bottom panels are not constrained by NAAQS status.

3.6 Summary of Statewide Emissions

Fig. 18a illustrates the net change in emissions related to criteria pollutants in California in the GHG-Step scenario versus the BAU scenario analyzed in the current study. Emissions of each pollutant are broken down by the major emissions categories analyzed in Section 2. The miscellaneous category is equivalent in the BAU and GHG-Step scenarios and hence is not plotted. Contributions below 0% indicate emissions reductions, while contributions above 0% indicate emissions increases. Each of these changes represents the statewide average for the sources within the indicated sector. Note that the changes within each sector may not be uniform across the entire state. The net change in total emissions is indicated by the black horizontal line for each species. It is immediately apparent that the emissions reductions illustrated in Fig. 18a are not uniform for all pollutants. Maximum reductions of ~60% are observed for CO_2 and particulate copper (Cu) emissions. In contrast, emissions of particulate SO_4^{2-}, gaseous CO and gaseous SOx actually increase under the GHG-Step scenario due to tradeoffs in the technologies adopted in the off-road mobile categories (rail, marine, aviation, etc) needed to optimize the overall GHG emissions across the state. Emissions of pollutants that experience increasing trends in Fig. 18a are minor in the present-day inventory and so that they do not currently trigger NAAQS violations. Changes in key, highly emitted pollutants fall in between the extreme cases described above (see results for particulate EC, particulate OC, and gaseous NOx). Each of these pollutants experiences a net decrease in total emissions averaged across California, but emissions changes are not uniform across all categories. Some technology and fuel changes cause higher emissions which are offset by savings in other categories. This complex mixture of tradeoffs reflects the optimal economic approach to GHG reductions determined by the CA-TIMES model.

The changing activity patterns, fuels, and technologies included in the GHG-Step scenario lead to changes in the emitted particle size and composition distribution. This leads to differences in the response of primary particulate matter with aerodynamic diameter less than 2.5 μm ($PM_{2.5}$) and less than 0.1 μm ($PM_{0.1}$; ultrafine particles). Ultrafine particles are an emerging pollutant of concern expected to influence public health (Delfino et al., 2005; Hoek et al., 2010; Knol et al., 2009). The results shown in Fig. 18a illustrate that the GHG-Step scenario leads to only a 4% decrease in primary $PM_{2.5}$ emissions but a much larger 36% reduction in $PM_{0.1}$ emissions. Recent epidemiology results indicate that $PM_{0.1}$ is associated with mortality in the California Teachers Study (Ostro et al., 2015). Likewise, toxicology studies indicate that ultrafine particles are more toxic than larger particles per unit mass (Donaldson et al., 2002; Donaldson et al., 2001; Elder et al., 2006; Kreyling et al., 2004; Oberdorster et al., 2002). Enhanced $PM_{0.1}$ emissions reductions could amplify the potential health benefits of the future GHG-Step scenario beyond the level expected from $PM_{2.5}$ emissions reductions.

Fig 18b. shows the net change in criteria pollutant emissions predicted using the expert analysis approach described by Shindell et al. (2012). These results are presented as a comparison point to the results illustrated in Fig. 18a and listed in SI Table S36 through Table S38. The expert analysis scenario focused on a small number of measures targeted for countries which are in the early stages of adopting policies to reduce GHG emissions or mitigate regional air quality problems. As a result, the measures described by Shindell et al. have a large impact on global public health but they will have a very minor impact on California (or any other major state or country that has already implemented significant emissions controls).

Comparison of Fig. 18a and Fig. 18b illustrates that only reductions in particulate EC are comparable in the Shindell et al. and CA-TIMES scenarios due to the mitigation of emissions from off-road diesel engines. CA-TIMES accomplishes this reduction through a combined switch in fuels and adoption of diesel particle filters on remaining diesel and bio-diesel sources to achieve a combined reduction in GHG emissions and criteria pollutant emissions. Shindell et al. assume uniform adoption of diesel particle filters on all off-road diesel engines with no fuel switching. Shindell et al. also specify the adoption of digesters for dairy waste and increased use of landfill gas as renewable methane sources. CA-TIMES predicts similar adoption resulting in a ~35-40% reduction in ammonia (NH_3) emissions from these sources. The CA-TIMES approach considered in the present study additionally considers how the emissions of bio-methane differ from the emissions of traditional natural gas. The only other significant measure specified by Shindell et al. that could reduce criteria pollutant emissions in California is a complete ban on burning of agricultural waste. California already limits agriculture burns to avoid stagnation periods. Thus, even the apparent savings associated with reduced agricultural burns shown in Fig. 18b are likely to have limited practical impact on air quality in the state. Shindell et al. do not consider the adoption of low carbon fuels or electrification of on-road vehicles which are necessary to achieve deep GHG reductions in CA.

Overall, the analysis presented by Shindell et al. (2012) is appropriately targeted at global health but the measures considered in this analysis do not achieve California's GHG objectives and the criteria pollutant emissions changes associated with them will not support calculations for future air quality in California. Energy economic models such as CA-TIMES represent a more realistic tool for development of scenarios in regions like California that have already considered all simple measures. Careful analysis is required to understand the resulting complex pattern of tradeoffs between emissions in different categories that results from these scenarios.

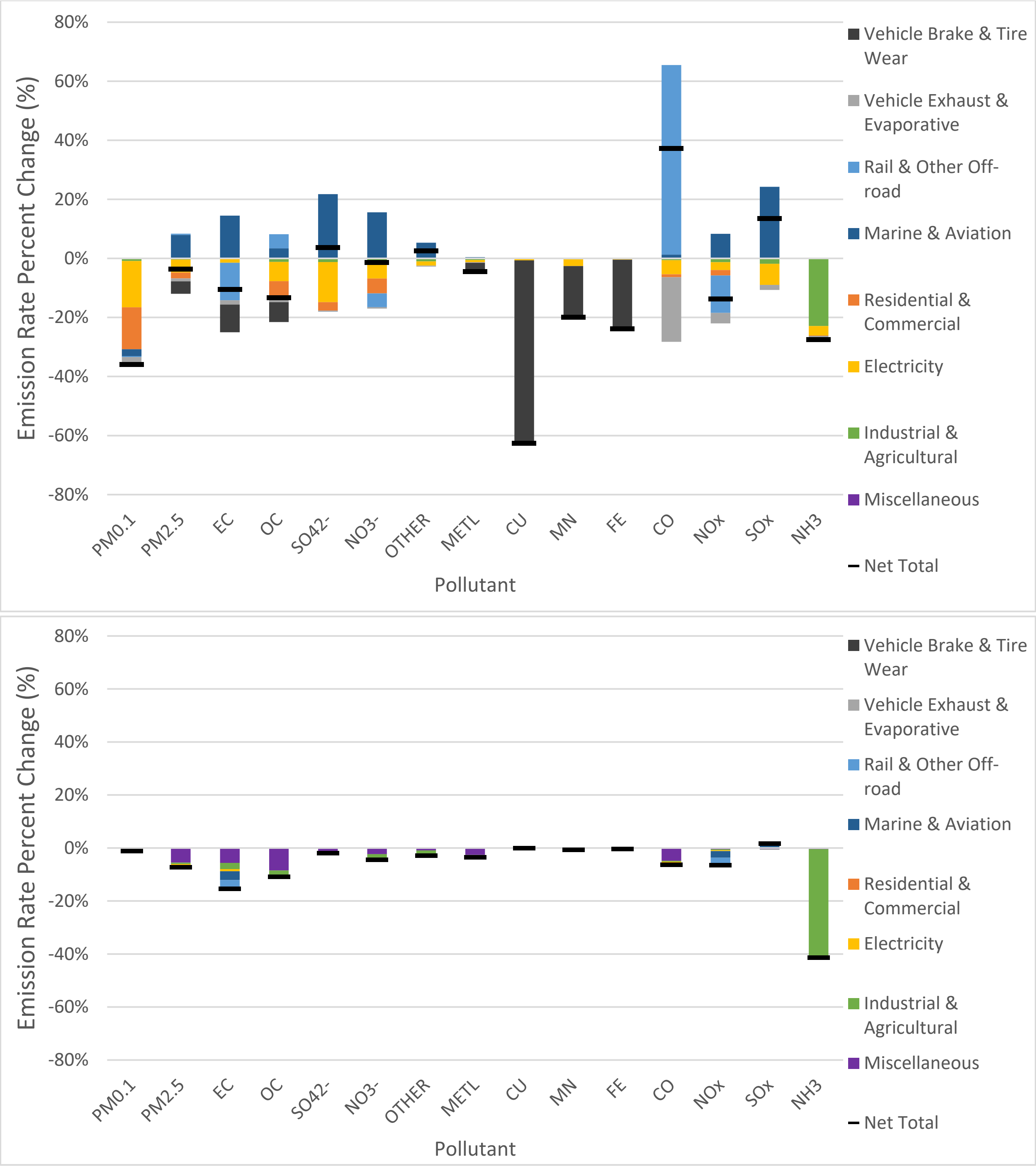


Figure 18: Change in pollutant emission rate relative to BAU scenario. Panel (a) represents GHG-Step analyzed in the current study using the CA-TIMES model. Panel (b) represents expert analysis presented by Shindell et al. (2012).

Fig. 19 illustrates examples of spatial patterns of emissions changes under the GHG-Step scenario predicted by CA-TIMES in the current study. The offsetting increasing and decreasing emissions changes illustrated in Fig. 18 do not occur uniformly over the state but instead appear as regions of localized increasing and decreasing emissions. As an even greater complication, the spatial pattern of increasing and decreasing emissions changes for each pollutant. The top panels of Fig. 19 illustrate changes in the commercial and residential sector for NOx emissions (Fig 19a) and OC emissions (Fig 19b) in central California. Patterns of emissions increases or decreases are similar in major urban centers (San Francisco and Sacramento) but different patterns are predicted for emissions of NOx and OC in the heavily polluted San Joaquin Valley (Fresno and Bakersfield). The lower panels of Fig. 19 illustrate even stronger variation in the spatial pattern of emissions changes in the off-road and rail categories in southern California. The spatial pattern of the change in particulate EC emissions (Fig. 19c) differs strongly from the spatial pattern of the change in particulate OC emissions (Fig. 19d).

All of the emissions illustrated in Fig. 19 will produce regions of increased or decreased pollutant concentrations. Given that each region is highly populated, these emissions patterns will have a direct effect on population exposure. Detailed analysis with regional air quality models at a resolution of 4km or finer will be required to understand the health implications of these changing emissions. California requires this level of fine-scale emissions analysis to accurately predict the air quality impacts of future GHG mitigation strategies in the state. Similar efforts will be required to analyze the effects of GHG mitigation strategies on criteria pollutants in other highly-populated regions that have already moved beyond simple emissions regulations banning obvious sources of air pollution.

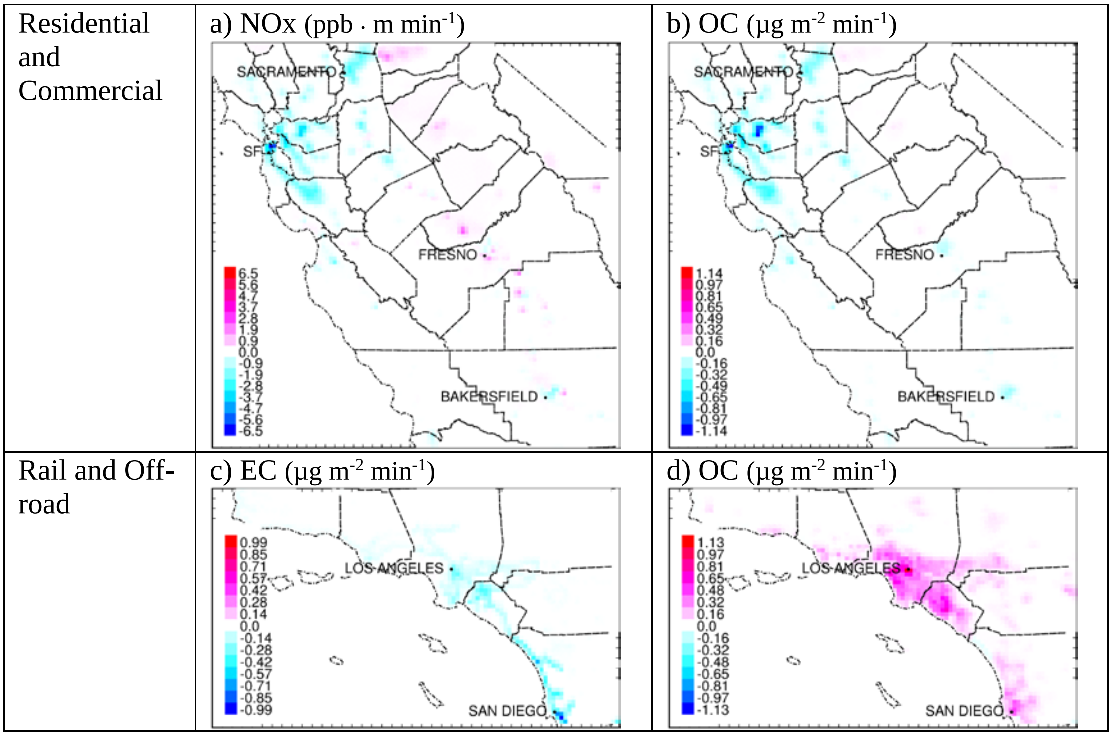


Figure 19: Change in emissions in the GHG-Step scenario relative to the BAU scenario . (a) NOx from residential and commercial sources (ppb · m min^{-1}), (b) particulate OC from residential and commercial sources (µg m^{-2} min^{-1}), (c) particulate EC from off road and rail sources (µg m^{-2} min^{-1}), and (d) particulate OC from off road and rail sources (µg m^{-2} min^{-1}).

The CA-REMARQUE projections for criteria pollutant emissions associated with optimal climate policies in California should not be directly extrapolated to other regions or countries. Instead, the methods used by CA-REMARQUE should be applied to each new region to fully consider the appropriate energy resources available, consumption patterns, equipment vintages, aftertreatment regulations and population and economic growth rates.

Each region may have a different optimal set of GHG mitigation technologies and policies that will lead to different rates and spatial patterns of emission compared to the changes predicted in California. Many developing regions will be able to select less expensive GHG mitigation strategies that also reduce GHG and criteria pollutant emission relative to their BAU scenario. Within developed regions such as other U.S. states, the elements of the mobile emissions inventory maintained by the U.S. EPA (MOVES and mobile portion of the National Emissions Inventory) can be adapted to replace the corresponding California information (EMFAC, mobile portion of the CARB inventory). Changes to off-road emissions would need to be estimated following procedures similar to those employed in the CARB off-road VISION model. Effort would be needed to estimate how changes to marine fuel sources would influence emissions at major ports. Studies would need to be conducted describing potential locations for new facilities producing low-carbon fuels and the resulting emissions from those facilities. This information would support a fully resolved analysis of the criteria pollutant emissions associated with climate policies outside of California.

4 Conclusions

The California REgional Multisector AiR QUality Emissions (CA-REMARQUE) model has been developed to translate optimized GHG mitigation policies to criteria pollutant emissions in California. Minimum-cost GHG policies are first selected with the energy economic model CA-TIMES. Tailored methods are then used to predict corresponding changes in criteria-pollutant emissions for individual categories including on-road vehicles, off-road vehicles, marine, aviation, rail, residential, commercial, electricity generation, industrial, and agricultural emissions. Translation methods account for efficiency improvements, changing technology, and changing fuels with corresponding changes to criteria pollutant emissions. Modifications to the composition of reactive organic gases and the size and composition of airborne particulate matter are considered. Translation methods also account for increased emissions associated with some measures, such as the need to produce new bio-fuels including bio-diesel, ethanol, and hydrogen.

The CA-REMARQUE model is demonstrated by predicting emissions in 2050 under a Business as Usual scenario (BAU) and an optimized GHG mitigation scenario (GHG-Step) in California. The results show that the optimal scenario for GHG mitigation produces increasing criteria pollutant emissions in some categories that are offset by decreases in other categories. These tradeoffs yield a complex pattern of emissions trends with sub-regions of increasing emissions and sub-regions of decreasing criteria pollutant emissions across California when viewed at 4km spatial resolution. In contrast, a simplified expert analysis scenario designed to address global GHG emissions may not necessarily reduce criteria pollutant emissions in California because many emission sources have already been controlled by the state's air pollution regulations. The expert analysis method does not consider complex fuel switching scenarios beyond the replacement of natural gas with biomethane. Choosing an economically optimal scenario of additional measures needed to achieve GHG mitigation goals in California requires tools beyond expert analysis opinions. Likewise, fully accounting for the corresponding changes to criteria pollutant emissions requires sophisticated analysis in fully developed countries and states with strict existing environmental regulations.

The California sub-regions of increasing and decreasing criteria pollutant emissions predicted in the current project occur in close proximity to major population centers and so they will almost certainly influence population exposure and public health. The emissions inventories created in the current study will be analyzed using regional air quality models in a future study to fully calculate impacts on public health.

4 Code and Data Availability:

CA-REMARQUE was developed and executed in the Linux programming environment using standard shell scripts and FORTRAN programs compiled using the Portland Group software. All of the data necessary to calculate changes to emissions inventories are published in full in the main text and supporting information section of the manuscript. The output emissions datasets are available free of charge at faculty.engineering.ucdavis.edu/kleeman/. The program code is currently being updated to use the latest version of the California EMFAC software and will be posted at faculty.engineering.ucdavis.edu/kleeman/ when complete. Note that the CA-REMARQUE v1.0 model is separate from the CA-TIMES energy-economic model and the California EMFAC model.

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