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Future emissions of particles and gases that cause regional air pollution in California under different greenhouse gas mitigation strategies

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HIGHLIGHTS

• Six future energy scenarios were translated into detailed emissions inventories for air pollution in California.

- Deep GHG mitigation scenarios significantly reduce emissions of air pollution precursors, yielding significant reductions in predicted ground-level PM_{2.5} concentrations.
- Carbon capture and sequestration strategies yielded only one third of the public health benefits compared to the deep GHG reduction approaches.
- Deep GHG mitigation scenarios that used additional natural gas experienced higher concentrations of ultrafine particles.

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ABSTRACT

Mitigating future climate change and managing future air quality are inter-related fields that have the potential to benefit from coordinated strategies that leverage the efforts in one area to achieve positive outcomes in the other area. California plans to reduce greenhouse gas (GHG) emission by 80% (relative to year 1990) by the year 2050. The changes required to meet this target also have the potential to improve air quality. Previous work developed an energy-economic model CA-TIMES and an emission inventory model CA-REMARQUE to study the possible pathways of meeting the GHG mitigation target and the air pollutant emissions associated with those pathways. Here we update the CA-TIMES and CA-REMARQUE model framework and analyze six different scenarios: (i) BAU - a business-as-usual future reference scenario, (ii) CAP30 - a loose GHG reduction scenario that meets current policy references but only achieves a 40% GHG reduction (relative to 1990 levels) by the year 2030, (iii) GHGAi - a climate-friendly 80% GHG reduction scenario featuring broad adoption of advanced technologies and renewable energies, (iv) CCS - a scenario that achieves 80% net GHG reductions but allows for more fossil energy combustion by focusing on adoption of carbon capture and sequestration technology, (v) NGB- a variation on the GHGAi scenario that allows for more natural gas combustion for residential and commercial buildings, and (vi) NGT - a variation of the GHGAi scenario that allows for more natural gas combustion for electricity generation. Results show that the GHGAi deep GHG mitigation scenario significantly reduces emissions (-41% PM_{0.1}, -8% PM_{2.5}, and -26% NO_X) and improves air quality (-1 µg m⁻³ PM_{2.5}) yielding public health benefits (+USD 20B yr⁻¹) relative to the BAU scenario. The CCS scenario achieves the same GHG reductions but increases emissions in some areas (+2.5% PM2.5) resulting in only one third of the public health benefits compared to the GHGAi scenario. The NGB and NGT scenarios show that an 18% increase in natural gas utilization in buildings or a 15% increase in natural gas power generation offsets 32% and 46% of the ultrafine particle emission reduction achieved in the GHGAi scenario but has little impact on PM2.5 concentrations, producing approximately 90% of the public health benefits of the GHGAi scenario. These public health benefits should be considered when making decisions about future GHG mitigation strategies.

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1. Introduction

Greenhouse gas (GHG) mitigation is a dynamic process that must recognize changes in population, attitudes towards policy enforcement, technology advancement, land-use and lifestyle (Brown et al., 2018; Keshavarzmohammadian et al., 2017; Penrod et al., 2014; Rudokas et al., 2015; Shi et al., 2017; Shindell et al., 2018; Trail et al., 2015; Zapata et al., 2018a, 2018b). GHG mitigation occurs within a framework of other environmental efforts with the potential for overlap in multiple areas, including air pollutant emissions and regional air quality impacts (Ellis and Wolf, 2011). California is the most populous state and the second largest greenhouse gas emitter in the U.S. due to its high economic activity. It is also home to seven of the ten most polluted cities in the U.S. (American Lung Association, 2021) primarily because of unfavorable topography and meteorology that keeps emissions trapped close to the surface. California is at the forefront of the leading economies across the world in the development of science-based policies to address climate change and air pollution issues. Since the passage of Assembly Bill 32 (CARB, n.d.) that calls for California to bring about an 80% reduction in GHG emissions (relative to 1990 levels) by year 2050, researchers have been working on constructing and analyzing possible pathways to bring about the decarbonization of the energy system (McCollum et al., 2012; Yang et al., 2014, 2015, 2016; Yang and Ogden, 2013). As part of this effort, a California-specific integrated multi-sector energy-economic-optimization model framework CA-TIMES (Yang et al., 2014, 2015) was developed to identify least-cost approaches to achieve target levels of GHG reduction subject to policy constraints. Soon after, an emission inventory model CA-REMARQUE (California Regional Multisector Air Quality Emissions Model) was developed to map changes in energy, technology and activity from the CA-TIMES model to changes in air pollutant emissions (Zapata et al., 2018a). The resulting analysis determined that the transition to a low carbon energy future could avoid \sim 25% of the premature mortality associated with air pollution in California (Zapata et al., 2018a). The public health savings associated with improved air quality are significant and must be considered in future planning exercises.

Although some of the technology advancements and fuel shifts needed to ensure a low-carbon sustainable future are well understood at this time (i.e. electric and fuel cell vehicles, wind and solar electricity generation), the optimal combination of other key technologies and their associated environmental impacts still require further analysis. For example, carbon capture and sequestration (CCS) technology is being widely discussed due to its promising GHG reduction benefits, but some studies note the potential for environment and public health disbenefits (Klein et al., 2011; Muratori et al., 2017; Rhodes and Keith, 2005; Sanchez and Kammen, 2016; Shindell et al., 2018; Wang et al., 2020). Several studies report that CCS deployed in fossil fuel power plants could increase emissions of GHGs, gas-phase oxides of nitrogen (NOx), and airborne particulate matter because of low carbon capture efficiency and the additional fuel consumption needed to power the CCS unit (European Environment Agency, 2011; Jacobson, 2019; Sanchez and Kammen, 2016). In contrast, CCS deployed in a biomass integrated gasification combined cycle (Bio-IGCC) is considered to be a promising negative carbon emission technology with competitive costs compared to other carbon mitigation strategies (Klein et al., 2011; Muratori et al., 2017; Rhodes and Keith, 2005; Zang et al., 2020). The air quality impacts of Bio-IGCC-CCS have not yet been evaluated, and this study provides information that will begin the process of addressing this gap. Natural gas is another energy resource that is being studied intensely in GHG mitigation plans due to its potential to provide a transition pathway from heavy petroleum fuel to renewable energy while simultaneously improving air quality. Natural gas power plants and the associated infrastructure contribute to carbon "lock-in" where current structures would remain in place for decades before phasing out (Sproul et al., 2020), but natural gas pipeline infrastructure can also distribute renewable gaseous fuel (e.g biomethane) (Mac Kinnon et al., 2018) yielding climate benefits. The fast response time for natural gas power plants also plays an important role in balancing electricity service load in grids that rely on intermittent renewable energy (i.e. wind and solar) (Demirhan et al., 2021). Despite these potential climate benefits, natural gas combustion emits significant amounts of ultrafine particulate matter ($PM_{0.1}$) (Venecek et al., 2019; Xue et al., 2018), potentially degrading some of the benefits provided by technologies with zero emissions (i.e. wind and solar). The optimal strategy for natural gas usage in a low carbon future that balances GHG emission and air quality remains to be explored.

In this study, we incorporate the latest energy system projections from the CA-TIMES model and update the accompanying emission inventory model CA-REMARQUE (Zapata et al., 2018a) to generate future air pollutant emission inventory under six different scenarios in California: (i) BAU - a business-as-usual future reference scenario, (ii) CAP30 - a loose GHG reduction scenario that meets current policy references but only achieves a 40% GHG reduction (relative to 1990 levels) by the year 2030, (iii) GHGAi - a climate-friendly 80% GHG reduction scenario featuring broad adoption of advanced technologies and renewable energies, (iv) CCS - a scenario that achieves 80% net GHG reductions but allows for more combustion to generate electricity by focusing on adoption of carbon capture and sequestration technology, (v) NGB - a variation on the GHGAi scenario that allows for more natural gas combustion for residential and commercial buildings, and (vi) NGT - a variation of the GHGAi scenario that allows for more natural gas combustion for electricity generation. The combination of the latest versions of CA-TIMES and CA-REMARQUE produced a California-specific, detail-rich air pollutant emission inventory with 4 km resolution. The two-model framework retains internally consistent new-technology and alternative-energy projections throughout the emission inventory, while also considering the appropriate spatial allocation of the emissions. The present study updates the BAU and GHG scenarios created using previous versions of CA-TIMES and CA-REMARQUE (Zapata et al., 2018a) and compares them to alternative scenarios. These results support calculations using chemical transport models to predict future air pollution concentration fields. The identification of potential benefits and disbenefits for future air quality can help policy makers minimize the undesirable outcomes of GHG mitigation efforts while simultaneously optimizing the energy-environment-economic relationship.

2. Methods

2.1. The CA-TIMES model and future scenarios

CA-TIMES is an integrated energy-engineering-environmentaleconomic systems model focusing on the transition of California's energy system (Yang et al., 2014). Built upon the MARKAL-TIMES optimization framework, CA-TIMES is rich in technological detail across all of the supply and demand sectors of the energy economy, including fuel production and conversion, electricity production, and energy consumption in the residential, commercial, industrial, transportation, and agricultural end-use sectors. CA-TIMES selects the economically-optimal mix of energy supplies to satisfy demand subject to the specified resource limits, policies, and any exogeneous constraints. Numerous scenarios have been generated by the CA-TIMES model to understand the transition costs and technology/resource implication of long-term strategies to decarbonize California's energy system (Yang et al., 2014, 2015). Six scenarios in the year 2050 were chosen for detailed air pollution emissions analysis in the current study.

 "BAU" - A "business-as-usual" scenario that serves as a future reference. This scenario incorporates current regulations to achieve the goal outlined in California AB32, which requires greenhouse gas emissions in 2020 to be below 1990 levels (427 MMTCO₂-e) but otherwise does not constrain future emissions beyond that date. Given that the official state-wide total GHG emissions were 425 MMtCO₂-e in 2018 and 418 MMtCO₂-e in 2019, it seems likely that the 2020 targets were achieved. The BAU scenario assumes that population and economic growth through 2050 will require a baseline level of energy service similar to current conditions and it incorporates the most important current policies that drive this energy system development (see Table S1). The BAU scenario provides an example for how California's energy system could potentially develop in the absence of any substantial effort to move toward a low-carbon society beyond 2020.

- 2) "CAP30" A loose GHG mitigation scenario that reduces GHG emissions to 40% below 1990 levels by the year 2030 but does not constrain or invest further in future GHG reduction. This scenario represents an intermediate future decarbonization situation.
- 3) "GHGAi" A "climate friendly" scenario that reduces 80% GHG emissions (relative to year 1990) by the year 2050. The deep decarbonization requires market mechanisms such as a cap-andtrade program to augment existing policy programs. The GHGAi scenario uses a "step" carbon cap, meaning GHG emissions are only limited at the 2020 level (=1990 level) between 2020 and 2049 but then dropped to 80% below 1990 emissions in the year 2050. This step-cap allows maximum flexibility to determine the optimum costeffective trajectory to meet the GHG mitigation target by adjusting the timing for adoption of different types of efficient resources and technologies.
- 4) "CCS" A scenario that focuses on the impact of deploying carbon capture and sequestration (CCS) technology. This scenario generates 24% of all electricity with Bio-IGCC-CCS, which results in over 80 M tons of CO₂-eq negative carbon emissions. The negative emissions in the electrical sector allow for more fossil fuel consumption in other sectors (especially transportation), while still achieving a net GHG reduction of 80% relative to 1990 levels (similar to that in GHGAi).
- 5) "NGB" A GHG mitigation variation scenario that focus on the impact of natural gas usage in residential and commercial buildings. The shift from natural gas appliances (furnaces, water heating, etc.) to electricity appliances is limited, resulting in 20% more natural gas usage in buildings compared to other deeply decarbonized scenarios such as GHGAi.
- 6) "NGT" A GHG mitigation variation scenario that focus on the impact of natural gas usage to generate electricity. Electricity generation from natural gas is allowed to increase from 10% in the GHGAi scenario to 30% in the NGT scenario.

2.2. The updated CA-REMARQUE model

The California Regional Multisector Air Quality Emissions (CA-REMARQUE_v1.0) model (Zapata et al., 2018a) was developed to predict changes to criteria pollutant emission inventories in California in response to sophisticated emission control programs and energy scenario projections provided by the CA-TIMES model. CA-REMARQUE achieves this goal by combining detailed information from each economic sector with the latest outputs from multiple models to better represent activity patterns and emission locations in a series of tailored algorithms. For example, the EMFAC model (CARB, n.d.) is used to project future on-road mobile emissions, the VISION (CARB, n.d.) scenario planning model is used to project future off-road transportation activities, the SWITCH-WECC model (RAEL, n.d.) is used to project future electricity load in different subregions of California, the GREET (ANL, n.d.; CARB, n.d.) model is used to predict emissions from biomass and hydrogen facilities, and the H2-TIMES model (Yang and Ogden, 2013) is used to project locations for new hydrogen production facilities. The CA-REMARQUE model also compiles the latest published values for pollutant emission factors from new energy and technologies. All of these features make the CA-REMARQUE model a high resolution, detail-rich emission inventory model catering specifically to California's needs. The original version of the model CA-REMARQUE model (v1.0) has been documented in a previous study (Zapata et al., 2018a).

CA-REAMRQUE was updated to version 2.0 in the current study to be compatible with the latest version of the CA-TIMES model and other related model outputs as summarized below.

In the on-road transportation sector, CA-REMARQUE_v2.0 incorporated the updated Emission Factors (EMFAC) 2014 model results, which allowed direct emission projection to the year 2050. CA-REMARQUE_v1.0 worked with the EMFAC 2007 model that only projected emissions to the year 2035 and required extrapolation from 2035 to 2050. The Emission Inventory Code (EIC) cross-reference table between EMFAC vehicle class and technology and CA-TIMES vehicle types was updated in CA-REMARQUE_v2.0 (see Table S2).

CA-REMARQUE_v2.0 was updated to require the adoption of diesel particle filter treatment technology for all of the off-road and agricultural equipment that run on diesel and biodiesel in the year 2050. This specific control technology was not fully implemented in CA-REMARQUE v1.0. Aircraft emissions in the Los Angeles region that were missing in CA-REMARQUE v1.0 were added in CA-REMARQUE v2.0. Additional emission scaling factors of 0.45 for SO_X and 0.85 for NO_x were applied to all of the Bio-IGCC-CCS power plants in the CCS scenario in CA-REMARQUE v2.0 because NaOH scrubbers are typically used to control flue gas SO₂ concentrations to avoid contamination of the amine-based carbon capture solvent (Koornneef et al., 2010; Young et al., 2019). Exhaust stack information was updated and carefully matched to corresponding emissions records in the electricity generation, industrial, and commercial sectors in CA-REMARQUE_v2.0 to ensure reasonable plume rise heights. The updates summarized above slightly alter the BAU and GHG-Step scenarios analyzed previously (Zapata et al., 2018a). The current study presents updated versions of the BAU and GHGAi scenarios as internally-consistent reference points for comparison to the CAP30, CCS, NGB, and NGT scenarios.

2.3. Air quality model

Future air pollution concentrations were predicted using the UCD/ CIT air quality model (Kleeman and Cass, 2001; Yu et al., 2019) with a spatial resolution of 4 km over central California and Southern California that contains more than 90% of the total population in the state. Simulations were conducted over 32 individual weeks (each including three days of spin up time) randomly selected over a ten-year window from 2046 through 2055. The resulting concentrations characterize the long-term average concentration in the presence of meteorological variability associated with the El Nino Southern Oscillation (ENSO). Large scale meteorological inputs were obtained from the Community Climate System Model (CCSM) (NCAR, 2011) under the Representative Concentration Pathway 8.5 (RCP8.5) (IPCC, 2014). Fine scale meteorology was downscaled using the Weather Research and Forecasting (WRF) model v3.4. Biogenic emissions were predicted using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.1. Wildfire emissions were assumed to be independent of the energy scenarios and so were not considered in the current analysis since they would not change the relative difference between each scenario.

2.4. Health impact model

The public health impacts of altered $PM_{2.5}$ concentrations were predicted using the BenMAP-CE v1.5 model maintained by the US EPA (Sacks et al., 2018). The $PM_{2.5}$ health impact function was taken to be an evenly-weighted average of four independent epidemiological studies (Krewski et al., 2009; Laden et al., 2006; Lepeule et al., 2012; Pope et al., 2002). Avoided mortality was translated to a monetary value using the standard value of a statistical life (VSL) recommended by US EPA yielding a VSL equivalent to USD 7.6 M. Avoided mortality per 1M residents was projected to total avoided mortality based on an expected population in California of 45M in the year 2050 (*Report P-1A: Total Population Projections, California, 2010–2060 (Baseline 2019 Population* *Projections; Vintage 2020 Release)*, 2021). The spatial distribution of population assumed in health impact calculations was consistent with the distribution assumed in emissions projections.

3. Results and discussion

3.1. Energy system transition and GHG emission

California's energy system must change significantly in order to reduce GHG emissions by 80%. Fig. 1 summarizes the primary energy portfolios used in 2010 and in the scenarios developed for the year 2050. Large differences are apparent between the energy mix used in current vs. future energy scenarios, and between future energy scenarios that meet the 80% reduction target vs. scenarios that have lower levels of GHG reduction. Fig. S1 presents the evolution of primary energy consumption (PJ) within each scenario from 2010 to 2050 with a 5-year time step. Renewable sources (solar, wind, biomass, and other renewables including hydraulic power) account for only 5% of total energy in the year 2010 but grow to \sim 30% of total energy in the 2050 BAU scenario and over 50% in the GHGAi, NGB and NGT scenarios.

GHG emissions are closely related to the portfolio of energy sources used in each scenario. Fig. 2 summarizes the CO₂-equivalent GHG emission in 2010 and each of the 2050 scenarios while Fig. S2 shows the evolution of GHG emissions between 2010 and 2050. The reference scenario BAU reduces GHG emissions by 39% relative to the base year 2010, mainly through decarbonizing the transportation sector (-24%)and the electricity generation sector (-9%). These predictions reflect the effectiveness of current policies that target mobile and power plant emissions. GHG emissions from the residential and commercial sectors increase in the 2050 BAU scenario compared to the year 2010 due to population growth. The GHGAi scenario eliminates 61% of the electricity generation GHG emissions, 87% of the building sector GHG emissions, and almost 100% of the transportation sector GHG emissions relative to the BAU. The CAP30 scenario produces electricity with less carbon intensity than the BAU scenario but shows no further decarbonization in either the building or the transportation sectors. The CCS scenario is able to generate negative GHG emissions due to the adoption of Bio-IGCC-CCS technology that captures carbon from the atmosphere during biomass accumulation and "eliminates" carbon from the atmosphere by storing it in underground reservoirs. The negative emissions are used to offset emissions from the transportation and residential sectors yielding a net reduction in total GHG emissions in the CCS scenario that are similar to the GHGAi scenario (87.5 M ton CO₂-eq). As expected, GHG emissions are higher in the NGB scenario (buildings) and in the NGT scenario (electricity generation) compared to the GHGAi scenario. It is also noteworthy that the GHG emissions contributions from non-energy sectors (soil, livestock, waste treatment, etc.) increase from 11% in the BAU scenario to 37% in the GHGAi scenario as the major GHG emissions sources undergo deep decarbonization.



Fig. 1. California energy system transition as represented by the percentage of different types of primary energy in 2010 and 2050 scenarios.



Fig. 2. Total greenhouse gas emission (M ton CO_2 -eq) from different economic sectors modeled by CA-TIMES in 2010 and 2050. Emissions scenarios are defined in Section 2.1.

3.2. Particulate and gaseous pollutant emissions from different future scenarios

Fig. 3 summarizes the changes to criteria pollutant emissions under the CAP30, CCS, GHGAi, NGB, and NGT scenarios relative to their reference scenarios. For CAP30 (Fig. 3(a)), CCS (3b) and GHGAi (3c) the reference scenario is BAU, and for NGB (3d) and NGT (3e), the reference scenario is GHGAi. The emissions changes contributed from sectors 1 to 7 are represented by the colored bars and the final value illustrates the net total change. For example, when looking at PM_{2.5} change in the GHGAi scenario (Figure 3(c), 2nd column), sectors 1 to 7 contributed -1.1%, -0.8%, -3.0%, +5.3%, +5.3%, -2.6% and -0.2% respectively, resulting in a net total PM2.5 emissions change of -7.6% relative to the BAU scenario. NOx emissions decrease by 26% in the GHGAi scenario relative to the BAU scenario, mainly from sector 3 (off-road equipment -12%) and sector 5 (residential and commercial buildings -9%). The CAP30 scenario also achieves noticeable $PM_{2.5}$ reductions from sector 3 (-1.2%) and sector 6 (electricity generation -3.1%) but overall, as a partial mitigation scenario, it results in less PM2.5 emissions reduction compared to the GHGAi scenario (-4.6% vs. -7.6%). The CCS scenario has elevated $PM_{2.5}$ emission (+2.5%) because there is a major emission increase from sector 4 (marine and aviation +5.3%) and no major reduction from sector 5 or sector 6 (+0.44%). SOx emissions increase in the CCS scenario because the Bio-IGCC plants emit more SO_X than other electricity generation processes even though the accompanying CCS section removes more than half of the increased SOx. PM2.5 emissions increase 1.8% and 2.3% in the NGB and NGT scenarios, respectively, due to increased natural gas utilization relative to the GHGAi reference scenario.

Emissions of ultrafine particles (Dp $< 0.1 \ \mu m$) change much more than emissions of fine particles (Dp $< 2.5 \,\mu$ m) and coarse particles (Dp $< 10 \ \mu m$) across all scenarios. This effect is illustrated in the first three columns of Fig. 3 and in panels a-c of Fig. S3. Fig. 4(a) summarizes sector contributions to PM_{0.1} emissions under different scenarios, while Fig. 4(b) compares the sector contributions to $PM_{0.1}$, $PM_{2.5}$ and PM_{10} emissions under the BAU scenario. $\ensuremath{\text{PM}_{2.5}}$ and $\ensuremath{\text{PM}_{10}}$ emissions pie charts for the other scenarios are not shown because they show minimal difference (<5%) as discussed with Fig. 3. Sector 5 (residential and commercial buildings) and sector 6 (electricity generation) together account for approximately 50% of the ultrafine particles emissions. Sector 3 (offroad equipment and railroad) and sector 4 (marine and aviation) account for approximately 25% of the total ultrafine particle emissions. The remaining $\sim 25\%$ of the ultrafine particle emissions come from sector 8 ("other processes") that does not vary between future scenarios. On-road mobile emissions (type 1 & 2) are projected to contribute only \sim 1% of the ultrafine emissions in the future.



Fig. 3. Pollutant emission change (%) in different scenarios relative to their reference scenario. Stacked colored bars represent contributions from different socioeconomic sectors.



Fig. 4. (a) Ultrafine particulate matter emissions from different sectors under different scenarios (b) Ultrafine, fine and coarse particulate matter emissions under the BAU scenario.

As Fig. 3 summarizes, the four major sectors contributing to emission changes are identified to be: residential and commercial buildings, electricity generation, marine and rail, and off-road equipment. The following sections discuss the fuel and technology changes leading to the observed emissions differences in each of these sectors.

3.3. Particulate and gaseous pollutant emissions from different socioeconomic sectors

Residential and commercial buildings. The residential and commercial building sector consumes large amounts of energy and therefore produces large amounts of emissions. This sector accounts for approximately 40% of end-use energy consumption in California and contributes to 13% of the GHG emissions from direct fossil fuel combustion in the year 2010, not including GHG emissions associated with electricity consumption in buildings (Yang et al., 2014). In 2050, the CA-REMARQUE_v2.0 model predicts that residential and commercial buildings will account for 20–50% of total $PM_{0.1}$ emissions and 15–20% of total $PM_{2.5}$ emissions. The majority (+70%) of the $PM_{2.5}$ emitted by commercial and residential buildings is composed of organic compounds.

Fig. 5(a–f) shows $PM_{0,1}$ emission rates (ug·m⁻²·min⁻¹) from northern and southern California domains under the six scenarios analyzed in the present study. Fig. 5 panel (a) presents the absolute $PM_{0,1}$ emissions rate from the reference BAU scenario, while panels (b)-(d) show the difference in the $PM_{0,1}$ emissions rate relative to the reference scenario. The GHGAi scenario generates the greatest amount of $PM_{0,1}$ emission reduction around the cities with large populations (San Francisco Bay Area, Greater Los Angeles). The spatial pattern of the $PM_{0,1}$ emissions are similar to the spatial pattern for the $PM_{2,5}$ and NO_X emissions (Figs. S14 and S28).

Changes to ultrafine particle emissions in the building sector can be linked directly to changes in natural gas combustion in the built environment. Buildings in California are projected to use either natural gas



Fig. 5. (a) $PM_{0.1}$ emission ($\mu g \cdot m^{-2} \cdot min^{-1}$) from residential and commercial sector in the BAU scenario, and (b–f) changes in $PM_{0.1}$ emissions ($\mu g \cdot m^{-2} \min^{-1}$) relative to the indicated reference scenario.

or electricity in the year 2050, but emissions from the latter energy source are tabulated in the electricity generation sector. Fig. S8 summarizes the demand for natural gas and electricity from residential and commercial buildings in 2050. The deep GHG reduction scenarios (GHGAi, NGB and NGT) are able to satisfy the same energy service demand with only 74% of the total energy needed in the BAU scenario due to the adoption of more efficient building appliances (efficiency gains of $1.7 \times$ for residential and $1.3 \times$ for commercial buildings appliances). Moreover, many natural gas appliances are replaced by electric appliances the GHGAi scenario, as reflected by the increasing share of electricity demand. These two factors combine to reduce natural gas consumption by 80% in residential buildings and 57% in commercial buildings under the GHGAi scenario. Measurements have shown that natural gas combustion in appliances emits particles exclusively in the ultrafine size range (Xue et al., 2018). The widespread use of natural gas as an energy source makes it a dominant contributor to ultrafine particle emissions but relatively low emissions rates dilute the contributions to PM_{2.5} emissions (Yu et al., 2019). Natural gas combustion in the building sector accounts for 28% of the PM_{0.1} emission change but only 5.3% of the PM_{2.5} emission change in the GHGAi vs BAU scenarios.

The share of natural gas increases from 21% to 39% of total building energy demand in the NGB scenario vs. the GHGAi scenario, resulting in more $PM_{0.1}$ emissions around major population centers (Fig. 5(e)). Although the building sector $PM_{0.1}$ emissions are still less in the NGB scenario than in the BAU scenario (see Fig. S13), the increased use of natural gas in the NGB scenario offsets a third of the $PM_{0.1}$ reduction achieved through electrification and efficiency improvement in the GHGAi scenario (GHGAi $PM_{0.1}$ –41.5%, NGB $PM_{0.1}$ –28.4% relative to BAU). These results emphasize the importance of limiting natural gas in the building sector if reducing $PM_{0.1}$ emissions is a priority.

Electricity generation. Fig. 6(a) shows the statewide $PM_{0.1}$ emissions from electricity generation under the BAU scenario and Fig. 6(b–f) show the change in emissions associated with other scenarios. Power plants are point sources but the BAU scenario assigns these emissions to the 4 km model grid, with a small number of major emissions cells and a much larger number of low-level emissions cells around the populated regions (see Fig. 6(a)). Changes to point source emissions in Fig. 6(b–f) are illustrated as circles with radius proportional to the emission values



Fig. 6. (a) $PM_{0,1}$ emissions ($\mu g \cdot m^{-2} \cdot min^{-1}$) from electricity generation in the BAU scenario, and (b-f) changes in $PM_{0,1}$ emissions ($\mu g m^{-2} min^{-1}$) relative to the indicated reference scenario.

to show the results more clearly. The CAP30, CCS and GHGAi scenarios all have major $PM_{0.1}$ emissions reductions compared to the BAU scenario due to reductions in natural gas combustion to generate electricity. The BAU scenario generates 676 PJ of electricity from natural gas power plants, with significant reductions for CAP30 (-80%), CCS (-96%) and GHGAi (-64%) scenarios. The GHGAi, NGB and NGT scenarios electrify across many sectors and therefore require much more total electricity generation than other scenarios. This extra electricity is mainly generated from renewable resources including wind, solar, geothermal, biomass and hydro. Natural gas accounts for only 9% of the electricity generation in the GHGAi and NGB scenarios. However, the share of natural gas electricity increases to 26% in the NGT scenario, resulting in a significant $PM_{0.1}$ emission increase centered at the natural gas power plants (Fig. 6(f)). Total $PM_{0.1}$ emissions still decrease by 22.4% under the NGT scenario relative to the BAU scenario but a significant portion of the 41.5% $\mbox{PM}_{0.1}$ emissions reduction in GHGAi scenario is eroded in the NGT scenario.

Despite the decrease of natural gas electricity and the increase of wind and solar power, the CCS scenario is drastically different from the other scenarios in the way that 24% of the electricity (379 PJ, see Fig. S9) comes from Bio-IGCC-CCS. Therefore, in the CCS scenario $PM_{2.5}$ and NO_X emission increase from the northern California biomass and solid waste power plants as shown in Fig. 7 and Fig. S29. It is noteworthy that the changes of $PM_{0.1}$ and $PM_{2.5}$ in the CCS scenario relative to BAU can go in different directions (Fig. 6(c) vs Fig. 7) because power plants with different technologies have different PM emission profiles that center in the ultrafine (natural gas electricity) or fine (biomass electricity) portion of the airborne particle size distribution.

Marine vessels and aircrafts. Sector 4 emissions are dominated by marine vessels including ocean-going vessels, shipping on inland



Fig. 7. $PM_{2.5}$ emission ($\mu g \cdot m^{-2} \cdot min^{-1}$) from electricity generation, CCS-BAU.

waterways, and recreational boating. PM2.5 emissions from sector 4 account for 10% of total PM2.5 emissions in the BAU scenario, but this contribution increases to 17% in the deep GHG reduction scenarios (GHGAi, NGB and NGT). Fig. S16 illustrates the spatial pattern of the increasing emissions from sector 4. PM2.5 emissions increase from shipping lanes far offshore in all scenarios (CAP30, CCS and GHGAi) relative to BAU, with significantly larger increases apparent in the CCS and GHGAi scenarios. Shipping activities far offshore currently use residual fuel oil (RFO), which is a heavy petroleum fuel. All of the RFO is replaced with biomass-based residual fuel oil (BRFO) in the BAU scenario to lower pollutant emissions (see Fig. S7). Supplies of BRFO are limited by available feedstocks, and the increased demand for biofuels in the CCS, GHGAi, NGB and NGT scenarios redirects most of those feedstocks to the production of transportation fuels or electricity generation. The CCS, GHGAi, NGB, and NGT scenarios therefore use RFO for most offshore shipping needs. A slight PM2.5 emissions decrease from nearshore shipping activities and inland waterway activities shown in the CAP30 scenario is the result of switching from diesel to biodiesel. The GHGAi, NGB and NGT shipping emissions are very similar.

Off road equipment and railroads. Figs. S17, S23 and S31 show the statewide $PM_{2.5}$, $PM_{0.1}$ and NO_X emissions rates from off road equipment and railroads respectively. $PM_{2.5}$ emissions rates uniformly decrease in the CAP30, CCS, and the GHG scenarios (GHGAi, NGB and NGT) relative to the BAU scenario in and around large cities and along the rail lines. This decrease results from replacing diesel with biodiesel in railroads and off-road equipment, electrifying railroads, and replacing gasoline with ethanol in off-road equipment. The fuel usage changes are

presented in Fig. S5 as total energy demand from different scenarios. PM_{0.1} and NO_X emissions do not decrease uniformly, but rather emissions for these pollutants increase in some locations and decrease in other locations (Figs. S23 and S31). For example, PM_{0.1} emissions in the GHGAi scenario decrease along the rail lines but increase at and around major cities (Fig. S23 d) because all railroads are electrified to eliminate PM_{0.1} emissions while replacing gasoline with ethanol in the off-road equipment increases PM_{0.1} emissions (Surawski et al., 2010; Zapata et al., 2018a). NO_X emissions decrease at most locations across California in the GHGAi, NGB and NGT scenarios (Fig. S30), but increase along the rail lines and at the Port of Los Angeles in the CAP30 and CCS scenarios. Replacing gasoline with ethanol reduces NOx emissions in off-road equipment (- 45%), while replacing biodiesel with diesel increases NOx emissions in railroads (+13%) and off-road equipment (+8%) (Zapata et al., 2018a). These results illustrate the complexity in predicting the effects of fuel switching on criteria pollutant emissions. Multiple factors acting in opposite directions dictate the net effect on overall emissions.

On-road vehicles. PM2.5 emissions from vehicle tailpipes account for only 0.8% of total PM2.5 emissions in the 2050 BAU scenario due to the implementation of existing standards. These tailpipe emissions further decrease to less than 0.1% of total $PM_{2.5}$ emissions in the deep GHG mitigation scenarios (GHGAi, NGB and NGT) as a result of largescale electric vehicle adoption. Fig. S4 shows that the share of electric and fuel cell vehicles is only 5.5% in the BAU scenario but increases to over 70% in the GHGAi, NGB and NGT scenarios. The CCS scenario allows more gasoline and diesel in vehicles compared to the GHGAi scenario (but still less than BAU) because of the negative GHG emissions from electricity generation. The NGB scenario further increases the share of electric and fuel cell vehicles to over 88% to compensate for the increased GHG emissions from residential and commercial buildings. Therefore, all scenarios reduce $PM_{2.5}$, $PM_{0.1}$ and NO_X emissions from tailpipes relative to the BAU scenario, with the GHGAi, NGB, and NGT scenarios getting close to zero tailpipe emissions in 2050 (Fig. S18 S24 and S32). $\ensuremath{\text{PM}_{2.5}}$ emissions from vehicle tire and brake wear accounts for 5-6% of the overall PM2.5 emissions in the year 2050 BAU scenario, exceeding emissions from tailpipes. CA-TIMES predicts the same vehicle miles traveled in all scenarios and so the differences in predicted tire and brake wear emissions are related to the adoption of various amounts of regenerative braking and vehicle weight in electric, hybrid electric and fuel cell vehicles (Fig. S19). Regenerative braking systems are estimated to reduce tire and brake wear PM emissions by 59% (Antanaitis, 2010).

3.4. Airborne particulate matter concentrations

The long-term (~10 year average) ground-level PM2.5 concentrations predicted under each of the emissions scenarios considered in the current study are summarized in Fig. 8. PM_{2.5} concentrations under the BAU scenario peak over urban areas such as Los Angeles and the San Francisco Bay Area, but concentrations are also high downwind of major electrical generating stations near Monterey Bay (south of San Francisco) and around intensive agricultural sources in the San Joaquin Valley between Fresno and Bakersfield (Fig. 8(a)). PM_{2.5} concentrations decrease under all scenarios that reduce GHG emissions, but the extent of the reductions and the spatial pattern depend on the details of the emissions changes (Fig. 8(b-f)). The CAP30 scenario and the CCS scenario produce similar levels of PM_{2.5} reduction in major urban centers, but increasing PM_{2.5} concentrations are predicted at locations outside of urban centers under the CCS scenario due to the increased use of fossil fuel combustion under this scenario. Much stronger $PM_{2.5}$ reductions are predicted across the entire study region under the GHGAi scenario, with more than 1 μ g m⁻³ of reductions across most populated regions in the study domain (Fig. 8(d)). PM_{2.5} concentrations under the NGB and NGT scenarios are slightly higher than the GHGAi scenario (Fig. 8(e and f)) due to the increased use of natural gas combustion but still significantly lower than concentrations under the CAP30 and CCS scenarios.



Fig. 8. (a) Long-term $PM_{2.5}$ concentrations predicted under the BAU emissions inventory, and (b–f) change in long-term $PM_{2.5}$ concentrations associated with changing energy portfolios relative to the indicated reference scenario in the panel title. All units $\mu g m^{-3}$.

3.5. Public health benefits

Fig. 9 illustrates the public health benefits associated with reduced $PM_{2.5}$ concentrations under the emissions scenarios considered in the current study. All GHG mitigation scenarios produce net health savings relative to the BAU across the study region, including the CCS scenario that produced some zones of increasing $PM_{2.5}$ concentrations. Air pollution mortality associated with $PM_{2.5}$ exposure was estimated at 23,875 deaths per year in the 2050 BAU scenario. The GHGAi scenario produced the greatest overall health benefits equivalent to approximately 3500 avoided deaths per year, and an annual public health benefit greater than USD 20B yr⁻¹. The less aggressive CAP30 and CCS scenarios produced only one third of these public health benefits due to more modest $PM_{2.5}$ reductions in these scenarios. The NGB and NGT scenarios are similar to the GHGAi scenario with approximately 90% of

the public health benefits (3300 avoided deaths per yr).

4. Conclusion

Six different future energy scenarios in California were analyzed for their emissions of particulate matter and gaseous pollutants related to regional air quality using the CA-TIMES and CA-REMARQUE model framework. These scenarios are informative examples of possible carbon emissions reduction strategies, not literal predictions of future energy consumption. The scenarios provide valuable information to understand the key resources and technologies (i.e. natural gas, CCS) while trying to simultaneously reduce GHG emissions and improve air quality.

The GHGAi scenario represents the most cost-effective pathway to reduce GHG emissions by 80% (relative to 1990 level) without the deployment of negative carbon emission technology. Strategies in the



Fig. 9. Avoided mortality due to improved air quality associated with changing energy portfolios relative to the BAU scenario. Total population in 2050 is assumed to be 45 million. Public health benefits estimation assumes a present-day value of a statistical life equivalent to USD7.6M. All calculations performed with BenMAP-CE.

GHGAi scenario include aggressive decarbonization of electricity generation, adoption of electricity for most end-use applications, efficiency improvements for appliances, and deployment of low-carbon transportation fuels and technologies. Major air quality and public health benefits are generated under the GHGAi scenario due to the significant emissions reductions for $PM_{0.1}$ (41%), $PM_{2.5}$ (8%), and NO_X (26%) relative to the reference future BAU scenario. Long-term air quality simulations predict that ground-level $PM_{2.5}$ concentrations will decrease by more than 1 µg m⁻³ across most of California's major population centers under the GHGAi scenario, reducing air pollution mortality by approximately 3500 deaths per yr with a public health benefit greater than USD 20B yr⁻¹.

The CCS scenario achieves the same GHG reductions as the GHGAi scenario, but the negative GHG emissions from Bio-IGCC-CCS technology allow more fossil energy consumption in transportation and built environment. $PM_{0,1}$ emissions in the CCS scenario decrease (-25%) relative to the BAU scenario as a result of less natural gas usage in buildings and power plants, but PM2.5 emission increase (+2.5%) suggesting potential air quality disbenefit associated with the CCS future especially around the Bio-IGCC-CCS power plant locations. Even though total PM2.5 emissions increased under the CCS scenario, overall public health improved relative to the BAU scenario because the increase in PM_{2.5} concentration occurs in sparsely populated areas. Despite these mitigating factors, the air quality benefits associated with the CCS scenario are three times lower than the air quality benefits associated with the GHGAi scenario (USD 7 $B \cdot yr^{-1}$ vs 22 $B \cdot yr^{-1}$). The strong difference in public health benefits should be taken into consideration as part of the cost associated with CCS technology when planning future GHG mitigation strategies. The strong difference in public health benefits should be taken into consideration as part of the cost associated with CCS technology when planning future GHG mitigation strategies.

The NGB and NGT scenarios tested the impact of loosening natural gas usage limitations in the buildings and power plants compared to the strict GHGAi scenario. Increasing the share of natural gas by 18% in buildings increased PM_{0.1} emissions (22%), PM_{2.5} emissions (1.8%) and NO_X emissions (2.5%) in the NGB scenario relative to the GHGAi scenario. Increasing the share of natural gas in electricity generation by 15% increased PM_{0.1} emissions (26%), PM_{2.5} emissions (2.3%) and NO_X emissions (1.5%) in the NGT scenario relative to the GHGAi scenario. Projected PM2.5 concentrations slightly increase across California in the NGB and NGT scenarios, and their associated public health benefits were slightly reduced relative to the GHGAi scenario. Such observations indicate that failing to strictly limit natural gas usage in buildings and power plants may not result in significant public health degradation associated with PM_{2.5} pollution in the future environment. The potential

health effects $PM_{0.1}$ pollution are not considered in the monetary estimation. Studies have shown the linkages between $PM_{0.1}$ pollution and asthma, hypertension, and ischemic cardiovascular disease (Schraufnagel, 2020), but the epidemiological evidence for $PM_{0.1}$ health effects is in the early stages of development (Ostro et al., 2015). The precautionary principle suggests that natural gas utilization in the built environment and electricity generation should be kept at a low level in order to maximize the air quality benefits gained from adoption of low carbon energy sources in California.

All emissions inventories described in the current manuscript can be downloaded free of charge at https://faculty.engineering.ucdavis.edu /kleeman/.

CRediT authorship contribution statement

Yin Li: created emissions inventories, Formal analysis, Writing – original draft. **Christopher Yang:** provided energy scenarios and helped integrate results with air quality analysis. **Yiting Li:** provided assistance running BenMAP calculations. **Anikender Kumar:** produced meteorological fields for air quality simulations. **Michael J. Kleeman:** designed the study, performed air quality simulations, performed BenMAP analysis, and revised the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2022.118960.

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