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Lifecycle Analysis of Air Quality Impacts of Hydrogen and Gasoline Transportation Fuel Pathways

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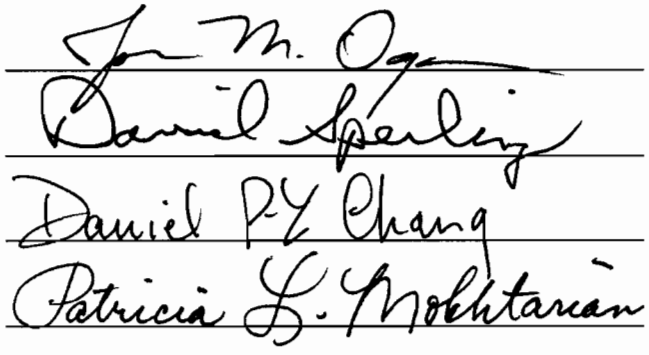

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To the memory of my father, to my mother, and to my wife Gengxin.

Guihua Wang
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Lifecycle Analysis of Air Quality Impacts of Hydrogen and Gasoline Transportation Fuel Pathways

Abstract

Hydrogen has been proposed as a low-polluting alternative transportation fuel. This dissertation analyzes the lifecycle air quality impacts of hydrogen and gasoline use in light duty vehicles, including impacts from fuel production, delivery, and vehicle use.

The analysis is conducted for various scenarios in Sacramento, California, for four pollutants: CO, NO_x, VOC, and PM_{10}. Three natural gas-based hydrogen supply pathways are considered: onsite hydrogen production via small-scale steam methane reforming (SMR), central SMR production with gaseous hydrogen pipeline delivery, and central SMR production with liquid hydrogen truck delivery. Four gasoline pathway scenarios, as compared to hydrogen pathways, are also investigated in the study. A new method is developed using travel demand model data to estimate air quality impacts of gasoline fleet operations, regression analysis is used to explore the relationship between lifecycle precursor emissions and secondary ozone formation for each hydrogen supply pathway, and a Gaussian atmospheric dispersion model is used to analyze ambient impacts.

The centralized/pipeline hydrogen pathway and the onsite hydrogen production pathway reduce pollution the most. The centralized hydrogen production with liquid truck delivery is the least clean option among the three means of hydrogen supply. The examined gasoline pathway, even with advanced new gasoline vehicles, would lead to much higher ambient concentrations of pollutants than the hydrogen pathways, producing 273 times greater CO, 88 times greater VOC, 8 times greater PM_{10}, and 3.5 times greater NO_x concentrations than those caused by the centralized/pipeline hydrogen pathway, assuming the same size vehicle population. The study also estimates the potential impacts of the above hydrogen pathways on secondary ozone air quality. The results indicate that adding a significant number of hydrogen fuel cell vehicles (FCVs) to the region would have a very small impact on secondary ozone pollution; in fact, it does not necessarily increase the peak ozone concentration, and may even cause it to decrease in some cases.

The results show that advanced gasoline vehicle technologies significantly reduce air quality impacts of light duty vehicles, but hydrogen vehicle technologies provide still greater benefits, reducing the contribution of light duty vehicles to ambient air pollutant concentrations to near-zero.

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1. Introduction

1.1. Research background

Hydrogen is a compelling alternative transportation fuel. Despite many technical and economic barriers, a hydrogen economy is quite attractive (NRC, 2004). Use of hydrogen in vehicles has many potential benefits (Sperling and Ogden, 2004). Hydrogen can be derived from a variety of sources such as natural gas (NG), coal, biomass, solar, wind, hydropower, and nuclear power, and could reduce oil supply insecurity (Ogden, 2002; Ogden, 1999a; Ogden, 1999b; Ogden et al., 2004). Hydrogen fuel cell vehicles (FCVs) produce no tailpipe emissions, and if made from renewables, decarbonized fossil fuels, or nuclear energy, hydrogen can also be produced and used with no emissions of greenhouse gases (GHGs), and could help mitigate global warming (Ogden, 2002; Ogden, 1999b; Ogden et al., 2004; Sperling and Ogden, 2004). In addition, fuel cell vehicles running on hydrogen could offer fuel economy around 2.5 times that of today's conventional internal combustion engine (ICE) vehicles (Ahluwalia et al., 2004; Farrell and Sperling, 2007; NRC, 2004).

In the U.S., the current petroleum-based transportation system emits significant amounts of carbon monoxide (CO), nitrogen oxides (NO_x), total organic gases (TOGs) or volatile organic compounds (VOCs), and particulate matter (PM_{10}, referring to particulates with an aerodynamic diameter less than 10 μm), as well as carbon dioxide (CO_2). To assure that transportation investments do not cause undermine efforts to attain ambient air

quality standards, the U.S. Clean Air Act requires that transportation plans for highway and transit projects be consistent with the air quality goals set by a state implementation plan (SIP) (U.S. DOT, 2007; U.S. EPA, 2007).

Although air quality in the U.S., in general, has been improving over the past several decades, it is still a challenging problem in many regions (designated to be in non-attainment). For example, mobile-source VOC and NO_x emissions are precursors to secondary ozone formation and aerosols. Particulates and ozone are the two criteria pollutants of greatest concern causing human health damage and leading to a social cost issue (ExternE, 1998; McCubbin and Delucchi, 1996; Murphy et al., 1999; Delucchi et al., 2002).

Hydrogen has been proposed as a low-polluting alternative transportation fuel that could help improve urban air quality. In particular, it has been suggested that hydrogen FCVs be introduced into the vehicle marketplace where zero emission vehicle (ZEV) mandates are enacted, e.g., in California (CARB, 2008a). To achieve high energy efficiencies and low overall emissions, the different pathways for producing hydrogen as a transportation fuel must be carefully examined (Wang, 2002). This dissertation analyzes the potential air quality impacts of hydrogen transportation fuel, using a lifecycle analysis (LCA) approach.

1.2. Research objectives

The overall objective of this study is to address the following questions.

(1) What would be the impact of hydrogen fuel cell vehicles on criteria pollutant emissions and air quality, considering all the emissions involved in the full fuel cycle, including producing, transporting, and using hydrogen?

(2) What changes in peak ozone pollution would occur if typical hydrogen supply pathways were introduced in Sacramento, California?

(3) What hydrogen supply strategy would be environmentally best for a specific county, region, or air basin in the U.S.? Is it onsite production or centralized production? Which delivery mode for centralized hydrogen is better, liquid truck or gaseous pipeline?

(4) What would the optimal, feasible spatial layout of hydrogen pathway steps be in a specific region, assuming that a possible hydrogen pathway type has been determined? and

(5) How do hydrogen FCVs compare to conventional or advanced gasoline vehicles in terms of the resulting impacts on emissions and air quality, from a lifecycle analysis perspective?

In this dissertation, a regional lifecycle analysis of air quality impacts is carried out to explore the hydrogen economy in more depth. We choose Sacramento, California as a site for our case study. The specific task-oriented objectives of this study are:

(1) To design typical, promising hydrogen pathways for the specific region, considering the regional feedstock resource availability;

(2) To compile emission inventories for several near-term alternative hydrogen pathways and both current and advanced gasoline fleet operations;

(3) To investigate the impacts of natural gas-to-hydrogen pathways on urban air quality of primary pollutants, using a Gaussian atmospheric dispersion model;

(4) To explore the relationship between secondary ozone formation and lifecycle precursor emissions from each hydrogen supply pathway, using regression analysis; and

(5) To develop a new methodological framework for estimating the air quality impacts of gasoline fleet operations, using travel demand model data and grid-level emission inventories.

1.3. Research approach: lifecycle analysis

This study uses the approach of lifecycle analysis (LCA), also written as lifecycle assessment. The LCA refers to the cradle-to-grave cycle of a product (Wang, 1999). More generally, LCA is a technique used to assess all the inputs and outputs of a product, process, or service (U.S. EPA, 2008). To assess the environmental aspects or potential impacts associated with a product, process, or service, the common procedure of LCA application usually has the following sequence (U.S. EPA, 2008).

(1) Lifecycle inventory. This refers to compiling a complete inventory of related energy and material inputs and outputs;

(2) Impact assessment. This refers to assessing the potential environmental impacts associated with inputs and outputs identified; and

(3) Lifecycle interpretation. This refers to interpreting the results to help people make a decision based on more comprehensive and complete information.

Lifecycle assessment of hydrogen production via natural gas steam reforming has been conducted in a number of studies, e.g., Spath and Mann (2000). The concept of a full fuel cycle is illustrated in Figure 1, using the case of hydrogen made from natural gas. A fuel cycle for a given transportation fuel includes, but is not limited to, the following three stages (Wang, 1999):

(1) The feedstock stage. Including feedstock extraction/production, transportation, and storage;

(2) The fuel stage. Including fuel production, transportation, storage, and distribution; and

(3) Vehicle operation. Also called downstream activities, including fuel combustion, evaporation, brake wear, and tire wear.

The full fuel cycle is also called well-to-wheels (WTW). In contrast, well-to-tank (WTT) includes all activities during both the feedstock and fuel stages; i.e., the vehicle operation stage is not included in WTT.

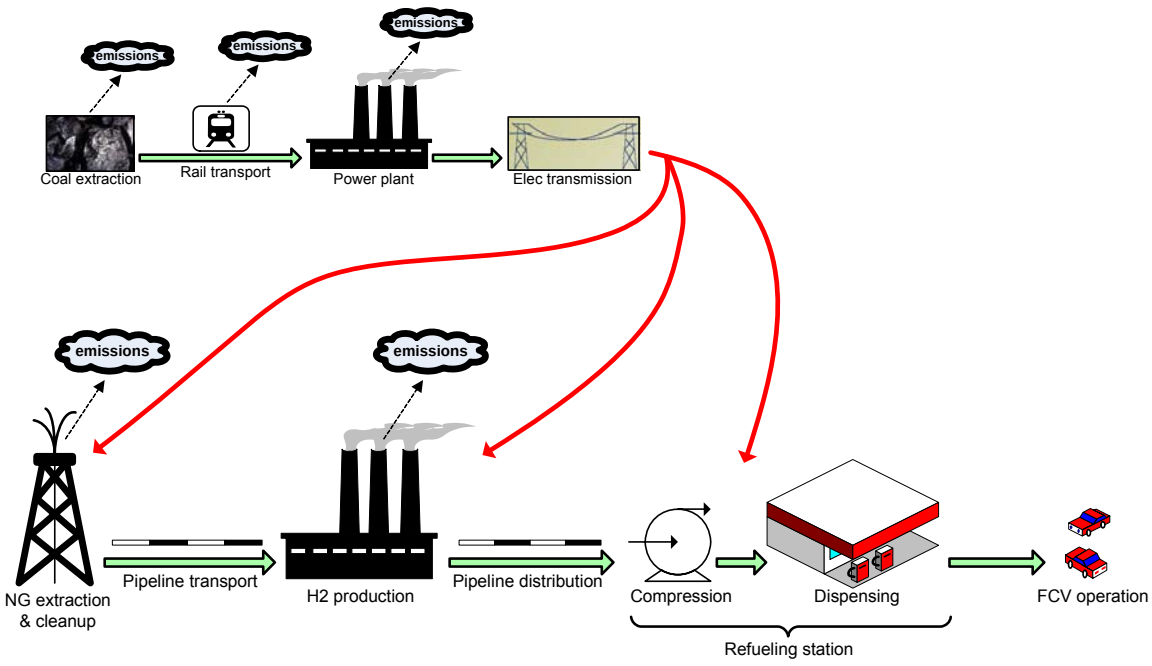

Figure 1. Full fuel cycle of hydrogen made from natural gas (i.e., hydrogen pathway). (The upper figure represents the sub-pathway of electricity consumed in steps of the primary fuel pathways.)

Several models have been developed for lifecycle analysis of alternative fueled vehicles (AFVs). To estimate lifecycle energy use and emissions for a transportation fuel, both the GREET and LEM models work very well.

The Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) model, developed by the Argonne National Laboratory, can calculate both the WTT and WTW results (Wang, 1999). About 100 fuel production pathways and 70 vehicle/fuel systems, covering all major vehicle technologies, are included and assessed in GREET (Wang, 1999).

The Lifecycle Emissions Model (LEM), developed by the University of California, Davis, can provide results for up to 30 countries and for the years 1970 to 2050 (Delucchi, 2003). The LCA boundary in LEM is defined more broadly, and it takes into account the following four individual lifecycles (Delucchi, 2003):

(1) Lifecycle of fuels and electricity;

(2) Lifecycle of vehicles. Including materials use, vehicle assembly, vehicle operation and maintenance, and secondary fuel cycle for transport modes;

(3) Lifecycle of materials. Including crude-ore recovery, finished-material manufacture, and the transport of finished materials to end users; and

(4) Lifecycle of infrastructure. Including energy use and materials production associated with the construction of highways, railways, etc.

Figure 1 shows a representative hydrogen pathway, which is a full fuel cycle for hydrogen made from natural gas. For a specific hydrogen pathway, the energy consumption and emissions vary with parameters such as the natural gas pipeline length, pipeline flow rate, plant capacity factor or plant size, plant conversion efficiency, truck distribution distance, etc. The emissions of criteria air pollutants (APs) and GHGs are closely related to pathway-specific situations and rely heavily on geographic or regional information.

Assuming that a certain number of hydrogen FCVs are operating in a specific region, Sacramento, California, this research investigates several possible fuel supply pathways to satisfy the vehicular demand for hydrogen fuel. Considering regional feedstock resource availability and commercially available hydrogen production technology, we focus on steam methane reforming (SMR) of natural gas in this dissertation.

Gasoline pathways, referred to as the petroleum-based fuel pathways (including both gasoline and diesel), are also examined for the purposes of comparison to hydrogen pathways. We investigate contributions of various current and advanced gasoline/diesel pathways to urban air pollution using travel demand model data.

Although some researchers have analyzed lifecycle emissions and energy use associated with hydrogen supply chains (Wang, 1999; Spath and Mann, 2000; Delucchi, 2003; Colella et al., 2005; Jacobson et al., 2005), no study has examined the change in air quality (i.e., pollutant concentrations rather than emissions only) due to a hydrogen transportation system. This study could provide the basis to quantitatively compare hydrogen and gasoline pathways in terms of the resulting air pollutant concentrations, and the results may be useful to policy makers in thinking about hydrogen.

1.4. Research contributions

The study makes an initial attempt to explore the lifecycle air quality impact of hydrogen pathways on a regional scale. It investigates the complicated but important relationships among transportation, energy, and the environment. The results of this research contribute to development of alternative fuel strategies, with important implications for policy makers. Below are specific original contributions.

(1) This study provides a quantitative basis for a better understanding of the air quality impacts of a hydrogen economy. It offers a more detailed and descriptive understanding of hydrogen systems than previous studies;

(2) This study develops new methods to quantitatively explore the air quality impacts of hydrogen-based transportation systems, for both primary criteria pollutants and secondary ozone formation. The results of comparison among the examined three hydrogen supply pathways are useful for determining hydrogen supply strategies;

(3) The methodological framework developed here for estimating the impacts of hydrogen pathways could be adapted to analyzing other alternative transportation fuel pathways, such as biofuels or coal-based hydrogen;

(4) This study depicts the region-specific empirical kinetic modeling approach (EKMA) property of secondary ozone formation, and points out that ozone formation is limited by NO_x in the summer and by VOC in the fall. This finding is useful for environmental policy makers to control ozone pollution more effectively;

(5) This study estimates the air quality impacts of light duty gasoline vehicles. The cases chosen span a range of possibilities for gasoline vehicles: from current technology to advanced technology. This allows us to estimate the air quality impacts resulting from cleaner, newer gasoline vehicles;

(6) This study develops a new systematic methodology for estimating contributions of mobile sources to urban air pollution, and it effectively connects transportation emissions and air quality. That would be useful for transportation conformity planning and mobile pollution control;

(7) The results, when comparing hydrogen/FCV scenarios with gasoline/ICE scenarios, will have meaningful implications for policy makers to evaluate

hydrogen, a promising alternative fuel, as compared to rapidly improving, low emission gasoline vehicles; and

(8) The results from this study are directly applicable for economists or scientists to estimate the external cost of air pollution causing human health damage (due to either hydrogen pathways or gasoline pathways), following such a methodological sequence: Source Emissions→ Ambient Pollution→ Physical Impacts→ Economic Evaluation.

1.5. Dissertation organization

The organization of this dissertation is as follows. Chapter 1 provides an introduction to the dissertation research and its framework. Chapter 2 estimates the lifecycle impacts of hydrogen supply pathways on urban air quality. Research emphasis is given to primary pollutants, including CO, NO_x, VOC, and PM_{10}, as well as SO_x. Going a step further, Chapter 3 explores the relationship between secondary ozone formation and lifecycle precursor emissions from hydrogen transportation systems by using regression analysis. Chapters 2 and 3 focus on hydrogen pathways. With a focus on gasoline fleet operations, Chapter 4 investigates the contributions of gasoline pathways to urban air pollution using traditional four-step travel demand model data. All the four important mobile-source pollutants, namely, CO, NO_x, VOC, and PM_{10}, are considered. In Chapter 5, both hydrogen and gasoline supply pathways are examined together, and their air quality impacts are compared from a lifecycle emissions perspective. Finally, Chapter 6

summarizes all the individual projects and presents the key findings of the entire dissertation research.

2. Lifecycle Impacts of Hydrogen Supply Pathways on Urban Air Quality of Primary Pollutants

2.1. Introduction

There is growing interest in hydrogen as a transportation fuel. To mitigate concerns about urban air pollution, zero emission vehicle (ZEV) mandates have been enacted in California (CARB, 2008a) and several other states in the U.S. Hydrogen fuel cell vehicles (FCVs) are a very promising ZEV option due to a number of potential advantages such as good performance of the FCV, various production sources of hydrogen fuel, and no CO_2 and criteria pollutant emissions throughout the vehicle lifetime (Ogden et al., 2004; NRC, 2004; Sperling and Ogden, 2004; Jacobson et al., 2005).

One of the key motivations for hydrogen is its potential to reduce emissions of air pollutants. In contrast, current mobile sources cause significant urban air quality degradation and damage to human health due to close proximity to people (Chaaban et al., 2001). In a European study, the damage costs of automotive air pollution were evaluated by using the impact pathways approach, and the impacts involved human health, agricultural crops, and building materials (Spadaro and Rabl, 2001). Although hydrogen FCVs emit no tailpipe emissions (Ogden et al., 1999), hydrogen must be produced from other sources and delivered to users. These steps can generate air pollutant emissions. Thus, the entire lifecycle from well to wheels must be considered in an assessment of hydrogen's air quality impacts. To achieve high energy efficiencies and low overall

emissions, the different pathways for producing hydrogen as a transportation fuel must be carefully examined (Wang, 2002). Lifecycle assessment of hydrogen production via natural gas steam reforming has been conducted in a number of studies, e.g., Spath and Mann (2000). Colella et al. (2005) examined the potential change in primary emissions and energy use from switching from the current U.S. on-road vehicle fleet to a hydrogen FCV fleet, using a lifecycle analysis (LCA) of alternative fuel supply chains, which provides positive evidence supporting the conversion to hydrogen FCVs for environmental and energy benefits. However, no study has quantitatively examined the changes in ambient concentrations (not just emissions) of pollutants resulting from hydrogen supply pathways on a regional scale.

Clearly, emissions for hydrogen (and associated environmental impacts) will depend on how hydrogen is made. Furthermore, air quality is related to emissions in complex ways that depend on many local factors, such as the mix of emission sources, meteorology, and geography. In this chapter, we are addressing the following two research questions.

(1) What would be the impact of hydrogen fuel cell vehicles on ambient concentrations of primary pollutants (NO_x, VOC, CO, and particulates, as well as SO_x), considering all the emissions involved in the full fuel cycle, including producing, transporting, and using hydrogen?

(2) What hydrogen supply strategy would be environmentally best for a specific county or region in the U.S.? For example, is onsite production at refueling

stations preferable to centralized production with delivery? Which delivery mode for centralized hydrogen is better, liquid truck or gaseous pipeline? and

(3) What would the optimal, feasible spatial layout of a hydrogen supply system be in a specific region, assuming that a possible hydrogen pathway type has been determined?

In this chapter, we develop hydrogen transportation scenarios for Sacramento, California, and estimate regional air quality impacts for three different hydrogen production and delivery pathways, based on steam reforming of natural gas (NG), which is currently the most common way of making hydrogen. Only primary pollutants that are directly emitted from emission sources are included, and no secondary atmospheric formation like secondary particulate matter or ozone (O_3) is considered in this chapter. Using a lifecycle analysis approach, this research compares these hydrogen supply options, presents the methodology to link hydrogen pathways to ambient air quality in urban Sacramento, California, and estimates the increases in ambient pollution corresponding to some key hydrogen supply chain steps.

2.2. Methodology

2.2.1. Overview of hydrogen pathway scenarios

From a lifecycle analysis (LCA) perspective, we estimate regional air quality impacts for three different hydrogen production and delivery pathways, based on steam methane

reforming (SMR) of natural gas, which is a commercially available technology for producing hydrogen today.

We examine distributed vs. centralized production. Distributed hydrogen is, in general, economically advantageous in the early hydrogen market, as hydrogen is produced onsite at the refueling station and no extra delivery is needed (Ogden, 1999a; Yang and Ogden, 2007; H2A, 2008). We also analyze the two important delivery modes for centralized hydrogen: gaseous hydrogen pipeline vs. liquid hydrogen truck. Centralized hydrogen tends to be more economically feasible at higher market penetrations of hydrogen (Ogden, 1999a; Yang and Ogden, 2007; H2A, 2008). A likely penetration pattern for natural gas-based hydrogen is: onsite SMR→ liquid truck SMR→ gaseous pipeline SMR.

In this chapter, the lifecycle emissions associated with each hydrogen pathway are used to determine the impact on urban air quality in Sacramento, California. Lifecycle emissions include all the emissions involved in producing and delivering hydrogen to vehicles, as well as emissions from electricity generation (for hydrogen compression or liquefaction) and petroleum use (diesel fuel for hydrogen truck delivery). Not only direct emissions resulting from the primary fuel pathway but also indirect emissions associated with secondary fuel pathways (sub-pathways) are taken into account.

To link hydrogen pathways to ambient air quality, we develop a methodological framework for hydrogen pathway scenarios, as shown in Figure 2 and described further in later sections.

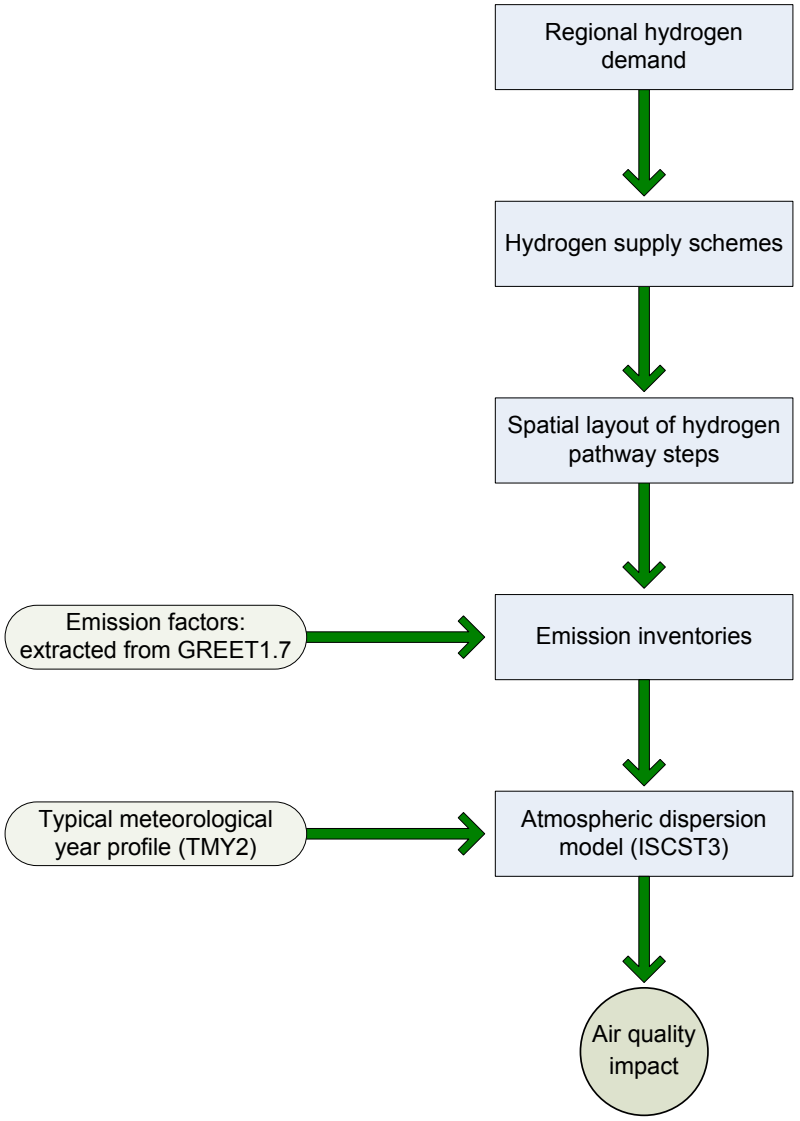

Figure 2. Methodological framework for hydrogen pathway scenarios

2.2.2. Estimating hydrogen demand

We use the urbanized Sacramento conventional light duty (LD) fleet in 2000 as the baseline. We consider two scenarios, where 9% and 20% of light duty vehicles in Sacramento are assumed to be hydrogen fuel cell vehicles (FCVs). We keep the number of gasoline vehicles constant in both scenarios, and add hydrogen vehicles and hydrogen

fuel supply systems to the Sacramento area. Thus, the total vehicle population is the sum of the year 2000 light duty gasoline fleet and the added hydrogen vehicles. This allows us to estimate the incremental impact of hydrogen energy systems on ambient pollution levels in the Sacramento area, without the complexities of simultaneously reducing the number of gasoline vehicles (see Section 2.3.1). Table 1 shows demographic data for Sacramento. From these and hydrogen vehicle assumptions we estimate regional hydrogen demand for vehicle use for each scenario (see Table 2).

In scenario 1, we add a number of hydrogen FCVs equal to 10% of the total LD fleet in urbanized Sacramento in the year 2000. In this study, we are not replacing gasoline vehicles with hydrogen FCVs; therefore, scenario 1 with 111,400 hydrogen vehicles would mean hydrogen FCVs operating at a market penetration of 9% ($=\frac{10\%}{1+10\%}$). In scenario 2, we add a number of FCVs equal to 25% of the urbanized Sacramento LD fleet in the year 2000. Similarly, Scenario 2 with 278,600 hydrogen vehicles would mean a hydrogen vehicle penetration of 20% ($=\frac{25\%}{1+25\%}$). The hydrogen demand is calculated for each case (the added 111,400 FCVs in scenario 1 and 278,600 FCVs in scenario 2).

Table 1. Demographic data for Sacramento

Parameters	Value
City Population in Sacramento (in 2000)[1]	1.393 million
LD gasoline vehicle ownership	0.8 vehicles/person
LD gasoline vehicle population (2000)	1.114 million

[1] Obtained from Population of U.S. Urbanized Areas from the 2000 U.S. Census (U.S. Census Bureau, 2006).

Table 2. Hydrogen vehicle assumptions and hydrogen demand

Parameters	Scenario 1	Scenario 2
Hydrogen FCV market penetration	9%	20%
Number of added hydrogen FCVs	111,400	278,600
Hydrogen fuel demand	78,000 kg/day	195,000 kg/day
Number of hydrogen stations	27	66
Fuel economy of the hydrogen FCV	60 miles/kg hydrogen	
Vehicle miles traveled (VMT)	15,000 miles/year/vehicle	
Hydrogen consumption	0.7 kg/day/vehicle	
Hydrogen station size	3,000 kg/day	
Liquid truck capacity	3,000 kg liquid hydrogen	

We consider only physical transport of conserved pollutants in this chapter. Thus, the assumed background ambient pollution levels do not influence the results for the incremental ambient concentrations due to hydrogen vehicles. Therefore, the above two market penetration scenarios (9% and 20%) can be presented in the following way. Scenario 1 explores the resulting concentrations of pollutants from a hydrogen system with 111,400 fuel cell vehicles, a number equal to 10% of the total LD gasoline fleet in urbanized Sacramento in the year 2000. Similarly, scenario 2 explores the resulting concentrations of pollutants from 278,600 fuel cell vehicles, a number equal to 25% of the total gasoline fleet in urbanized Sacramento in 2000.

Note that year 2000 was a somewhat arbitrary choice (based on available information at the time of analysis initially conducted) to provide a real-world basis for the number of hydrogen vehicles to analyze.

2.2.3. Hydrogen supply schemes

The delivered cost of hydrogen made from natural gas was examined in detail in many studies (Ogden, 1999a; Ogden, 1999b; Yang and Ogden, 2007; Mintz et al., 2006). The hydrogen supply cost via any pathway depends on many factors, e.g., the scale of supply, demand level, feedstock cost, and other key inputs (Leiby et al., 2006). A complete technical and cost analysis of hydrogen production and delivery is conducted by U.S. DOE's H2A program (H2A, 2008).

For each hydrogen scenario examined in this study, we assume that hydrogen supply meets a steady state regional hydrogen demand on a daily basis. The following three hypothetical hydrogen supply pathways, all natural gas-based, are investigated in the study, as they are likely to be the lowest cost near-term options of supplying hydrogen over the next few decades (NRC, 2004).

(1) The onsite pathway. Figure 3 illustrates onsite hydrogen production at the refueling station;

(2) The pipeline pathway. Figure 4 illustrates centralized hydrogen production with gaseous hydrogen pipeline delivery systems; and

(3) The truck pathway. Figure 5 illustrates centralized hydrogen production with liquid hydrogen (LH2) truck delivery systems.

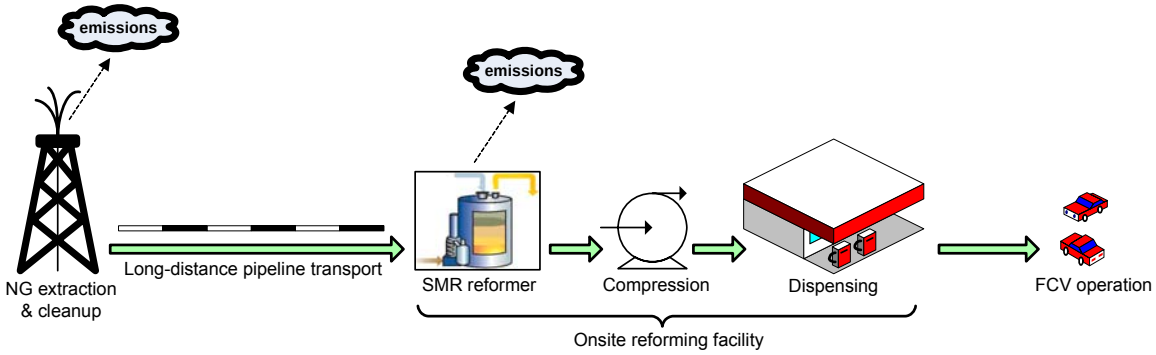

Figure 3. Natural gas-to-hydrogen pathway with onsite production

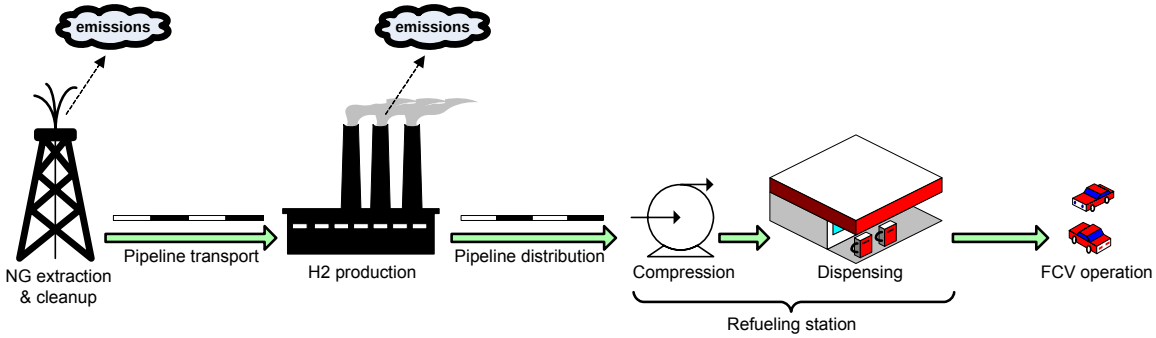

Figure 4. Natural gas-to-hydrogen pathway with pipeline delivery systems

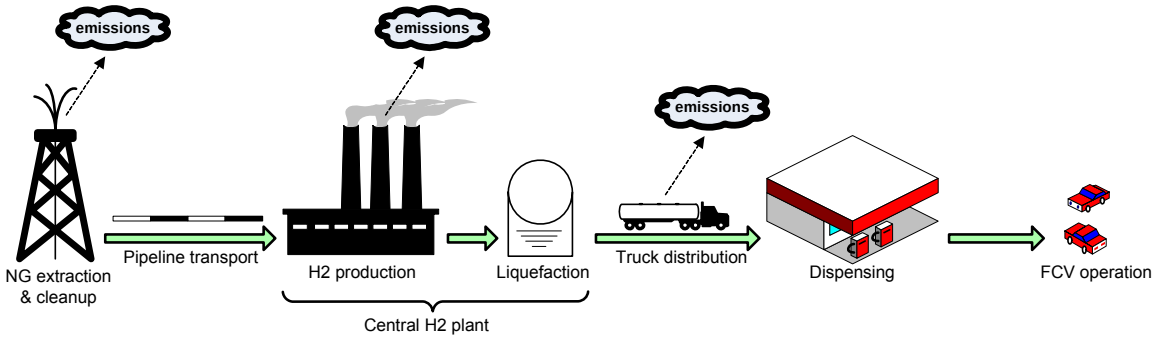

Figure 5. Natural gas-to-hydrogen pathway with liquid hydrogen truck delivery systems

For simplicity, emissions associated with electricity used for compressing, liquefying, or transporting hydrogen are not shown in Figures 3-5, although in our calculations we do add these emissions into the total. Further details on the hydrogen supply scenarios, e.g., the number of hydrogen stations, are presented in Table 2.

2.2.4. Spatial layout of hydrogen pathway steps

To estimate the environmental impacts of hydrogen vehicles, we consider all emissions associated with the system, including the following processes: feedstock extraction and transport; fuel production, storage, distribution, and dispensing; and vehicle operation (Wang, 1999).

Figure 6 presents the lifecycle of one of the three integrated natural gas-to-hydrogen pathways considered. The parts of the lifecycle system included in this analysis are enclosed by the dashed line (Wang and Delucchi, 2005). The parts of the system outside the dashed line are assumed to be either remote enough or low-emitting enough to have little or no impact on air quality in urban Sacramento. As described below, emissions from the pathways steps outside the “dashed lines” could impact air quality in regions outside the Sacramento area, with an attendant impact on human health. Thus, by focusing on the Sacramento region, our method somewhat underestimates the global impact of the hydrogen pathway.

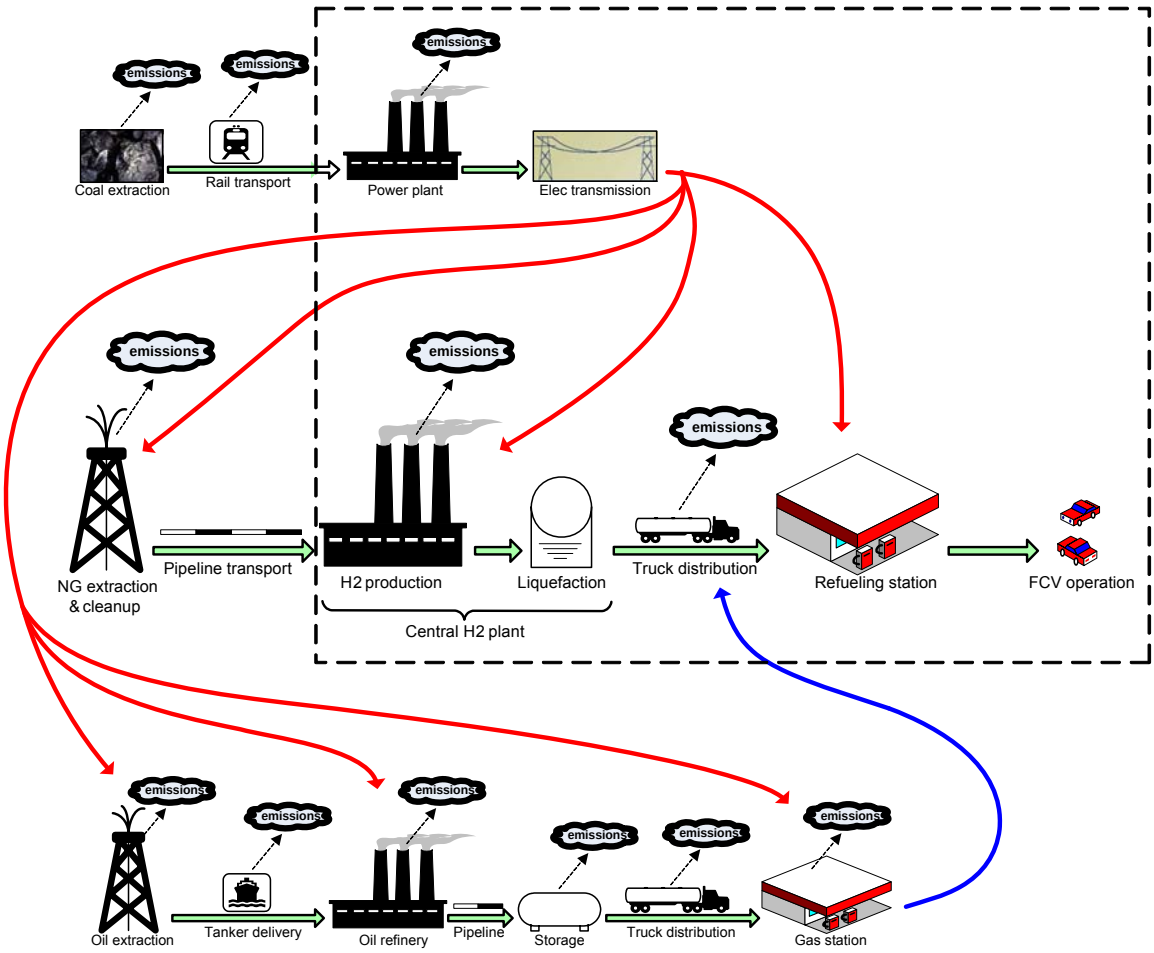

Figure 6. Integrated NG-to-H_2 pathways (liquid hydrogen illustration). (The upper figures represent the sub-pathway of electricity consumed in steps of the other fuel pathways. The lower figures depict the sub-pathway of diesel fuel used by the hydrogen-delivery trucks.)

The spatial locations of emission sources associated with the various hydrogen pathway supply steps have a strong influence on the regional air pollutant concentrations. In this study, we assume particular spatial locations for each step of the hydrogen pathway: natural gas extraction, hydrogen production, hydrogen delivery, and refueling stations, as well as hydrogen vehicle operation. The spatial layouts of the hypothetical stations and hydrogen plant are shown in Figures 7 and 8. Emission locations of each pathway step are described in detail below and summarized in Table 3.

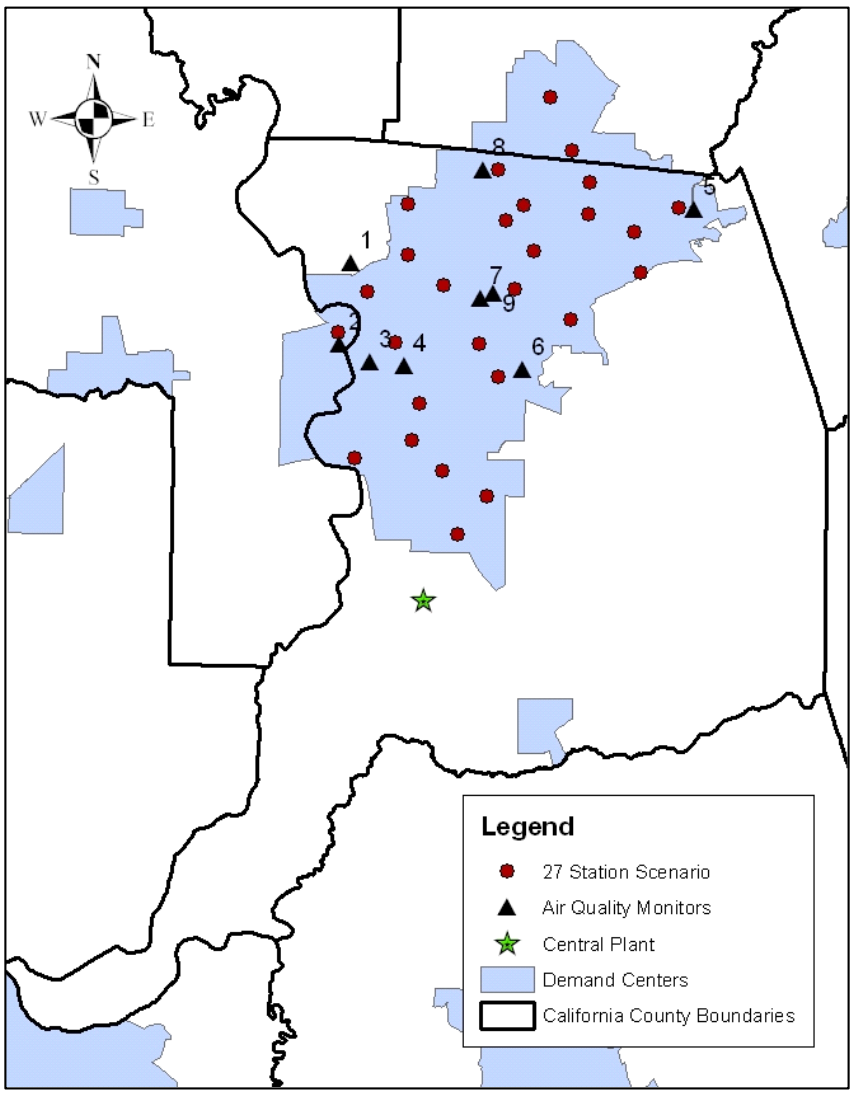

Figure 7. Spatial layouts of refueling stations, central plant, and air quality receptors in Sacramento (9% market penetration)

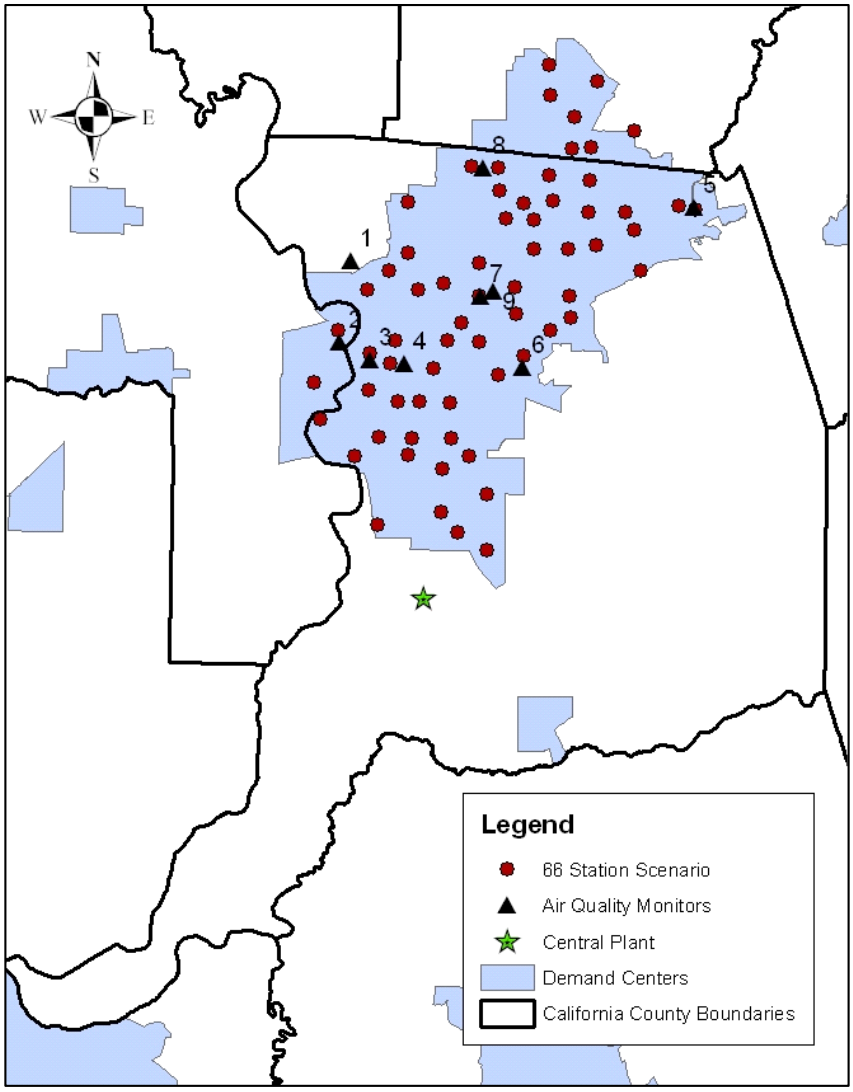

Figure 8. Spatial layouts of refueling stations, central plant, and air quality receptors in Sacramento (20% market penetration)

(1) Natural gas extraction and transport. This pathway step is not included in the research on air quality. Natural gas fields are located far away from Sacramento, and therefore the impacts of natural gas extraction and pipeline transport on air quality in urban Sacramento are neglected;

(2) Centralized hydrogen production. Because of the availability of natural gas, a central hydrogen production plant is assumed to be near currently existing natural gas-fired power plants in south Sacramento (see Figures 7 and 8), and is treated as a point source of emissions;

(3) Onsite hydrogen production at refueling stations. Emissions associated with hydrogen production from small steam reformers at refueling stations occur at the station sites. They are assumed to be a point source of emissions;

(4) Electricity for hydrogen liquefaction at the central plant. Actual locations of power plants in the Sacramento area are used to estimate incremental emissions associated with hydrogen liquefaction at the hydrogen plant. Although electricity is consumed at the central plant, the locations of emissions occur at those power plants which are assumed to be a point source;

(5) Electricity for hydrogen compression and pipeline delivery at the central plant. This is similar to electricity use for hydrogen liquefaction (see above);

(6) Liquid hydrogen truck delivery. Heavy heavy-duty diesel-fueled trucks (HHDTs) delivering liquid hydrogen are a mobile source of emissions. Liquid hydrogen trucks are assumed to travel on real-world highways and the actual route that each truck follows from the central hydrogen plant to the station is determined using geographic information system (GIS) data on a minimum travel time basis. The number of truck trips is estimated

based on the assumed station size and truck capacity (see Table 2). At the steady state, the road segments of the truck routes are treated as a thin-and-long area source of emissions;

(7) Refueling stations. We choose sites for hydrogen stations from among existing gasoline station locations in Sacramento. Hydrogen station sites are selected to minimize the average travel time from home to the closest station for all customers, given a certain number of stations. Customer locations are approximated using traffic analysis zones (TAZs). The method employs GIS data and optimization techniques and is described in detail by Nicholas (2004). The locations of stations in our study are shown in Figures 7 and 8. They are assumed to be a point source of emissions. Figure 7 corresponds to the scenario of a 9% market penetration and 27 refueling stations. Similarly, Figure 8 corresponds to the scenario of a 20% market penetration and 66 refueling stations;

(8) Electricity for hydrogen compression at refueling stations. To efficiently store and dispense gaseous hydrogen, some electricity is consumed at refueling stations. The treatment of emissions associated with electricity consumption at refueling stations is similar to electricity use for hydrogen liquefaction at the central plant (see above); and

(9) Vehicle operation. Hydrogen fuel cell vehicle operation is assumed to emit none of the air pollutants examined in this study (Ogden et al., 1999). Therefore, vehicle locations are not important for the analysis. Note that water vapor is the only emission if hydrogen

is used with fuel cells; in contrast, there are traces of NO_x generated if hydrogen is burned in air (Ogden, 1999b).

Table 3. Description of hydrogen pathway steps, locations, and emissions

Hydrogen pathways	Pathway steps included in the research[1]
Onsite hydrogen production	3(7)→8→9 In this case, steps 3 and 7 are essentially the same.
Centralized hydrogen production with pipeline delivery	2→5→7→8→9
Centralized hydrogen production with liquid hydrogen truck delivery	2→4→6→7→9

[1] The numbers in Table 3 refer to the pathway steps listed above in the text.

2.2.5. Lifecycle emission inventories

By using both emission rates and emission locations, we can develop spatially deterministic emission inventories which are one of the most important inputs to the subsequent air quality model. Only increases in air pollution due to emissions of primary criteria pollutants and ozone precursors are estimated; i.e. the focus is on the following directly emitted pollutants: carbon monoxide (CO), nitrogen oxides (NO_x, referring to both NO and NO_2), volatile organic compounds (VOCs, in some cases also called non-methane organic carbon, or NMOC), and particulate matter (PM_{10}, referring to particulates with an aerodynamic diameter less than 10 μm), as well as sulfur oxides (SO_x; this refers roughly to SO_2 here in the study). We do not account for re-entrainment of PM_{10} and particulates from tire wear and brake wear, too. (In the next chapter, we will

consider ozone production from its precursor emissions from hydrogen supply pathways examined in the study.)

Based on location information and emission source assumptions in the above sections, we consider the following direct emission sources: hydrogen plant or onsite production stations, electric power plants, and diesel-fueled delivery trucks (see Figure 6).

2.2.5.1. Hydrogen production infrastructure

To assess energy consumption and emissions of each hydrogen pathway step, models of emissions factors and hydrogen infrastructure engineering/economic designs are used. A full fuel cycle energy use and emissions model, GREET1.7, which is developed and maintained by the Argonne National Laboratory, is the source of those data on emission factors and energy consumption of hydrogen infrastructure such as the hydrogen plant or onsite production stations (GREET1.7, 2006; Wang, 1999). The technologies making up the hydrogen energy supply (e.g., hydrogen production, compression, or liquefaction) are assumed to have efficiency and emissions levels corresponding to current (year 2005) technologies (see Table 4 for efficiencies) (GREET1.7, 2006).

Because hydrogen systems will not be commercialized until a future year around 2020, we here compare the technology efficiency levels of hydrogen supply for 2005 vs. 2020, shown in Table 4 (GREET1.7, 2006). Table 4 indicates that most technology efficiencies will slightly increase over time. Therefore, it is conservative that we estimate the air

quality impacts of each hydrogen pathway using currently available energy supply technologies which correspond to the scenario year 2005 represented in GREET1.7.

Table 4. Hydrogen supply technology efficiencies for 2005 vs. 2020, on a lower heating value (LHV) basis

Key technology efficiency	2005			2020		
	Onsite pathway	Pipeline pathway	Truck pathway	Onsite pathway	Pipeline pathway	Truck pathway
Conversion efficiency	69.0%	71.5%	71.5%	70.5%	73.0%	73.0%
Compression efficiency[1]	94.0%	92.5%	N.A.	94.0%	92.5%	N.A.
Liquefaction efficiency	N.A.	N.A.	70.5%	N.A.	N.A.	72.0%

[1] Electric compressors apply to both the central plant and onsite stations.
[2] Efficiency data are extracted from GREET (GREET1.7, 2006).

2.2.5.2. Electric power plants

In this case study, we have chosen to neglect the impact of spatially distant pathway steps (such as natural gas extraction and oil refining) on air quality in Sacramento. However, we do consider emissions from the electricity used in hydrogen pathways steps. The emission factors for electricity consumption are extracted from GREET1.7.

Electricity consumed in both the primary hydrogen pathway and the sub-pathways is assumed to come from the average power mix for Sacramento. The electric generation mix in Sacramento is derived from the U.S. Department of Energy's eGRID2002 dataset for the year 2000 (eGRID, 2006). The power control area (PCA) of interest is specified as the Sacramento municipal utility district. There are 17 power plants serving the region and their profiles are shown in Table 5. The electric generation mix (i.e., percentage of

each kWh of electricity generated in 2000) by fuel type is summarized in Table 6. Clean renewables (i.e., solar, wind, and hydro) in Sacramento accounts for more than 42% of electric generation, which means the electric grid is less polluting than other regions when a large amount of electricity is consumed to compress or liquefy hydrogen.

Table 5. Sacramento PCA power plant profiles in 2000

Plant name	County name	Primary fuel	Generator capacity (MW)	Annual net generation (MWh)
CAMINO	EL DORADO	Hydro	154	429969
CAMP FAR WEST	PLACER	Hydro	7	31560
CARSON ICE CG	SACRAMENTO	NG	126	556594
HEDGE PV	SACRAMENTO	Solar	0.2	362
JAYBIRD	EL DORADO	Hydro	154	612984
JONES FORK	EL DORADO	Hydro	12	22297
KIEFER LF	SACRAMENTO	Biomass	9	74731
LOON LAKE	EL DORADO	Hydro	82	98011
MCCLELLAN	SACRAMENTO	NG, Oil	74	15743 (NG), 7 (Oil)
PVUSA	YOLO	Solar	1	253
ROBBS PEAK	EL DORADO	Hydro	30	49464
SCA	SACRAMENTO	NG	150	649213
SOLANO WIND	SOLANO	Wind	7	6774
SOLAR	SACRAMENTO	Solar	2	1887
SPA	SACRAMENTO	NG	174	1404149
UNION VALLEY	EL DORADO	Hydro	47	139504
WHITE ROCK	EL DORADO	Hydro	230	592124
PCA total			1257	4685626

Table 6. Sacramento PCA electric generation resource mix in 2000

Power plant type	Generation mix
Oil	0.0001%
Biomass	1.59%
NG	56.04%
Coal	0.00%
Nuclear	0.00%
Solar	0.05%
Wind	0.14%
Hydro	42.17%
Total	100.00%

2.2.5.3. Diesel-fueled delivery trucks

Heavy heavy-duty diesel-fueled trucks (HHDTs) delivering liquid hydrogen are considered as a mobile source of emissions. Liquid hydrogen trucks are assumed to travel along a fixed route from the central plant and arrive at a refueling station, and then come back along the same truck route. The emission factors of delivery trucks are from GREET1.7. For simplicity, the truck routes, which are determined by using a GIS-based optimization algorithm, are treated as a line source of vehicle exhaust. The number of truck trips is estimated based on the assumed station size and truckload capacity (see Table 2).

2.2.6. Atmospheric transport and urban air quality

We employ a complicated simulation model (ISCST3, introduced in later sections) for atmospheric transport of pollutants to estimate increases in pollutant concentrations in the

Sacramento area for each hydrogen supply case. No chemical transformation of pollutants is involved. We employ the spatial layouts in Figures 7 and 8 for the 9% and 20% market penetration scenarios, respectively. We estimate incremental concentrations at nine "receptor sites" in Sacramento, which are actual locations of air pollution monitors in EPA's monitoring network. These nine stations are shown in Figures 7 and 8 as triangles numbered 1 to 9. This allows us to compare the incremental changes in ambient concentrations due to hydrogen against actual measured ambient concentrations.

2.2.6.1. Atmospheric transport

We assume that each emission source in a hydrogen pathway emits pollutants at a constant rate. We further assume that pollutants disperse on an urban or regional scale, and the distance from an emission source to any air quality monitor of concern is less than 100 km, which assures that the above pollutants can be considered as conserved pollutants (ExternE, 2005). Studies by other researchers show that incremental annual concentrations are of much more interest than hourly or daily fluctuations, as they are more feasible and simpler to use to estimate yearly external costs associated with human exposure to ambient pollution (ExternE, 2005; Delucchi and McCubbin, 2004; McCubbin and Delucchi, 1996).

Only physical transport of the pollutants is taken into account, without considering chemical transformation or decaying of pollutants in the atmosphere. The equation below predicts the time-average concentrations downwind of an elevated point source,

accounting for superposition due to reflection from the ground (ExternE, 2005; Seinfeld and Pandis, 1998; Heath et al., 2005), shown as

$$C(x, y, z; H_E) = \frac{Q}{2\pi u \sigma_y \sigma_z} \exp\left[-\frac{y^2}{2\sigma_y{}^2}\right]\left\{\exp\left[-\frac{(z-H_E)^2}{2\sigma_z{}^2}\right] + \exp\left[-\frac{(z+H_E)^2}{2\sigma_z{}^2}\right]\right\},$$

(Equation 1)

where

H_E: effective stack height. H_E = physical stack height (h) + plume rise (ΔH);

$C(x, y, z; H_E)$: concentration of the pollutant at a receptor location (x, y, z);

Q: steady-state mass emission rate of the pollutant;

u: mean wind speed at the effective stack height. $u = x/t$, where t is the travel time of the pollutant from the release point to the location (x, y, z);

σ_y: transverse dispersion parameter. This is the standard deviation of the transverse concentration distribution at the downwind distance x; and

σ_z: vertical dispersion parameter. This is the standard deviation of the vertical concentration distribution at the downwind distance x.

2.2.6.2. The ISC model

To estimate atmospheric concentrations of pollutants, we run a steady state Gaussian plume dispersion model, Industrial Source Complex Short Term (ISCST3), maintained by U.S. EPA (ISCST3, 2006; U.S. EPA, 1995). It works directly for point, area, volume, and open pit sources of pollution, and by approximation to a sequence of long, thin area sources or volume sources, a line source of pollution can be simulated as well (U.S. EPA, 1995). It also can be used to assess air pollution from a variety of sources simultaneously. We use this model to estimate air quality at the receptor locations.

2.2.6.3. The TMY2 dataset

Like most air quality models, ISCST3 needs an annual cycle of local or regional meteorological information to predict the pollutant dispersion. The Typical Meteorological Year (TMY2), developed by the National Renewable Energy Laboratory (NREL), is a complete annual cycle of hourly meteorological data extracted from the 30-year period spanning 1961-1990 to represent a typical long-term meteorological condition in a specific region (TMY2, 2006). To run ISCST3, the hourly meteorological data such as the hour of day, wind direction, wind speed, ambient temperature, atmospheric stability class, rural mixing height, and urban mixing height are needed. The TMY2 dataset for Sacramento County is adopted in this research to predict changes in ambient air pollution under a historically representative meteorological condition rather than a worst-case condition (TMY2, 2006; Heath et al., 2005; Heath, 2005).

Figure 9 illustrates the Sacramento windrose for 2005 (including wind speeds and directions) (WRCC, 2008), and this windrose pattern is very typical of this region although it is not derived based on the TMY2 dataset. Note that the regional prevailing wind direction is from southwest to northeast, shown in Figure 9.

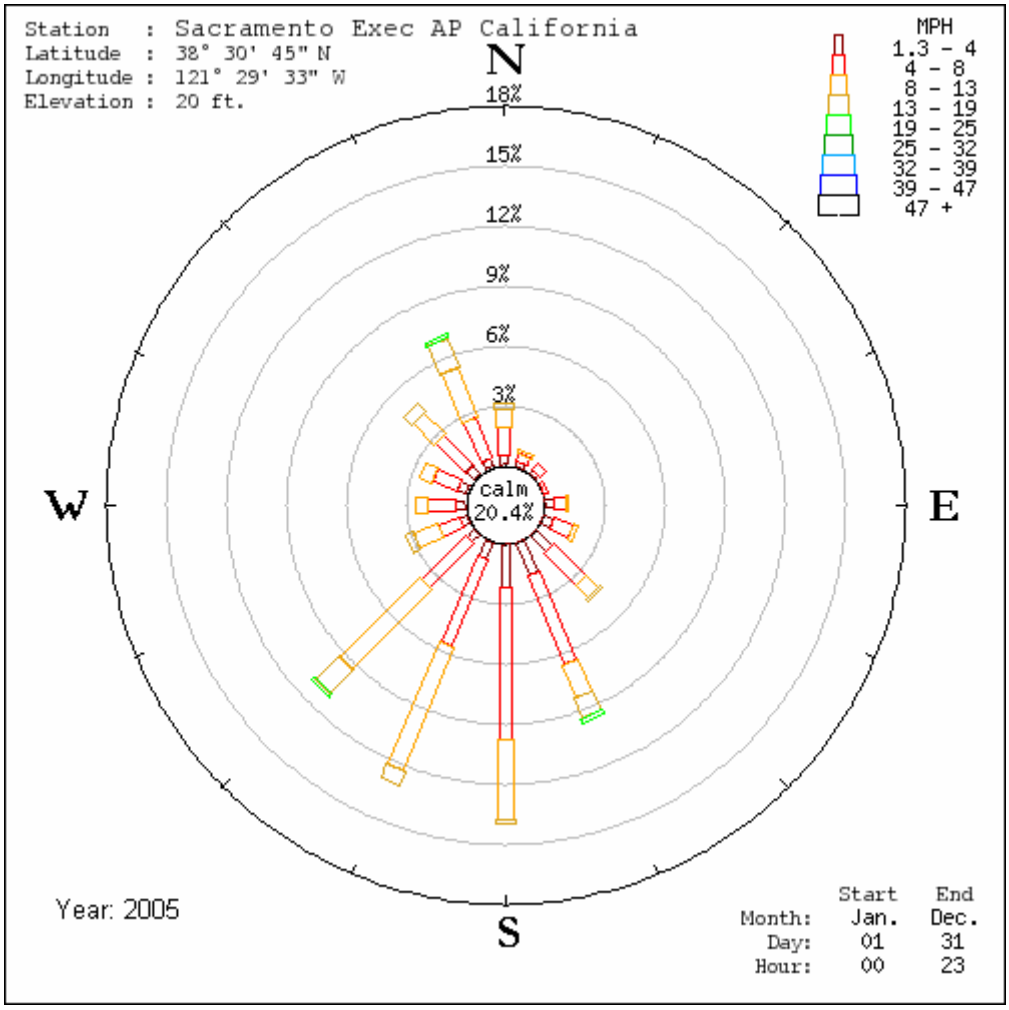

Figure 9. Sacramento windrose for 2005 (including wind speeds and directions)

2.2.6.4. The Air Quality System (AQS) for air pollution monitors

The Air Quality System (AQS) maintained by U.S. EPA contains ambient air pollution data and profiles of thousands of air quality monitoring stations throughout the country;

states, local and tribal agencies submit their data directly to AQS, and EPA itself also collects data (AQS, 2006). There are nine appropriate air monitoring stations officially maintained within or close to urban Sacramento based on the AQS system. These stations serve as receptors of pollutants in the research, and their profiles are shown in Table 7 (AQS, 2006). Figures 7 and 8 present their spatial layouts in Sacramento. The individual incremental concentrations at these receptors and their average values represent the ambient pollution level attributable to each of hydrogen pathways. Note, however, that a receptor is not necessarily a typical representative of urban air quality when it happens to be located very close to a truck route or a refueling station.

Table 7. Air quality monitors in urban Sacramento (i.e., receptors of pollutants)

Monitor	Name and address
1	Sacramento-3801 Airport Road
2	West Sacramento-15th Street
3	Sacramento-T Street
4	Sacramento-Health Dept Stockton Blvd
5	Folsom-Natoma Street
6	Sacramento-Branch Center Road
7	Sacramento-El Camino
8	North Highlands-Blackfoot Way
9	Sacramento-Del Paso Manor

2.2.6.5. The NAAQS standards and actual measurements

The Clean Air Act, last amended in 1990, requires U.S. EPA to set the National Ambient Air Quality Standards (NAAQS) to protect public health, and Table 8 shows NAAQS

primary standards (NAAQS, 2006; CARB, 2006). Table 8 also presents the actual measurements of pollution level in Sacramento in 2000, which is calculated based on the AQS dataset above. It is important to keep these ambient "baseline" concentrations in mind, as we discuss the incremental concentrations due to additional large numbers of hydrogen vehicles.

Table 8. NAAQS and ambient measurements in Sacramento in 2000

Pollutant	NAAQS (EPA,1990)		Sac. 2000 annual aver. conc. ($\mu g/m^3$)[1]
	Primary standards	Averaging times	
CO	9 ppm (10 mg/m^3)	8-hour	639.69
	35 ppm (40 mg/m^3)	1-hour	
VOC	No standards	N.A.	74.80 (NMOC)
NO_2	0.053 ppm (100$\mu g/m^3$)	Annual (Arith. Mean)	56.64
PM_{10}	50 $\mu g/m^3$	Annual (Arith. Mean)[2]	22.45
	150 $\mu g/m^3$	24-hour	
SO_x	0.03 ppm (80 $\mu g/m^3$)	Annual (Arith. Mean)	7.92
	0.14 ppm (365 $\mu g/m^3$)	24-hour	

[1] The Sacramento 2000 annual average ambient measurements are calculated based on data from U.S. EPA's Air Quality System (AQS).
[2] EPA revoked the annual PM_{10} standard in 2006 (effective December 17, 2006).

2.3. Results and discussion

2.3.1. Incremental pollution attributable to hydrogen pathways

We use the ISCST3 program to estimate the additional pollution at a receptor for each of our three hypothetical hydrogen pathways, at each of two market penetrations, 9% and 20%, respectively. Figures 10-19 present the magnitudes of incremental annual average concentrations of conserved pollutants due to existence of hydrogen pathways. There are

three pathways (i.e., the onsite pathway, the pipeline pathway, and the truck pathway), five pollutants (i.e., CO, VOC, NO_x, PM_{10}, and SO_x), and nine pollution receptors (denoted by R1 through R9). It is easy to see that environmental impacts vary with receptor site, which reflects the location variations and geographic factors, even when they are attributable to the same hydrogen pathway.

The first thing to note is that all three hydrogen supply pathways result in very small incremental amounts of pollution compared to ambient pollution levels, on the order of 0.1% increase in concentrations at the 20% market penetration, and often much less. This is in contrast to recent results for natural gas-based distributed generation of electricity in California, which resulted in more air pollution than central power plants (Heath et al., 2005).

For the truck pathway, emissions tend to be higher than for the other two supply pathways, though they are still small. As shown in Figures 20-24, most of the emissions for the liquid truck pathway are due to diesel truck emissions resulting from the delivery of the liquid hydrogen and to the electricity used to liquefy the product hydrogen.

For the onsite scenario, there are no hydrogen delivery emissions since all the hydrogen fuel is produced and dispensed onsite at the refueling stations. Also, the emissions are distributed throughout the metropolitan area so the wind direction has little impact on the average air pollution at receptors.

It can be seen in the following graphs that the change in air quality due to the onsite scenario is comparable to that caused by the central hydrogen pathway with pipeline systems, and both are very clean. The truck pathway also results in very low incremental pollution levels, but higher than concentrations resulting from the other two pathways.

Meteorological conditions, especially wind directions, have a large impact on the effect of emissions from the central plant. The prevailing wind in Sacramento is from southwest to northeast and seldom from east to west. The site of the central plant can be strategically located so as to minimize the effect on urban air quality. In our example, the site is somewhat advantageous in that it is only occasionally upwind of the urban area. The site for the central plant could be further improved by placing it east of the metropolitan area since this location is almost always downwind of the urban region. Furthermore, it would be meaningful to carry out a sensitivity analysis regarding the central hydrogen plant siting, although it is beyond the scope of this research. Again, geographic conditions have a significant effect on the impact of hydrogen production on urban air quality.

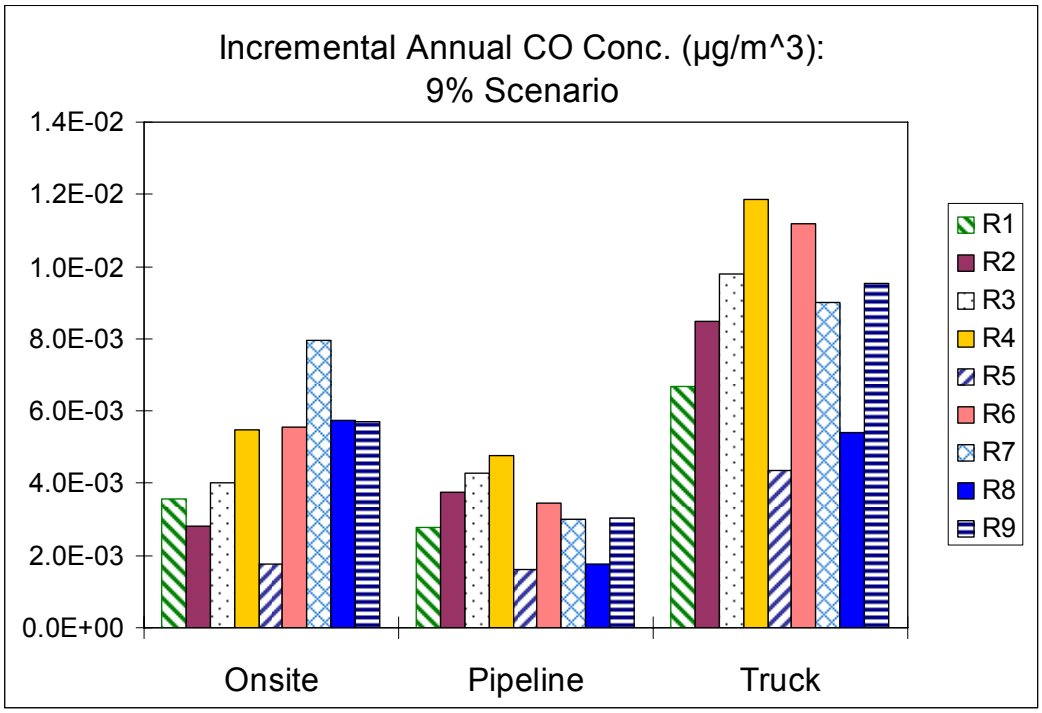

Figure 10. Incremental annual average concentrations of CO by receptor (9% scenario)

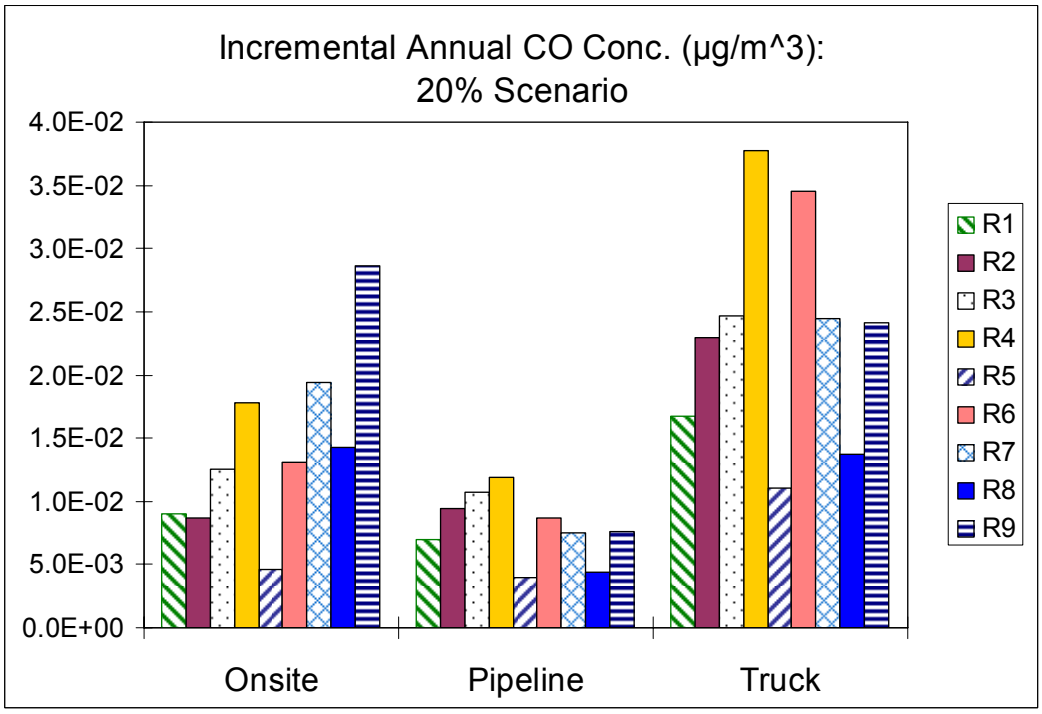

Figure 11. Incremental annual average concentrations of CO by receptor (20% scenario)

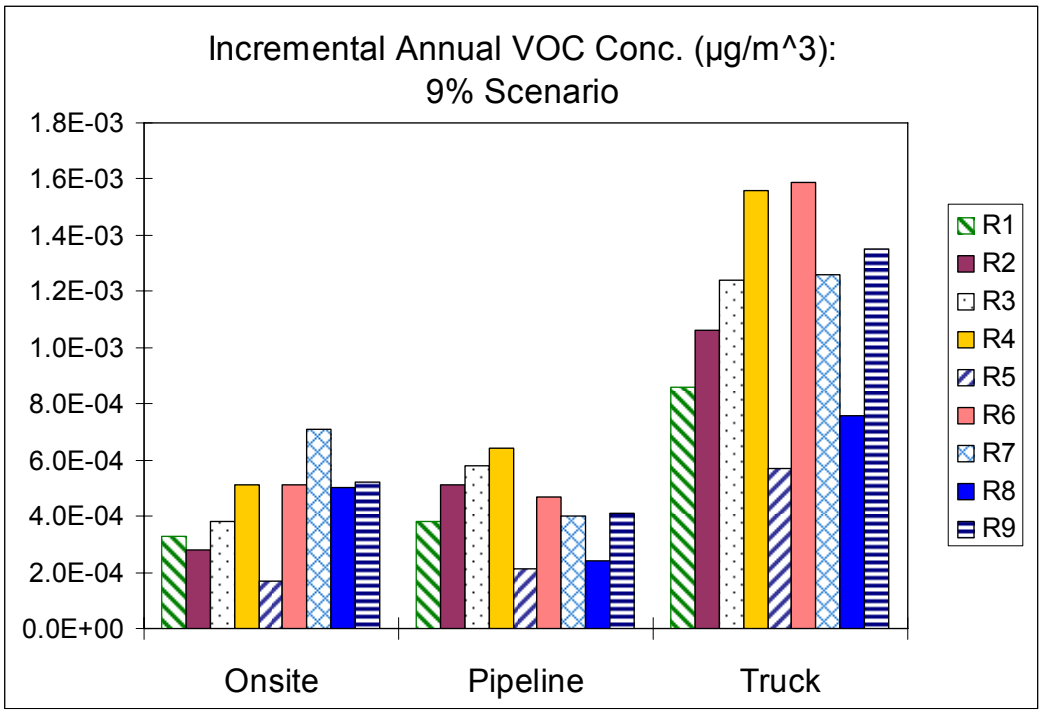

Figure 12. Incremental annual average concentrations of VOC by receptor (9% scenario)

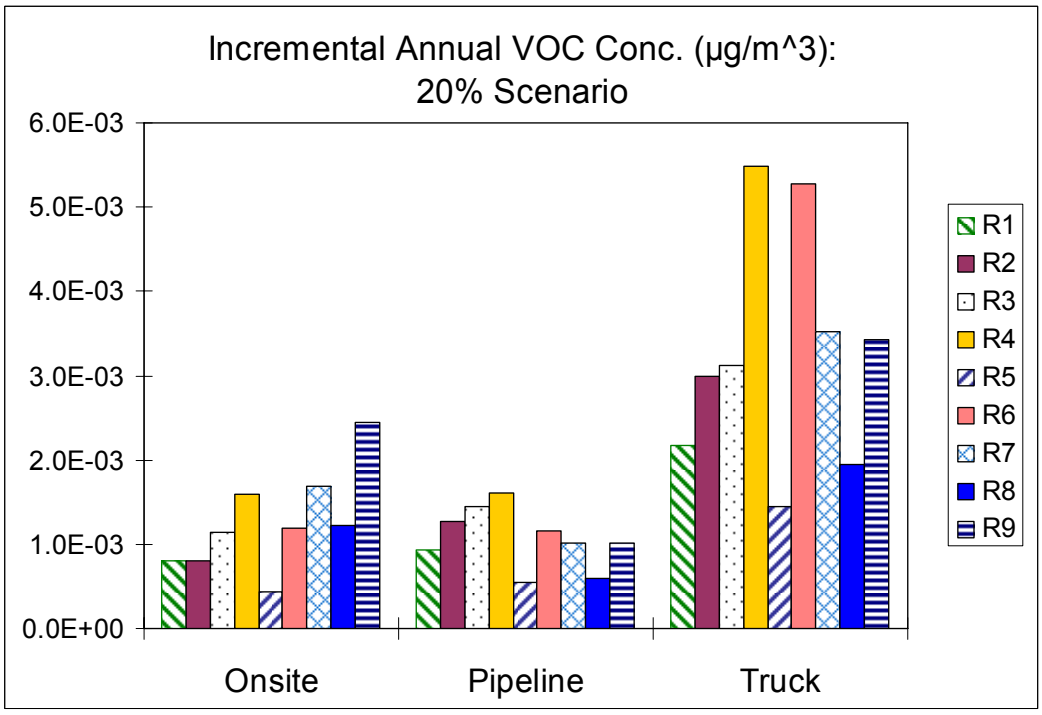

Figure 13. Incremental annual average concentrations of VOC by receptor (20% scenario)

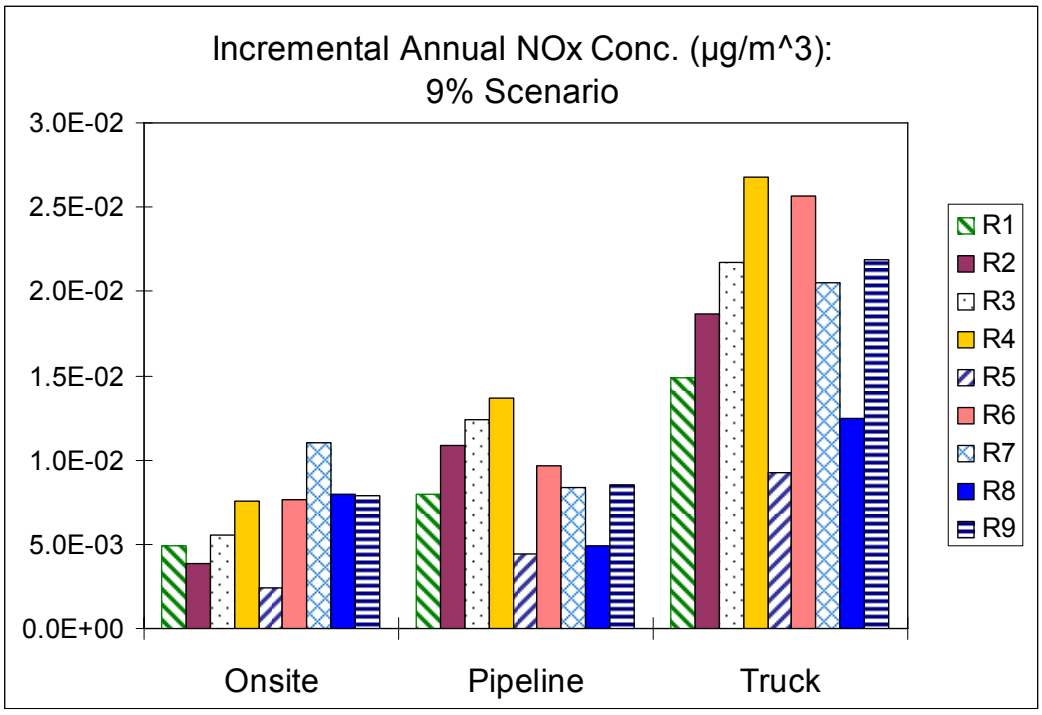

Figure 14. Incremental annual average concentrations of NO_x by receptor (9% scenario)

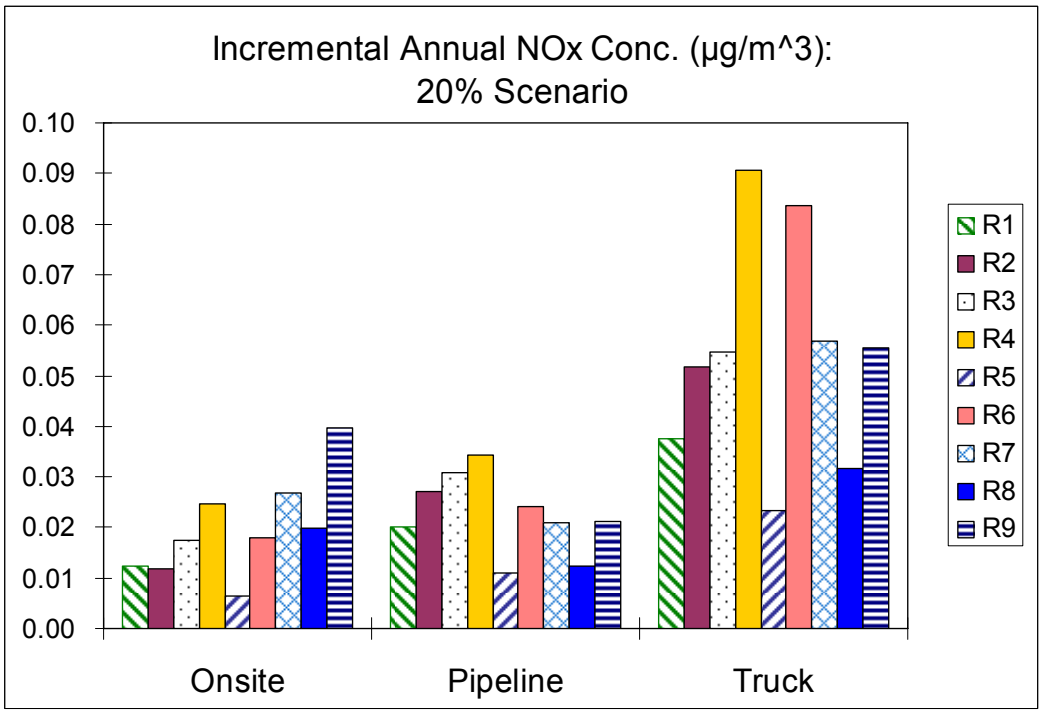

Figure 15. Incremental annual average concentrations of NO_x by receptor (20% scenario)

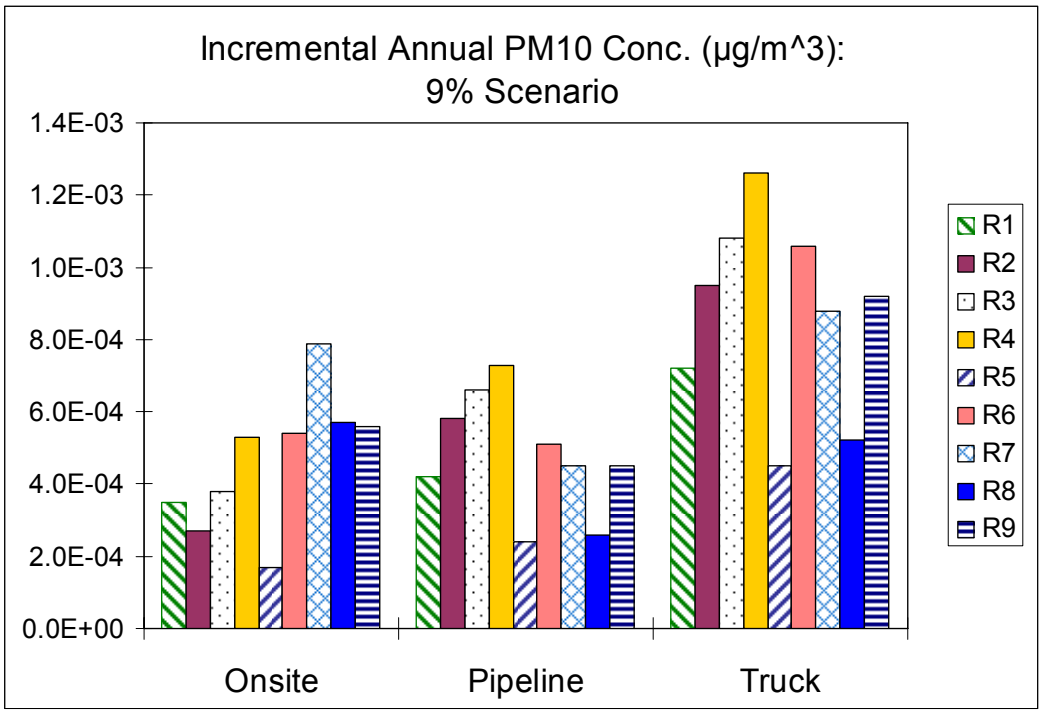

Figure 16. Incremental annual average concentrations of PM_{10} by receptor (9% scenario)

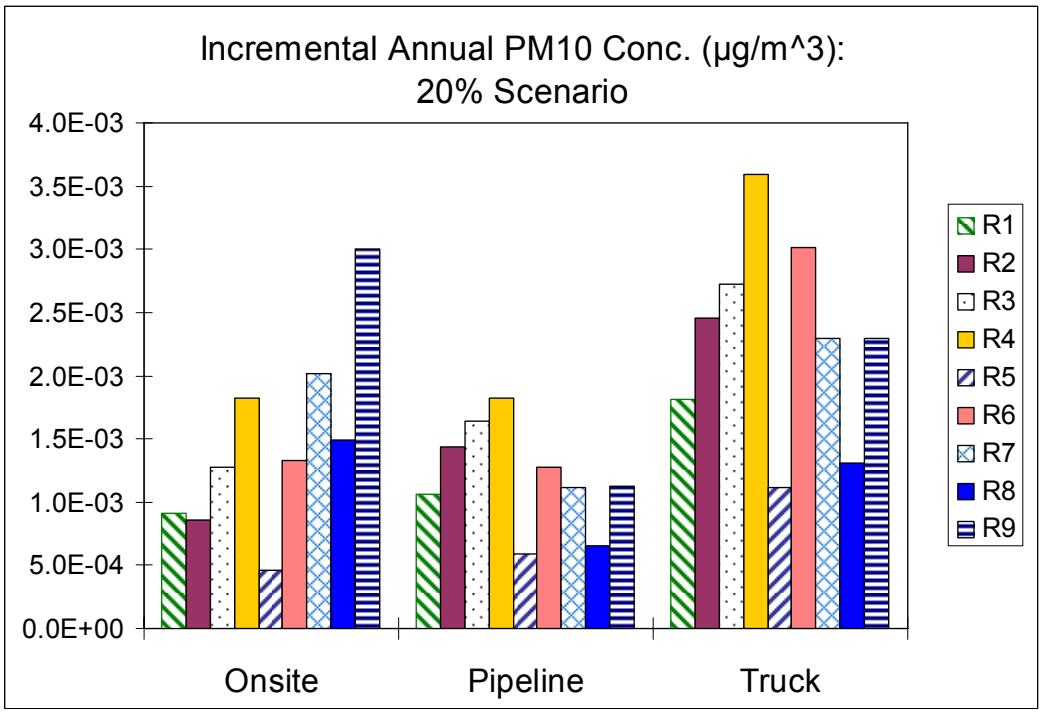

Figure 17. Incremental annual average concentrations of PM_{10} by receptor (20% scenario)

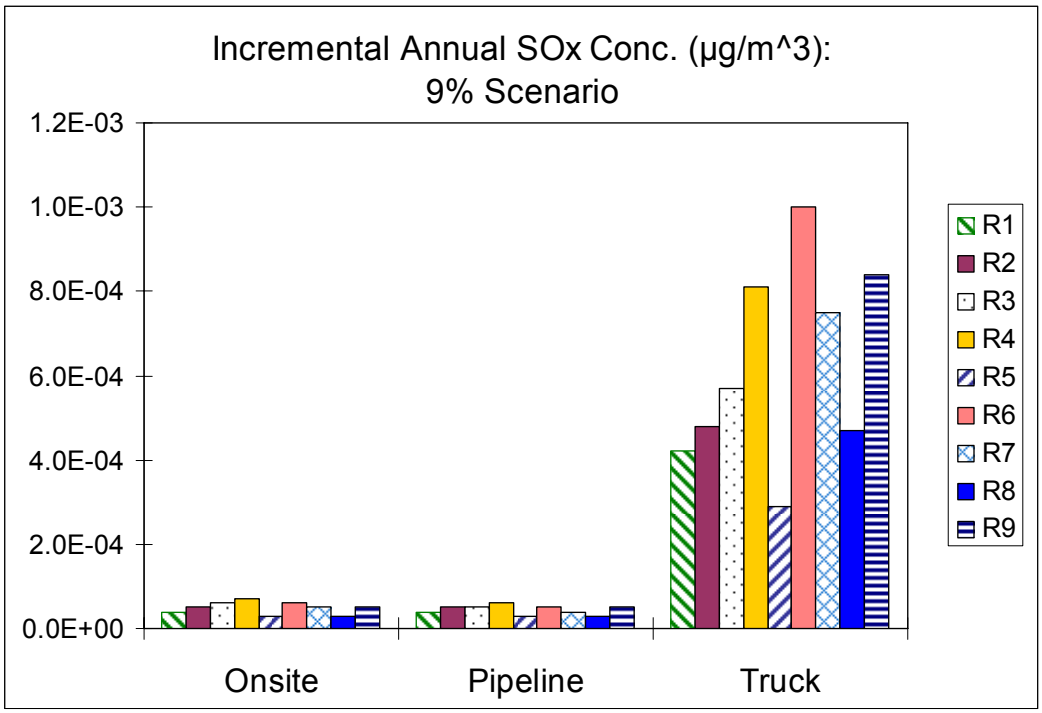

Figure 18. Incremental annual average concentrations of SO_x by receptor (9% scenario)

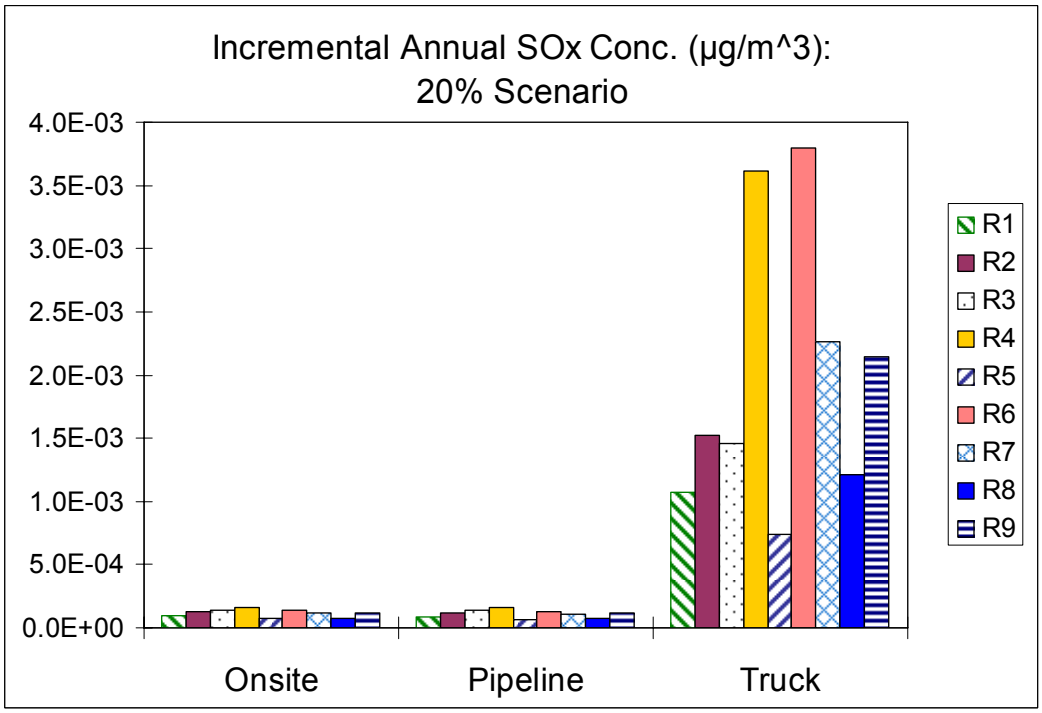

Figure 19. Incremental annual average concentrations of SO_x by receptor (20% scenario)

2.3.2. Comparison to the current ambient measurements

Table 9 compares the estimated incremental pollution, resulting from adding large numbers of hydrogen vehicles, to the actual measured mean concentration of pollutants in Sacramento, averaging over all the nine receptors. For the 9% market penetration scenario, the onsite pathway leads to incremental pollution fractions ranging from 0.0006% (SO_x, or approximately CO) to 0.0116% (NO_x) of current ambient concentrations. The pipeline pathway leads to pollution fractions ranging from 0.0005% (CO) to 0.0158% (NO_x), and the truck pathway leads to pollution fractions ranging from 0.0013% (CO) to 0.0337% (NO_x). For the 20% market penetration scenario, the onsite pathway leads to pollution fractions ranging from 0.0015% (SO_x, or approximately CO) to 0.0347% (NO_x), the pipeline pathway leads to pollution fractions ranging from 0.0012% (CO) to 0.0396% (NO_x), and the truck pathway leads to pollution fractions ranging from 0.0036% (CO) to 0.0952% (NO_x).

Relatively speaking, hydrogen pathways contribute the least fractions to ambient CO and the most fractions to ambient NO_x. This makes sense because most CO is released from on-road mobile sources (as compared to hydrogen supply pathways involving mainly stationary emission sources), whereas NO_x tends to be released from large stationary sources resulting from combustion, e.g., to produce the high temperature steam which is used in the central hydrogen plant or in a power plant.

In summary, for all scenarios, the incremental pollution due to adding hydrogen cars at a 9% or 20% market penetration is negligible. Truck pathways contribute more than onsite or central/pipeline pathways, but all lead to extremely low air pollution.

Table 9. Comparison of estimated incremental ambient pollution due to hydrogen pathways and ambient measurements

Pollutant	Market penetration	Onsite pathway		Pipeline pathway		Truck pathway	
		Mean conc. ($\mu g/m^3$)	Pollution fraction	Mean conc. ($\mu g/m^3$)	Pollution fraction	Mean conc. ($\mu g/m^3$)	Pollution fraction
CO	9%	0.00473	0.0007%	0.00316	0.0005%	0.00848	0.0013%
	20%	0.01423	0.0022%	0.00791	0.0012%	0.02331	0.0036%
VOC	9%	0.00043	0.0006%	0.00043	0.0006%	0.00114	0.0015%
	20%	0.00126	0.0017%	0.00107	0.0014%	0.00326	0.0044%
NO_x	9%	0.00654	0.0116%	0.00896	0.0158%	0.01909	0.0337%
	20%	0.01967	0.0347%	0.02241	0.0396%	0.05394	0.0952%
PM_{10}	9%	0.00046	0.0021%	0.00048	0.0021%	0.00087	0.0039%
	20%	0.00146	0.0065%	0.00119	0.0053%	0.00229	0.0102%
SO_x	9%	0.00005	0.0006%	0.00004	0.0006%	0.00063	0.0079%
	20%	0.00012	0.0015%	0.00011	0.0014%	0.00198	0.0250%

2.3.3. Further comparison among hydrogen pathways

Table 9 also shows a comparison of pathways in terms of resulting regionwide mean pollution. The truck pathway results in more pollution especially for SO_x, with concentrations more than an order of magnitude higher than those from the other pathways. Liquid hydrogen trucks fueled with sulfur-containing diesel make the biggest contribution to ambient SO_x concentrations. This is due to several factors: the trucks run on U.S. conventional diesel with an estimated sulfur mixing ratio of 200 ppm by mass

(GREET1.7, 2006); steam reforming of natural gas is very clean in terms of sulfur-containing emissions; and electric generation is relatively clean in Sacramento because renewables account for a very large share of production (see Table 6). The onsite pathway and the pipeline pathway result in very similar pollution levels, especially in terms of VOC, PM_{10}, and SO_x. However, the onsite pathway leads to more CO and less NO_x pollution than the pipeline pathway.

The incremental pollution due to each of hydrogen pathways, with the exception of the pipeline pathway, is not exactly proportional to the regional hydrogen demand, represented by the different hydrogen FCV market penetrations in this study (see Table 9). When the added hydrogen vehicle population increases by 2.5 times (from 10% of the year 2000 light duty fleet up to 25% of the fleet), the pollution ratio increases by slightly more than 2.5 times, with the exception of the onsite pathway, whose SO_x pollution is slightly lower than 2.5 times. For the pipeline pathway, it is 2.5 times greater because it is assumed that the NG to hydrogen conversion efficiency remains the same as hydrogen demand goes up, holding the electric generation mix constant.

2.3.4. Source contributions to incremental ambient pollution

Based on the locations of emissions, the sources of ambient pollution are categorized into the following groups (ignoring the other emission sources that are spatially far away from urban Sacramento).

(1) Hydrogen plant. This group includes the central hydrogen plant or onsite hydrogen production stations. Only emissions directly released at these locations are taken into consideration, and electricity consumed in a hydrogen plant is traced back to power plants that are referred to as another source contributor to ambient pollution;

(2) Power plant. This group includes all the 17 power plants that contribute to the electric generation mix in Sacramento; in fact, only 5 power plants contribute to the urban air quality since the other 12 power plants, accounting for 42.36% of power mix, are on a clean energy basis (i.e., solar, wind, and hydro). For simplicity, only emissions directly released at the power plants are taken into consideration, i.e. ignoring emissions upstream of power plants; and

(3) Truck route. This group only applies to the hydrogen pathway with liquid hydrogen truck delivery systems. The direct emissions are mainly diesel truck tailpipe emissions.

The source contributions to ambient pollution averaged over nine urban receptors of interest are presented in Figures 20-24. For the pipeline pathway, the hydrogen plant accounts for the largest share of pollution, and its contributions (for all the nine air quality monitors) are typically larger than 70%, and in some cases even larger than 80%. The exception is SO_x pollution, which is almost 100% from power plants.

For the onsite pathway, the hydrogen production stations account for the largest share, typically more than 70%. And again, SO_x pollution is the exception as power plants account for almost all of the SO_x pollution. Some receptors are affected by onsite stations much more severely, especially receptors that are next to and downwind from one or more stations. On average, hydrogen stations contribute around 90% of incremental pollution at receptors.

For the truck pathway, there are mainly three pollution components: the truck routes, hydrogen plant, and power plants. For all the five pollutants, truck routes and power plants are very important. The hydrogen plant contributes the smallest share, around 10% to 30%, and essentially 0% in terms of SO_x. Truck routes contribute 20% to 40% of pollution at a receptor, and particularly lead up to 70% in terms of SO_x pollution. Power plants contribute around 30% of pollution at a receptor for NO_x and SO_x, and they contribute around 50% pollution for the other three pollutants.

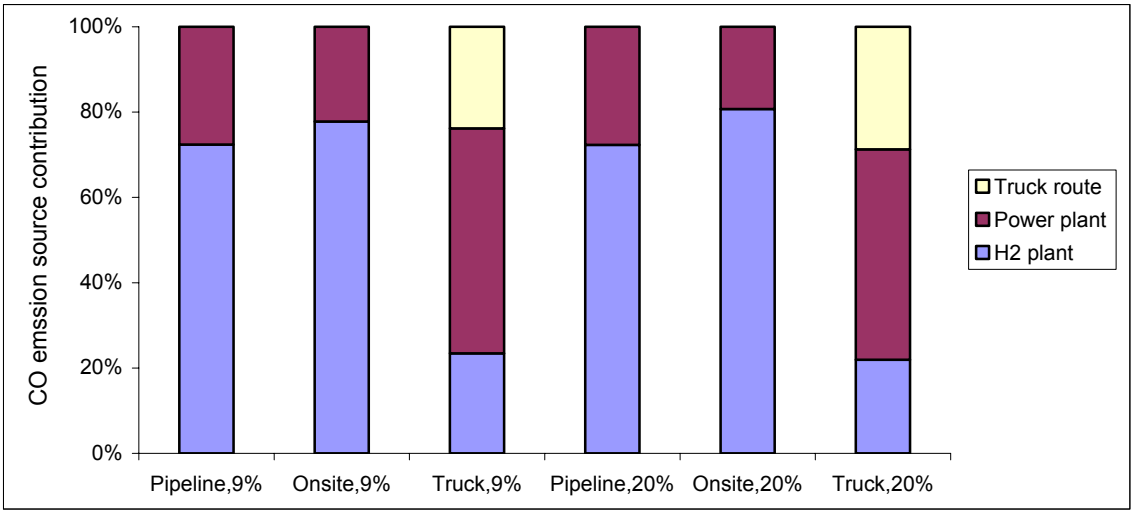

Figure 20. Source pollution shares averaged over all receptors (CO)

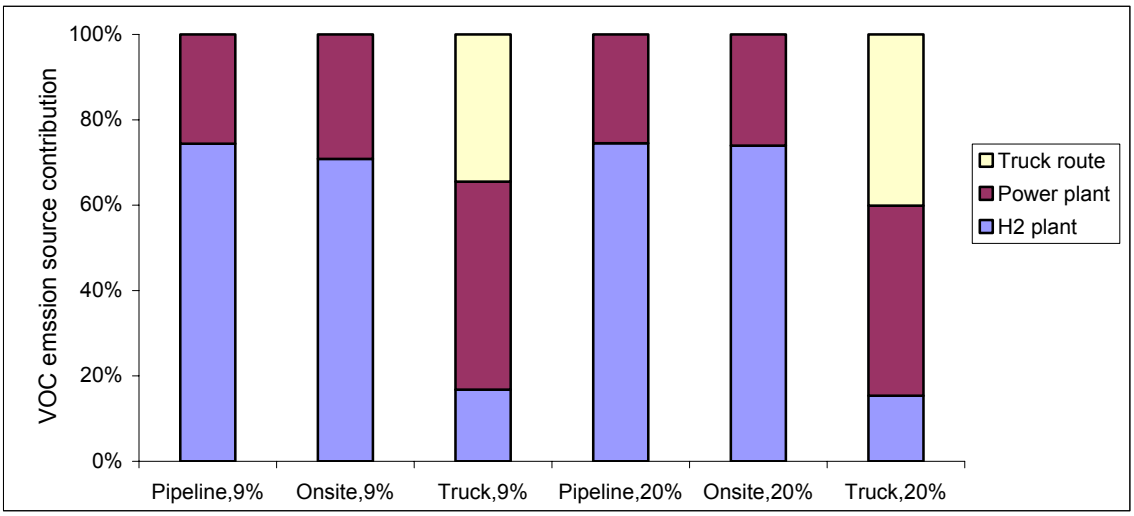

Figure 21. Source pollution shares averaged over all receptors (VOC)

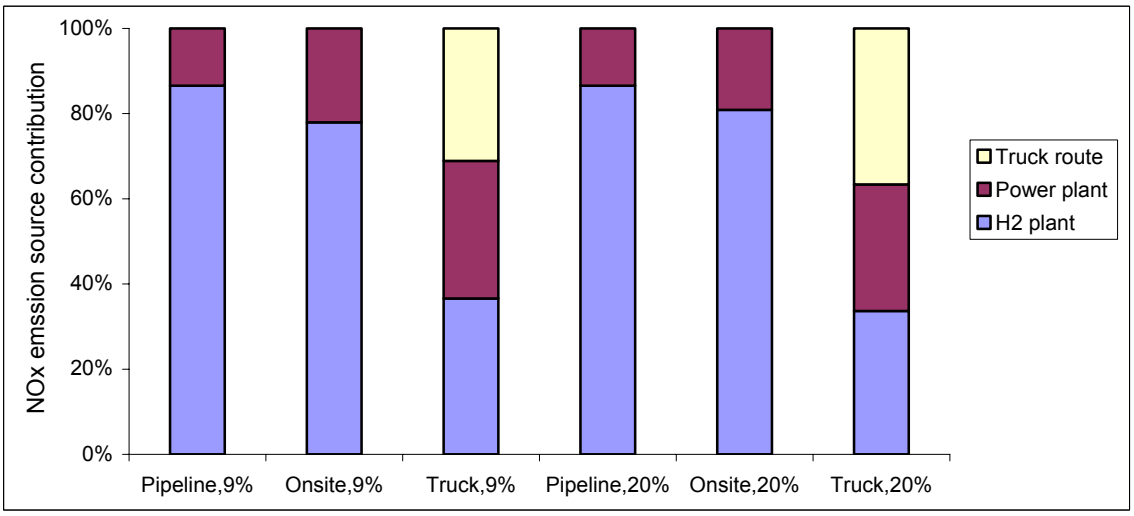

Figure 22. Source pollution shares averaged over all receptors (NO_x)

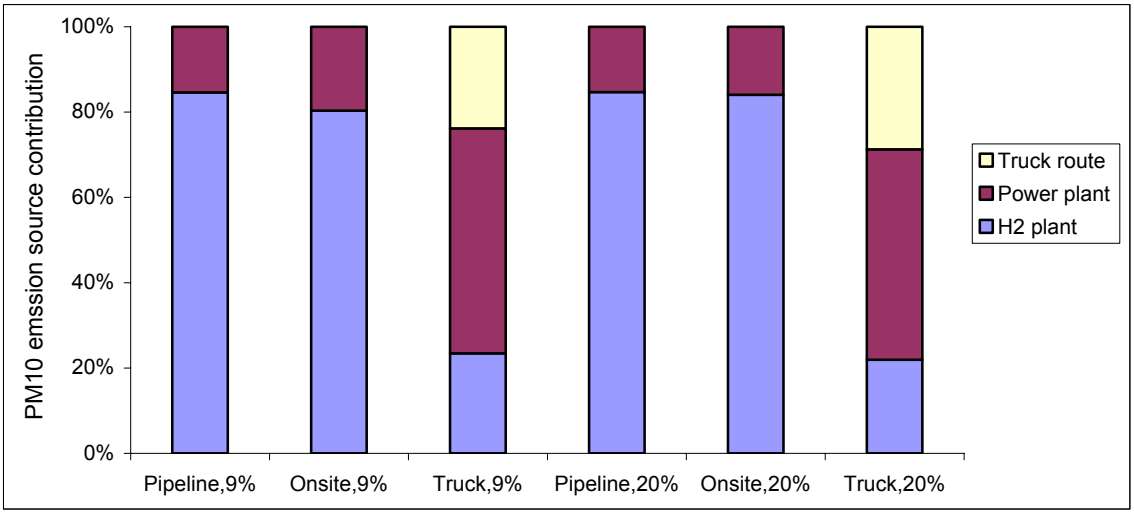

Figure 23. Source pollution shares averaged over all receptors (PM_{10})

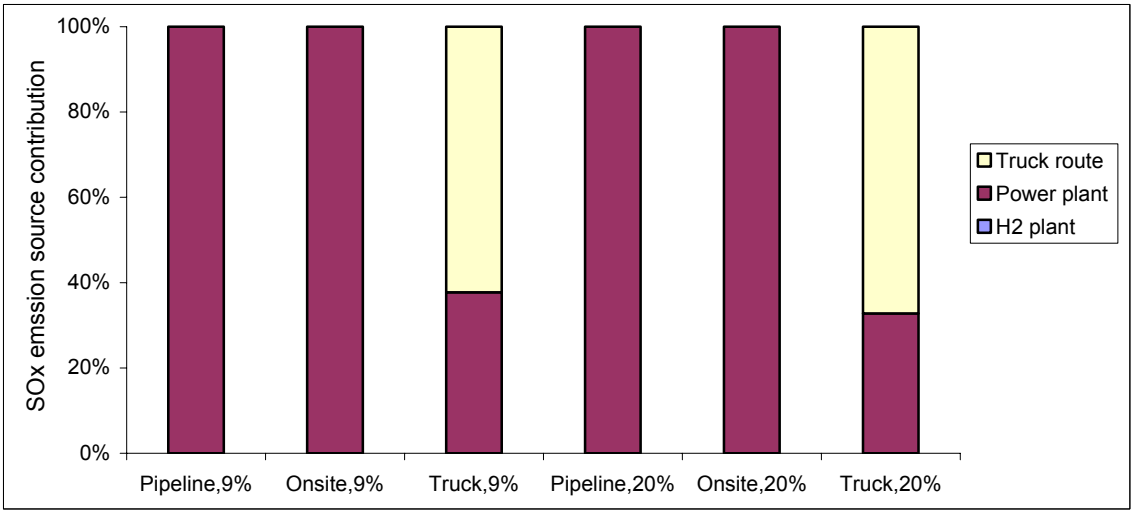

Figure 24. Source pollution shares averaged over all receptors (SOx)

2.4. Conclusions

We have examined the potential regional air quality impacts of hydrogen transportation fuel from a lifecycle analysis (LCA) perspective, including impacts from fuel production,

delivery, and vehicle use. The analysis is a case study for a specific region, Sacramento, California. We considered two levels of market penetration where 9% or 20% of the light duty fleet are hydrogen fuel cell vehicles. Three natural gas-based hydrogen supply pathways were considered: onsite production via small-scale steam methane reformer (SMR), large central SMR with liquid truck delivery, and large central SMR with pipeline delivery.

The contributions of each hydrogen pathway to ambient air pollution were estimated using a physical transport model for primary air pollutants NO_x, CO, VOC, particulates, and SO_x. (We used a Gaussian plume dispersion model for the atmospheric transport of pollutants. However, it does not include chemical production of secondary pollutants such as O_3 formed by the precursors VOC and NO_x in the presence of sunlight. Therefore, these results are not a complete air quality impact assessment of potential hydrogen pathways.)

The pollution levels associated with each of the hydrogen scenarios are dependent upon the location of emitters and receptors, regional meteorological conditions, and geographic factors. The spatial layout of pathway steps therefore plays an important role in determining ambient pollution levels at air quality monitoring stations. We find that all the hydrogen pathways considered are associated with extremely low pollution levels relative to current ambient air concentrations of NO_x, CO, VOC, particulates, and SO_x. For the 9-20% hydrogen scenarios examined, the results are typically less than 0.1% of the current ambient pollution.

Among the hydrogen supply options, it is found that central SMR with pipeline delivery is the least polluting option available provided the plant is located to avoid transport of pollutants into the city via prevailing winds. The onsite hydrogen pathway is comparable to the central hydrogen pathway with pipeline systems in terms of the resulting air pollution. The pathway with liquid hydrogen trucks has a greater impact on air quality relative to the other pathways due to emissions associated with diesel trucks and electricity consumption to liquefy hydrogen. The truck pathway causes more pollution than the onsite pathway and the pipeline pathway. The truck pathway causes around 15 times more SO_x, and around 3 times more of the other pollutants, than the other two pathways. For the pipeline or onsite pathway, hydrogen production accounts for the largest share of pollution. For the electricity-intensive liquid hydrogen truck pathway, emissions from diesel-truck delivery and electric generation at power plants are much more important than hydrogen plant emissions in terms of the resulting pollution. Again, compared to measured ambient concentrations, all the three hydrogen pathways result in negligible air pollution in the region.

In summary, the results in this chapter show that emissions from near-term hydrogen production and delivery systems would make negligible contribution to ambient urban air pollution. In later chapters, we compare the emissions from hydrogen systems to those from advanced gasoline vehicles.

3. Estimating Changes in Urban Ozone Concentrations Due to Lifecycle Emissions from Hydrogen Transportation Systems

3.1. Introduction

This chapter builds on methods and results from Chapter 2 to examine the potential impact of introducing a hydrogen-based transportation system on urban ambient ozone concentrations.

Hydrogen has been proposed as a low-polluting alternative transportation fuel that could help improve urban air quality. The tailpipe emissions of a hydrogen fuel cell vehicle (FCV) are strictly zero under all operating conditions (Ogden et al., 1999), but there would be some emissions related to hydrogen considering the full fuel cycle, including hydrogen production and delivery. These processes directly emit primary criteria pollutants and precursors to secondary ozone (O_3), including nitrogen oxides (NO_x, i.e., NO_2 and NO) and volatile organic compounds (VOCs, also called non-methane organic carbon, or NMOC). Ozone is of great concern because it is harmful to human health and agricultural crops and thus can become a social cost issue (ExternE, 1998; McCubbin and Delucchi, 1996; Delucchi et al., 1998; Murphy et al., 1999; Delucchi and McCubbin, 2004). In this chapter, we are addressing the following two research questions.

(1) What changes in peak ozone pollution would occur if typical hydrogen supply pathways were introduced in Sacramento, California, considering all the

emissions involved in the full fuel cycle, including producing, transporting, and using hydrogen? and

(2) What hydrogen supply strategy would be the best for the specific region, Sacramento, California, in terms of the resulting secondary ozone pollution?

In the previous chapter the three common natural gas-to-hydrogen pathways were examined, and the incremental pollution of NO_x, VOC, carbon monoxide (CO), particulate matter (PM_{10}), and sulfur oxides (SO_x, referring roughly to SO_2 here) was quantified based on the atmospheric physical transport of directly emitted pollutants. However, no atmospheric chemical reactions like ozone formation were considered. Going a step further, this chapter develops a region-specific regression model to predict atmospheric ozone formation. From a lifecycle analysis (LCA) perspective, the same three hydrogen pathways are compared in terms of the resulting changes in peak ozone pollution in urban Sacramento. Predictions of the potential ozone pollution caused by each of the hydrogen pathways are compared to the current ambient pollution levels.

3.2. Literature review on predictors of ozone formation

Ozone pollution and episodes mainly occur during the daylight hours of the summer months (NRC, 1991). In summary the “high ozone days” are likely affected by such parameters as the ground-level temperature, upper air temperature, dew point temperature, wind speed, wind direction, solar radiation or cloud cover, and relative humidity or precipitation (NRC, 1991). In addition to the meteorological conditions that lead to

ozone episodes, the characteristics and chemical composition of VOC have an impact on ozone formation, and different species of VOC differ in their photochemical ozone creation potential (Derwent et al., 1996). Prediction of ozone formation has improved over the decades with the development of three-dimensional photochemical transport models, but they often include a hundred or more coupled reactions just to describe gas phase changes along with detailed meteorology, and yet may only yield results accurate to about 25%. For the purposes of this study, a more efficient region-specific method of estimating the magnitude of the effects of different hydrogen pathways on ozone production was sought.

One such method might be to generate region-specific ozone isopleth data. Ideally, ozone isopleth diagrams can be produced by smog chamber experiments. In practice, the empirical kinetic modeling approach (EKMA) developed by the U.S. EPA relates the maximum hourly average ozone concentrations with the 6:00-9:00am average of precursor concentrations in a region, and both standard and city-specific ozone isopleths can be generated (Kinoslan, 1982). Figure 25 shows a typical pattern of ozone isopleths used in EPA's EKMA (NRC, 1991, citing Dodge's work). The NO_x-limited region in the ozone isopleth diagram is typical of locations downwind of urban and suburban areas, and in contrast the VOC-limited region is typical of highly polluted urban areas (NRC, 1991). From such studies it is recognized that the VOC and NO_x precursors to atmospheric ozone formation often yield a peak ozone when the VOC/ NO_x ratio is around 7-10 (Chang et al., 1989), and that ozone formation is retarded by additional NO_x emissions when the VOC/ NO_x ratio is less than 5.5 (Seinfeld and Pandis, 1998).

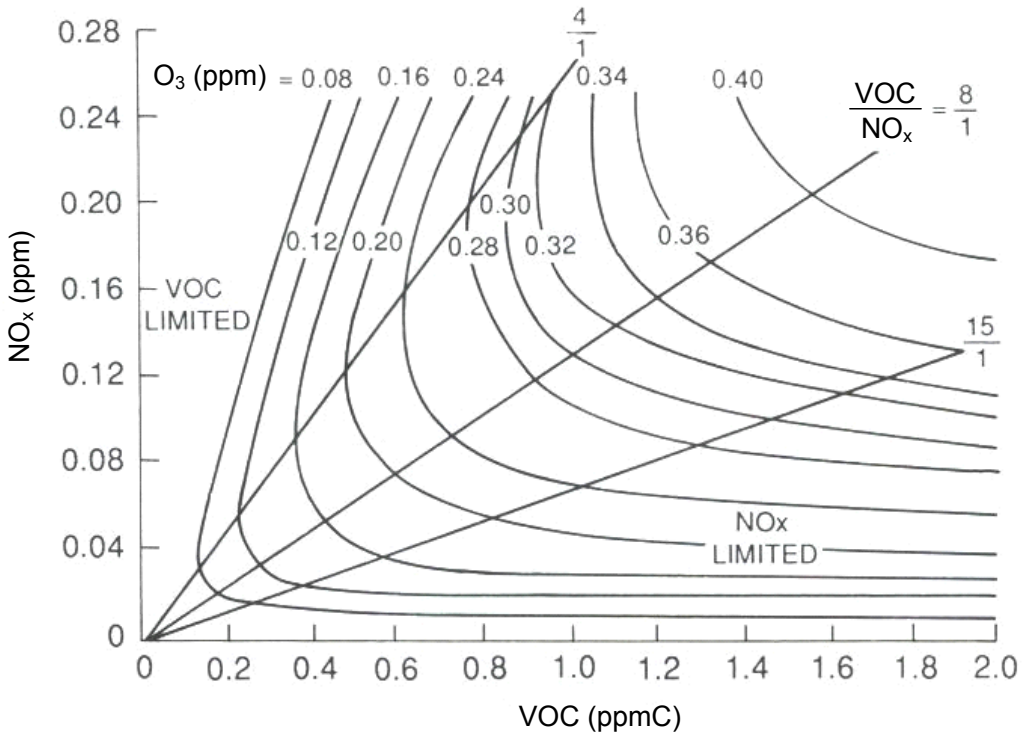

Figure 25. Typical pattern of ozone isopleths used in EPA's EKMA (NRC, 1991, citing Dodge's work). (Note that the values in the figure are not necessarily true in reality for all cities, as ozone isopleths are usually developed empirically for a specific region.)

Regression modeling approaches yield useful region-specific information when sufficient measurements are available. Based on daily air quality monitoring results for the downtown Los Angeles station for the months of August, September, and October, and using the 3-hour (6-9am) averages of total hydrocarbon and NO_x concentrations and the maximum hourly average oxidant concentration occurring on that day, an empirical model of ozone production was derived (Merz et al., 1972). Multiple regression modeling was also conducted to simulate the peak ozone produced by Los Angeles air in outdoor smog chambers, using the independent variables HC (i.e., hydrocarbon), NO_x, and the

average daily temperature (Kelly and Gunst, 1990). That study was the first published to quantify the effect of temperature on peak ozone formation in captive air studies (Kelly and Gunst, 1990).

3.3. Methodology

3.3.1. Overview of methodology

This research develops a region-specific regression model to predict atmospheric ozone formation associated with a hydrogen transportation system. Our methodology has several steps.

First, we estimate the emissions of ozone precursors from the various steps (and locations) along selected hydrogen supply pathways. We then use an atmospheric dispersion model ISCST3 to find the precursor concentrations throughout the Sacramento area, using data for typical meteorological conditions. For more details, see chapter 2. Next, we use the meteorological information and air quality data from 2004 to derive the relationship between ozone formation and its precursor concentrations by using regression analysis. Finally we use the regression-based model to estimate the incremental ozone concentrations due to hydrogen pathways.

This methodology should give a reasonable estimate of ozone concentrations in future years when hydrogen might be widely used, say in 2025 or beyond.[1]

3.3.2. Hydrogen pathway scenarios and dispersion model applications

Consistent with earlier chapters, this part of the research is built upon the same hydrogen pathway assumptions and methodological framework used for estimating primary pollution, resulting from physical transport only and without considering chemical transformation. See Chapter 2 for more details on the hydrogen pathway scenarios and air quality model applications.

We assume that in Sacramento, California, the current light duty gasoline fleet is held constant and still on the road in the same numbers, whereas additional hydrogen fuel cell vehicles have been introduced and are operating at market penetrations of 9% and 20%, respectively. Thus, the total vehicle population is the sum of the current light duty fleet

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[1] We used the 2004 ambient VOC and NO_x concentrations as the "baseline" from which changes were calculated on a daily basis. The rationale is as follows: first, there is no ambient air quality standard for VOC, and second the NO_x air quality in 2004 (see Figure 27) met the air quality standard. Thus, while one can try to project improved VOC and/or NO_x air quality in a future year, say 2025, based upon a state implementation plan (SIP), the degree of improvement is difficult to estimate since regional growth will likely reduce any gains due to emission control strategies. Also, we used daily air quality data to predict ozone for a given day and there is no good way to predict the VOC and NO_x concentrations on a daily basis in a distant future year.

To estimate the typical changes in VOC and NO_x concentrations, the year 2004 meteorology was not used; instead, a typical meteorological year (TMY2) was used, which was extracted for the region statistically from 1961-1990 and is the most representative meteorology (TMY2, 2006). The TMY2 is a complete annual cycle of hourly meteorological data extracted from the 30-year period to represent a typical, rather than a worst-case, long-term meteorological condition in a specific region.

The choice of using 2004 initial VOC and NO_x concentrations in conjunction with the TMY2 meteorology as inputs to the regression model could be considered "inconsistent," i.e., in the sense that a high initial VOC and NO_x concentration level could have been used on a day with good ventilation (meteorology), however running the analysis for an entire season averages out the impact of such occurrences.

and added hydrogen vehicles. Table 10 shows the two scenarios of estimated hydrogen demand for regional vehicle use. See Section 2.2.2 (in Chapter 2) for more details and discussions.

Table 10. Regional hydrogen demand for vehicle use (part of Table 2)

	Scenario 1	Scenario 2
Hydrogen FCV market penetration	9%	20%
Number of hydrogen FCVs	111,400	278,600
Hydrogen fuel demand	78,000 kg/day	195,000 kg/day
Number of hydrogen stations	27	66

We assume that in a steady state, hydrogen demand meets hydrogen supply on a daily basis. To meet the hydrogen demand, three hypothetical hydrogen supply pathways were considered in this study: onsite hydrogen production, centralized hydrogen production with pipeline delivery, and centralized hydrogen production with liquid hydrogen truck delivery.

The lifecycle emissions of ozone precursors associated with each hydrogen pathway are used to determine the impact on ozone production. Based on the lifecycle emission inventories and location information, a Gaussian dispersion model ISCST3 was run, together with Typical Meteorological Year (TMY2) data for the region as the meteorological inputs to the model (ISCST3, 2006; U.S. EPA, 1995). The TMY2 dataset is a complete annual cycle of hourly meteorological data extracted from the 30-year period spanning 1961-1990 to represent a typical, rather than a worst-case, long-term

meteorological condition in a specific region (TMY2, 2006). Therefore, the typical incremental concentrations of ozone precursors at each receptor (i.e., the air quality monitoring stations, shown in Figures 7 and 8) due to atmospheric transport of emissions associated with hydrogen pathways can be determined. The incremental VOC and NO_x concentrations were then added to the baseline VOC and NO_x concentrations and used to estimate subsequent ozone formation, though not necessarily in proximity to the origin of the emissions.

3.3.3. Data and the ozone regression model

The air quality data used in this research are selected from the Air Quality System (AQS), which is maintained by U.S. EPA and also contains profiles of many air quality monitoring stations throughout the country (AQS, 2006). However, the data on VOC and ozone are not complete for the year 2004. Even though more than 10 air quality monitoring stations were operating in Sacramento County in 2004, only one of them (see station 9 shown in Figures 7 and 8) has relatively good-quality VOC data available.

Figure 26 illustrates the Sacramento windrose for 2004 (WRCC, 2008), and this windrose pattern is very typical of this region. Note that the prevailing wind in the region is commonly in the direction from southwest to northeast in the summer months (see Figure 26), therefore station 5 is very often downwind of station 9, and it makes sense to assume that the early morning (say, 6:00 to 9:00am) pollution level at station 9 provides the initial VOC and NO_x concentrations that form ozone that reaches a maximum at the downwind station 5 (see Figures 7 and 8). This study uses data for 93 days in the summer

(most of the period July 3, 2004 through October 26, 2004), the season during which ozone pollution mainly occurs.

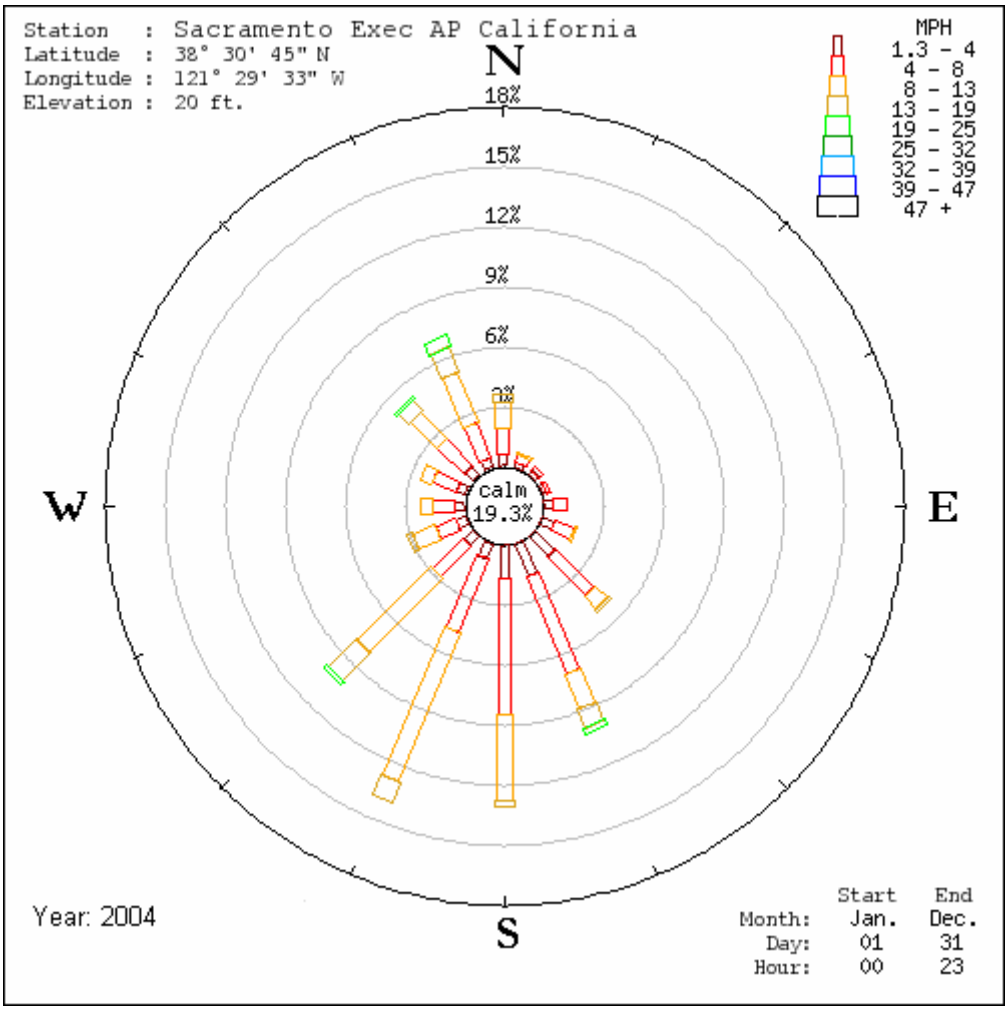

Figure 26. Sacramento windrose for 2004 (including wind speeds and directions)

An intrinsically linear regression model was developed to explore the relationship of ozone formation specific to the region. Consider initially that the peak ozone concentration is related to a number of factors, shown as

$$O_3(\max) = f(VOC, NO_x, Temp(\max), RH(avg), Solar\ radiation, Wind\ speed, etc.),$$

(Equation 2)

where

f: represents a certain functional relationship;

O_3(max): the peak ozone concentration ultimately reached (i.e., 1-hour maximum ozone concentration of the day) at the receptor station 5 (representing a location downwind of the urban area and often reflecting the maximum ozone pollution level observed in Sacramento), in units of parts per billion (ppb) by volume;

VOC (or NO_x): the initial ambient VOC (or NO_x) concentration at air quality monitoring station 9 (representing a typical central urban Sacramento pollution level). They are the 3-hour average of the ambient VOC (or NO_x) concentrations during 6:00 to 9:00am, in units of ppbC (or ppb) by volume;

Temp(max): the 1-hour maximum temperature of the day, degrees Celsius; and

RH(avg): the daily average relative humidity, %.

Using regression analysis, only four factors, namely the initial VOC concentration, initial NO_x concentration, maximum hourly temperature of the day, and daily average relative

humidity, were observed to be statistically significant and theoretically meaningful, and were therefore selected. The initial ambient VOC, the initial ambient NO_x, and the observed ambient peak ozone during the period of regression are shown in Figure 27, and the meteorological conditions used in the regression model are shown in Figure 28.

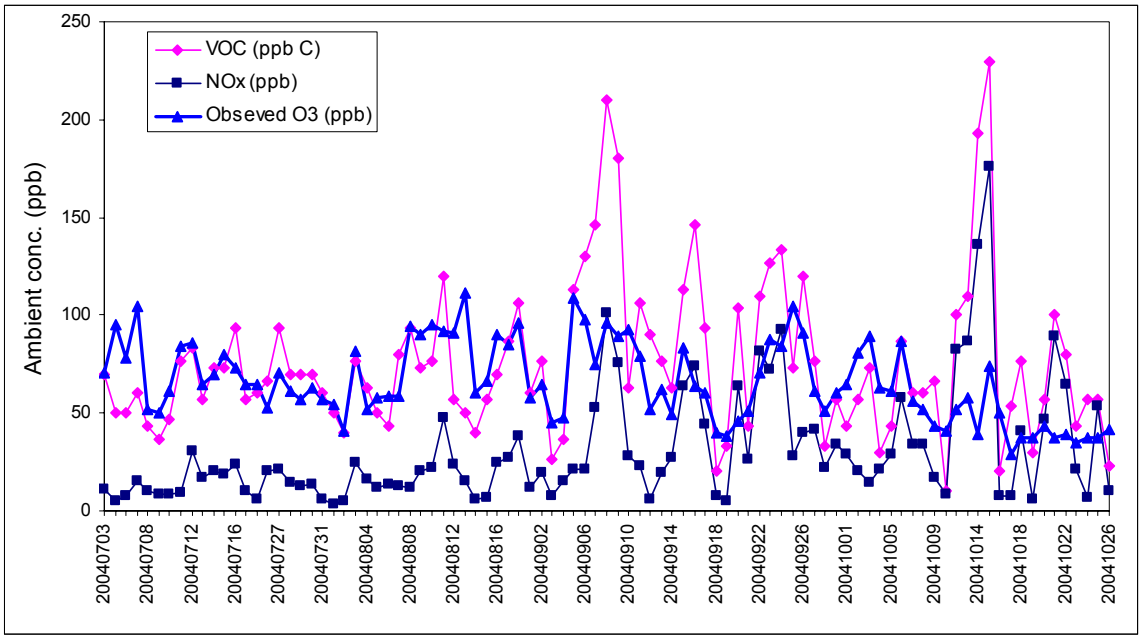

Figure 27. Actual initial ambient concentrations of pollutants used in the regression (6:00-9:00am)

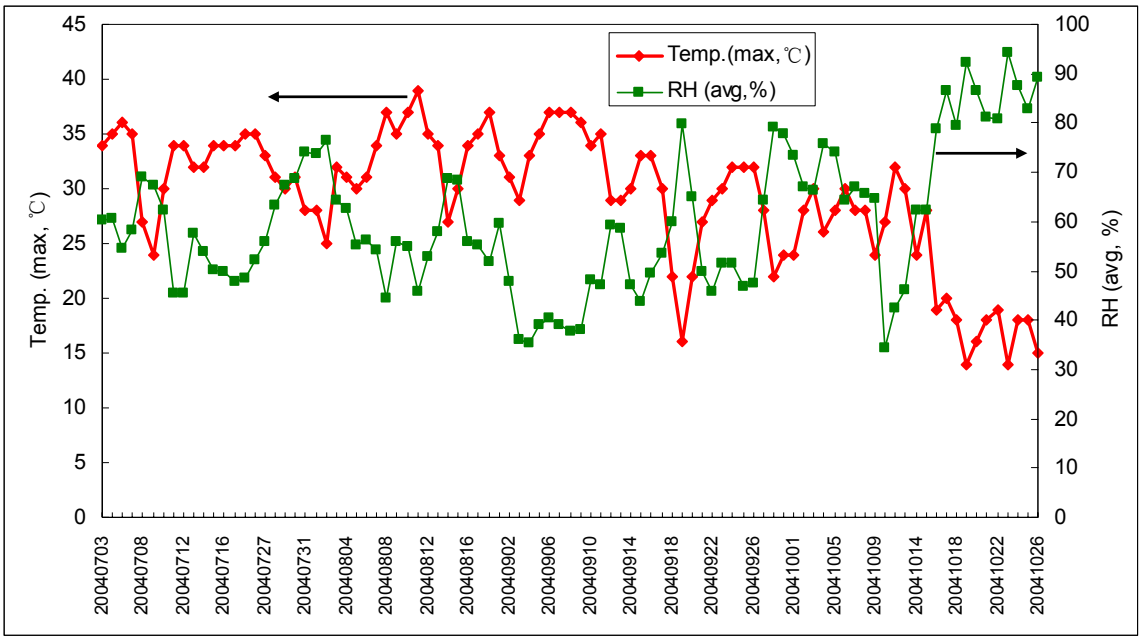

Figure 28. Actual meteorological conditions used in the regression

The following linear regression model (or equation) was estimated with Ordinary Least Squares (OLS) and ascertained to best correspond to all the groups of air quality data (see Equation 3).

$$O_3(\max) = -54.268 + 3.069\,Temp(\max) + 0.406\,RH(avg) + 0.474\,NO_x - 0.521\,NO_x^{\,2}/VOC$$
$$\text{(-2.644)} \quad \text{(7.628)} \qquad\qquad \text{(2.478)} \qquad\qquad \text{(2.376)} \qquad \text{(-2.017)}$$

(Equation 3)

The coefficient of determination is R^2=0.65 for the regression, and the sample size is N=93. The numbers in parentheses under the equation are t-statistics for the corresponding regression coefficients. The t-distribution table shows that at a significance level of 0.05, the critical value of t-statistic is 1.9873, with 88 degrees of freedom for the

above regression. That means if the magnitude of the t-statistic for a regression coefficient exceeds 1.9873, we can say with at least 95% confidence that the regression coefficient is significantly different from zero. The Durbin-Watson test gives DW=1.580, which provides no evidence of the existence of autocorrelation in the model specification using the time series data.

The regression is limited to the training dataset and hence to the corresponding region and time period. As we are only interested in the relationship between ozone and its precursors VOC and NO_x, the explanatory variables Temp(max) and RH(avg) are the control variables for our analysis.

The explanatory variable, Temp(max), is extremely important, and its coefficient is significantly different from zero (t-statistic is 7.628). The temperature effect is correlated with sunlight and also other meteorological conditions associated with the build-up of pollutants, e.g., low wind speed, so it is possible that several meteorological factors are included in Temp(max). However, the effect of RH appears to be largely separate from that of Temp(max), as model hypothesis testing and auxiliary regressions (equivalently, variance inflation factors, or VIFs) have not identified strong collinearity between Temp(max) and RH(avg) for this training dataset even though RH is often inversely related to temperature.

Based on the standard ozone isopleth diagram produced by EKMA (see Figure 25), there can be divergent predictions of ozone formation (increment or decrement) depending, to

some extent, on the ambient ratio of initial VOC to NO_x as the concentration of NO_x increases. This is reflected by the regressor NO_x^2 / VOC, which is equal to the NO_x concentration divided by the ratio of VOC to NO_x. The ratios for the training dataset happen to be within $0 < VOC / NO_x \leq 20$ (see Figure 29). There is no case in which the ratio VOC / NOx is greater than 20 in the dataset of the research, so the regression equation should perhaps not be applied to situations where $VOC / NO_x > 20$. Intuitively, the functional relationship where $VOC / NO_x > 20$ is very likely to be, or be close to, a straight line parallel to the VOC axis based on a standard EKMA ozone isopleth diagram when plotted in terms of initial VOC and NO_x (see Figure 25).

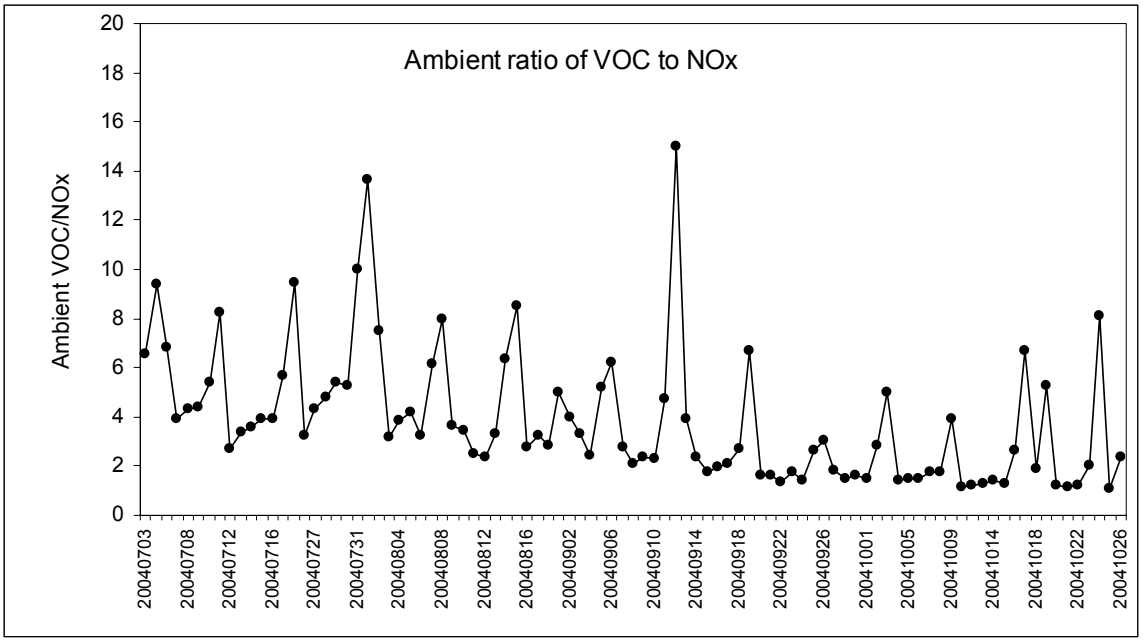

Figure 29. Ambient ratio of VOC to NOx at station 9 (6:00-9:00am). (Note that not all calendar days are present in the available data, due to lack of good-quality measurements of VOC and ozone. This study uses data for 93 days in the summer, i.e., most of the period July 3 - October 26, 2004, the season during which ozone pollution mainly occurs.)

The comparison of the observed and predicted peak ozone concentrations at receptor station 5 are shown in Figure 30. Even though the prevailing wind commonly blows from southwest to northeast in the summer, due to the wind speed/direction variability the geographical location where peak ozone concentrations occur may differ from receptor station 5 (as can be observed from photochemical grid model simulations), and thus the predicted concentration at station 5 is simply an approximation of the observed or theoretical concentration derived from smog chamber experiments or chemical reaction mechanisms. That in part explains why R^2 is not higher in the regression; that is, about 65% of the variation in peak ozone concentrations can be accounted for by the regression variables selected, but the regression cannot account for the day-specific variation of spatial transport of the predicted emissions to receptor 9 by the ISCST3 model or of subsequent ozone formed by those emissions to receptor 5.

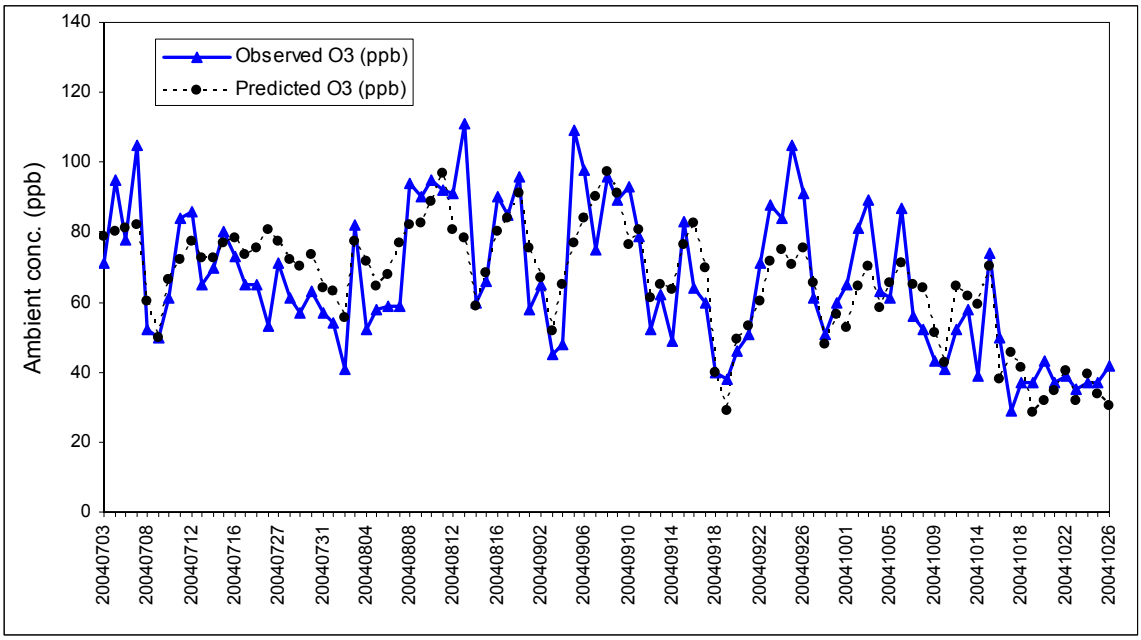

Figure 30. Comparison of the observed and predicted peak ozone concentrations

3.3.4. Applying the regression model

Three hydrogen supply pathways are considered, and for each of them there are two sets of market penetrations: 9% and 20%. Therefore, there are a total of six scenarios. Based on a previous study (see Chapter 2), the changes in ambient concentrations of primary pollutants, including VOC and NO_x, at monitoring stations have been determined. Below are the steps to estimate the changes in ozone air quality due to lifecycle emissions of each hydrogen pathway. We should sequentially:

(1) Estimate the incremental VOC and NO_x concentrations (i.e., the 3-hour average of 6:00-9:00am or the daily average, at station 9), caused by atmospheric physical transport and associated with each of six hydrogen supply scenarios. This step is accomplished using atmospheric dispersion models along with typical meteorological year data. No atmospheric chemical transformation is considered and only directly emitted primary pollutants are investigated for this step; therefore, secondary ozone pollution has not been included so far in the analysis. The percentage change in VOC or NO_x is expressed as Equations 4 and 5.

$$\%\Delta VOC = \frac{new\,VOC - baseline\,VOC}{baseline\,VOC} \times 100\% = \frac{incremental\,VOC}{baseline\,VOC} \times 100\%,$$

(Equation 4)

$$\%\Delta NO_x = \frac{new\ NO_x - baseline\ NO_x}{baseline\ NO_x} \times 100\% = \frac{incremental\ NO_x}{baseline\ NO_x} \times 100\%,$$

(Equation 5)

(2) Add the incremental VOC and NO_x to the baseline VOC and NO_x concentrations (i.e., the current ambient background VOC and NO_x in 2004), respectively, and use the sum as inputs to the ozone-and-precursors regression model developed in this study (see Equation 3);

(3) Calculate the new peak ozone concentrations day by day using the same meteorological data as in the regression model, as we treat meteorological factors as control variables. The calculated results are for station 5 (see Figures 7 and 8); and

(4) Compute the difference between the new ozone and the previously predicted ozone that is estimated using ambient background VOC and NO_x as inputs to the regression model. Now, the changes in ozone air quality, denoted by $\Delta O_3(\max)$, and the percentage changes in peak ozone levels, denoted by $\%\Delta O_3(\max)$, associated with a hydrogen pathway can be determined. Below are the formulas (see Equations 6 and 7).

$$\Delta O_3(\max) = new\ O_3(\max) - baseline\ O_3(\max), \qquad \text{(Equation 6)}$$

$$\%\Delta O_3(\max) = \frac{new\ O_3(\max) - baseline\ O_3(\max)}{baseline\ O_3(\max)} \times 100\%, \qquad \text{(Equation 7)}$$

Note that the changes in initial VOC and NO_x are small relative to the baseline pollution level, so it makes sense to apply the regression model to these "new" input data since they remain within the range of observations in the region.

3.4. Results and discussion

3.4.1. Incremental 3-hour average pollution of ozone precursors

The incremental 3-hour average concentrations of VOC and NO_x at receptor station 9 are shown in Tables 11 and 12. Those numbers represent additional pollution, caused by lifecycle emissions from six hypothetical hydrogen pathways respectively and occurring at station 9, which subsequently results in ozone pollution at station 5 commonly downwind of station 9. Relative to the actual ambient pollution level at station 9, Figures 31-34 compare the incremental 3-hour average pollution of physically transported VOC and NO_x associated with each hydrogen pathway.

At the 9% market penetration, the onsite pathway causes additional VOC concentrations of 0% to 0.007%, the pipeline pathway causes 0% to 0.014% (there is one outlier of 0.041%), and both are much smaller than the truck pathway that causes additional VOC concentrations of 0% to 0.027%. At the 20% market penetration, the onsite pathway causes additional VOC concentrations of 0% to 0.058%, the pipeline pathway causes 0% to 0.034% (again, there is an outlier of 0.102%), and the truck pathway causes additional VOC concentrations of 0% to 0.067%.

At the 9% market penetration, the onsite pathway causes additional NO_x concentrations of 0% to 0.140%, the pipeline pathway causes 0% to 0.381% (one outlier is 0.795%), and the truck pathway results in additional NO_x concentrations of 0% to 0.750%. At the 20% market penetration, the onsite pathway causes additional NO_x concentrations of 0% to 0.443%, the pipeline pathway causes 0% to 0.952% (the outlier is 1.987%), and the truck pathway results in additional NO_x concentrations of 0% to 1.861%.

In conclusion, compared to the background initial VOC and NO_x (the 3-hour averages, 6:00-9:00am), the truck pathways have the greatest impact on both VOC and NO_x pollution, the onsite pathways have the smallest impact, and the pipeline pathways are between them (even though the pipeline pathways and the onsite pathways are almost comparable in terms of the resulting additional VOC or NO_x pollution). In particular, the real-world NO_x pollution at station 9 was often relatively low in 2004 (see Figure 27), and diesel hydrogen-delivery trucks emit substantial amounts of NO_x, which explains why the truck pathway at the 20% market penetration can lead to up to a 2% increase of the current NO_x pollution levels.

Table 11. Descriptive statistics for incremental 3-hour average VOC concentrations, ppbC

Scenario	N	Range	Minimum	Maximum	Mean	Std. deviation
onsite, 9%	93	0.00398	0.00000	0.00398	0.00066	0.00074
pipeline, 9%	93	0.00817	0.00000	0.00817	0.00080	0.00164
truck, 9%	93	0.01479	0.00000	0.01479	0.00206	0.00301
onsite, 20%	93	0.03617	0.00000	0.03617	0.00282	0.00487
pipeline, 20%	93	0.02043	0.00000	0.02043	0.00200	0.00410
truck, 20%	93	0.03689	0.00000	0.03689	0.00520	0.00748

Table 12. Descriptive statistics for incremental 3-hour average NO_x concentrations, ppb

Scenario	N	Range	Minimum	Maximum	Mean	Std. deviation
onsite, 9%	93	0.02015	0.00000	0.02015	0.00299	0.00330
pipeline, 9%	93	0.06097	0.00000	0.06097	0.00516	0.01089
truck, 9%	93	0.07298	0.00000	0.07298	0.01063	0.01588
onsite, 20%	93	0.18763	0.00000	0.18763	0.01369	0.02490
pipeline, 20%	93	0.15242	0.00000	0.15242	0.01290	0.02721
truck, 20%	93	0.18194	0.00000	0.18194	0.02686	0.03932

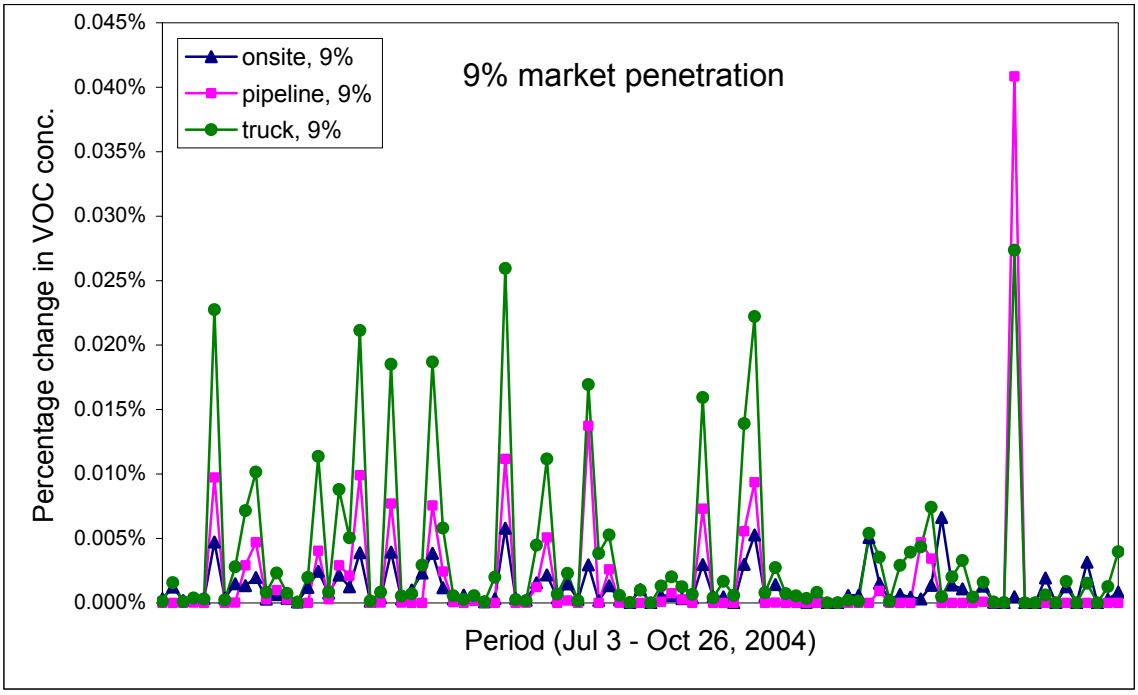

Figure 31. Comparison of percentage changes in 3-hour average VOC concentrations (9% market penetration)

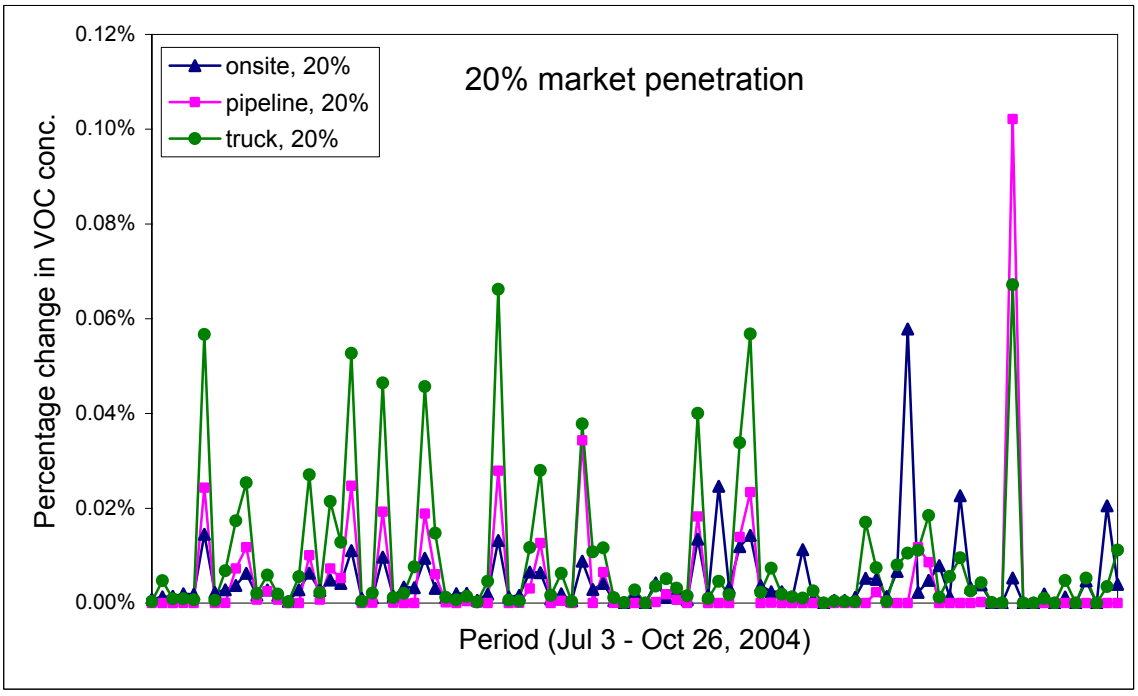

Figure 32. Comparison of percentage changes in 3-hour average VOC concentrations (20% market penetration)

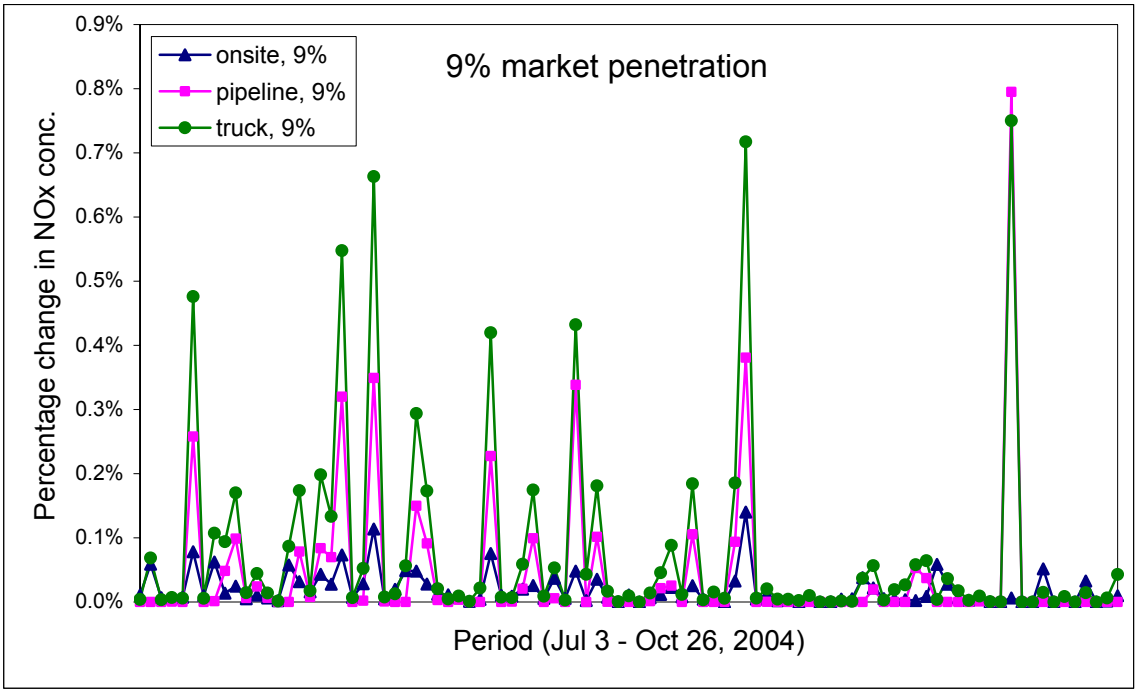

Figure 33. Comparison of percentage changes in 3-hour average NO_x concentrations (9% market penetration)

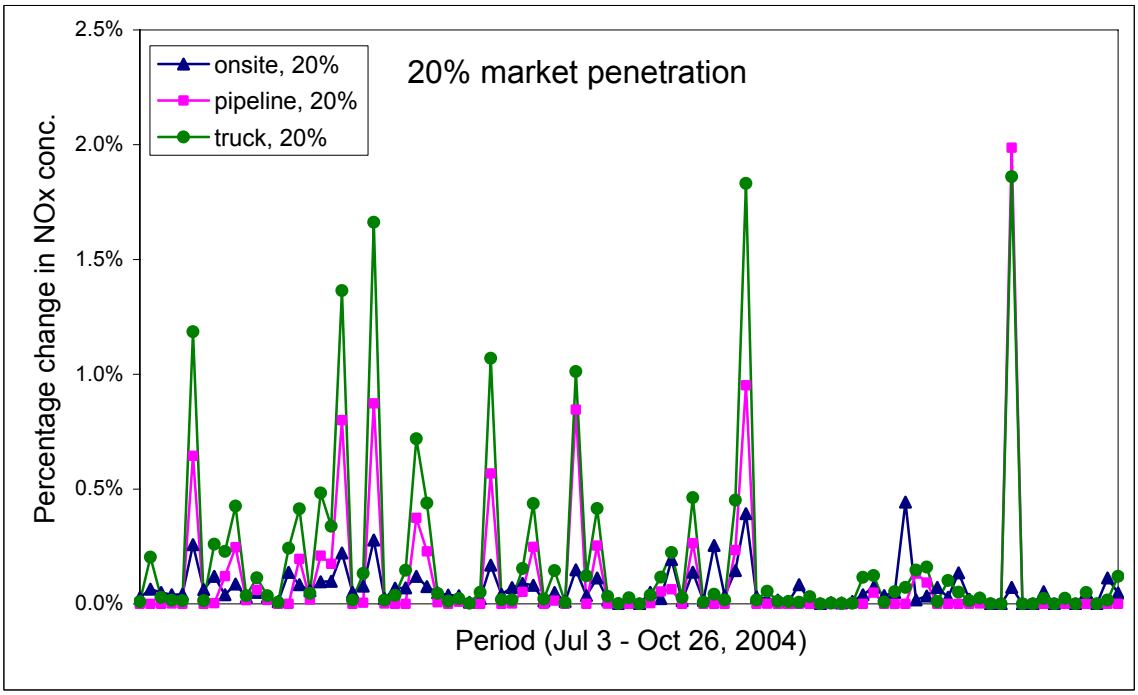

Figure 34. Comparison of percentage changes in 3-hour average NO_x concentrations (20% market penetration)

3.4.2. Changes in peak ozone concentrations, $\Delta O_3(\max)$

Ozone formation in the atmosphere is a complicated issue. Table 13 summarizes the estimated changes in 1-hour peak ozone concentrations due to lifecycle emissions of each hydrogen pathway. The range of $\Delta O_3(\max)$ results increases as the market penetrations increase from 9% (corresponding to 27 refueling stations) to 20% (corresponding to 66 refueling stations). Given the same market penetration, truck pathways correspond to the widest range of $\Delta O_3(\max)$ results, which means that truck pathways tend to result in much more variation in degradation or improvement of ozone air quality.

As shown in Table 13, all the minimum $\Delta O_3(\max)$ results are negative and all the maximum $\Delta O_3(\max)$ results are positive, which means that increases in initial VOC and NO_x concentrations do not necessarily increase the peak O_3 concentration, and may even result in a decrease. This is consistent with a standard EKMA ozone isopleth (see Figure 25). Accounting for the meteorological conditions, this phenomenon depends on the ambient ratio of initial VOC and NO_x, i.e., relative abundance of initial VOC and NO_x. NO_x is relatively abundant in the NO_x-rich zone (or so-called "VOC-limited" zone) on a typical ozone isopleth diagram, which corresponds to the situations where VOC / NO_x is generally less than 7-10 (Chang et al., 1989; NRC, 1991). However, VOC is relatively abundant in the VOC-rich zone (or so-called "NO_x-limited" zone) on an ozone isopleth diagram, which corresponds to the situations where VOC / NO_x is generally greater than 7-10 (Chang et al., 1989; NRC, 1991).

Table 13. Descriptive statistics for changes in peak ozone concentrations, $\Delta O_3(\max)$, ppb

Scenario	N	Range	Minimum	Maximum	Mean	Median	Std. deviation
onsite, 9%	93	0.00512	-0.00237	0.00275	0.00031	0.00019	0.00083
pipeline, 9%	93	0.01761	-0.00574	0.01187	0.00081	0.00000	0.00213
truck, 9%	93	0.02684	-0.00648	0.02036	0.00155	0.00018	0.00371
onsite, 20%	93	0.03351	-0.02523	0.00828	0.00016	0.00084	0.00472
pipeline, 20%	93	0.04401	-0.01437	0.02964	0.00201	0.00000	0.00530
truck, 20%	93	0.06702	-0.01641	0.05061	0.00377	0.00046	0.00926

3.4.3. Percentage changes in peak ozone concentrations, $\%\Delta O_3(\max)$

Figure 35 presents the comparison of $\%\Delta O_3(\max)$ results associated with these three hydrogen supply pathways. For the 9% market penetration scenario, the onsite pathway causes $\%\Delta O_3(\max)$ within the range of -0.007% to 0.008%, and the pipeline pathway causes $\%\Delta O_3(\max)$ within the range of -0.008% to 0.021%. The truck pathway causes $\%\Delta O_3(\max)$ within the range of -0.009% to 0.039%, which is a wider range than for the onsite and pipeline pathways.

For the 20% market penetration scenario, the onsite pathway causes $\%\Delta O_3(\max)$ within the range of -0.075% to 0.022%, and the pipeline pathway causes $\%\Delta O_3(\max)$ within the range of -0.020% to 0.052%, shown in Figure 36. Moreover, Figure 36 also demonstrates that the truck pathway causes $\%\Delta O_3(\max)$ within the range of -0.023% to 0.100%, which is also wider than the range for the onsite and pipeline pathways.

During the modeling period (July 3, 2004 through October 26, 2004), two obviously different ozone pollution trends appear. One occurs in the days preceding September 15, 2004, when the incremental VOC and NO_x from hydrogen pathways typically lead to an increase in the ozone level, and the resulting ozone pollution goes up in the following order: onsite pathway < pipeline pathway < truck pathway. That is, during the worst summer months for ozone pollution, July, August, and September, the changes in $\%\Delta O_3(\max)$ are almost all positive, therefore a worsening of ozone air quality would occur. The other trend occurs after September 15 (mostly in October), and all pathways often lead to no ozone pollution or even a decrease in ozone formation; especially onsite and truck pathways lead to greater decreases in ozone pollution.

At the 20% market penetration, the greatest increase can be up to 0.1% of the current ozone level (corresponding to the truck pathway), and the greatest decrease can be around -0.1% of background ozone pollution (corresponding to the onsite pathway). In summary, all the pathways result in very small changes in ozone air quality. However, Figures 35 and 36 both show that the truck pathway causes $\%\Delta O_3(\max)$ to fluctuate much more than do the other two hydrogen pathways (especially in July, August, and September). Therefore, just in terms of ozone pollution, all the three hydrogen pathways in some cases would result in a better ozone air quality, corresponding to a negative $\%\Delta O_3(\max)$, and in some cases will result in a worse ozone air quality, corresponding to a positive $\%\Delta O_3(\max)$, but there is little doubt that the truck pathway tends to lead to a much wider fluctuation in degradation or improvement of ozone air quality.

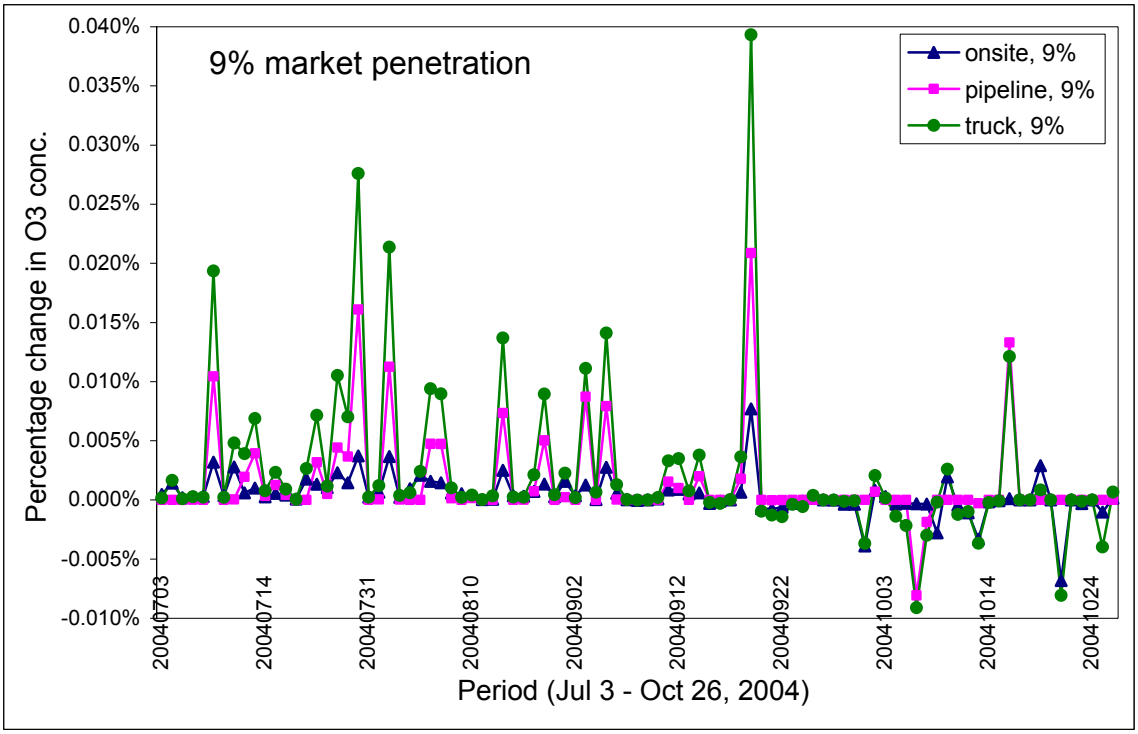

Figure 35. Comparison of percentage changes in peak ozone concentrations (9% market penetration)

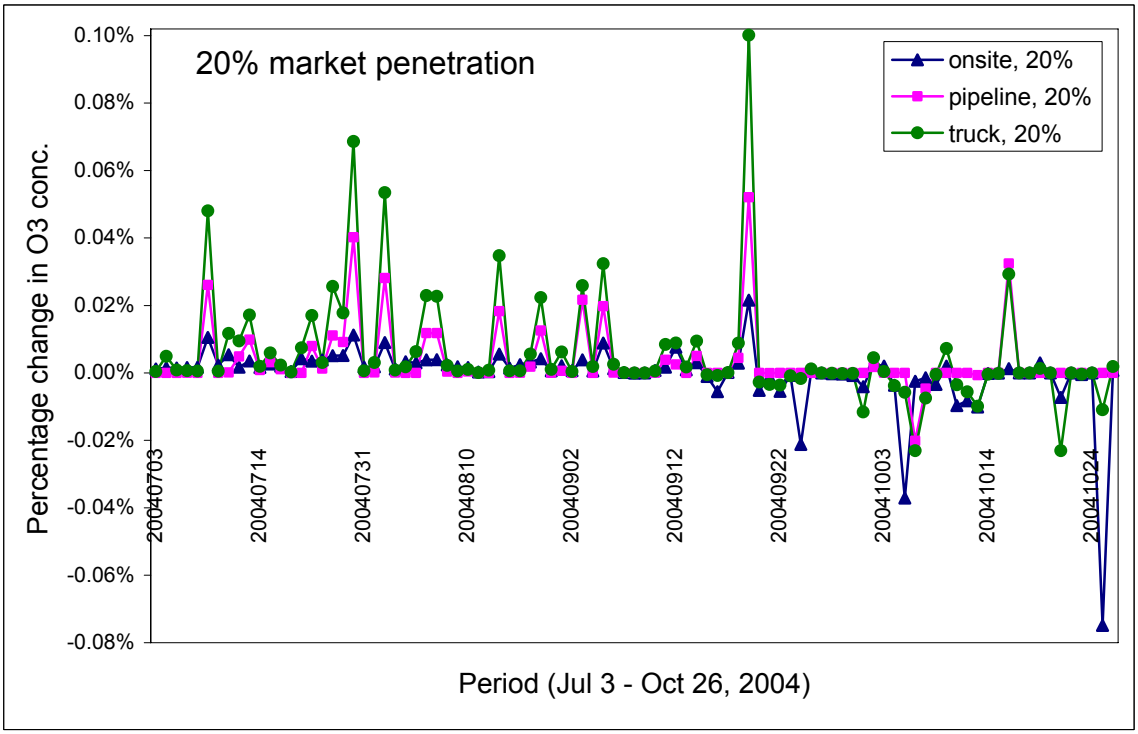

Figure 36. Comparison of percentage changes in peak ozone concentrations (20% market penetration)

3.4.4. Further discussion on ozone pollution

The ozone pollution caused by the truck pathways fluctuate most widely; percentage changes in peak ozone concentrations are approximately -0.01% to 0.04% for the 9% market penetration scenario, and approximately -0.03% to 0.1% for the 20% market penetration scenario. Note that the 20% onsite pathway can cause a decrease around -0.1% of background ozone pollution. The federal ozone standard is 80 ppb within the 8-hour averaging time (NAAQS, 2006), and the California ozone standard is 70 ppb within the 8-hour averaging time and 90 ppb within the 1-hour averaging time (CARB, 2006). Figures 27 and 30 show that the California 1-hour ozone standard is violated on some days during the modeling period, but the ambient peak ozone is often in the vicinity of the standard. Therefore, the truck pathways (and even more so the onsite and pipeline pathways) are unlikely to lead to a serious ozone problem in Sacramento.

Since the same meteorological conditions for each day are used when deriving the regression model and applying the model to the analysis of hydrogen supply scenarios, the changes in peak ozone concentrations shown in Figures 35 and 36 are due only to the variation in VOC and NO_x inputs to the model. Most of the largest positive peaks in $\%\Delta O_3(\max)$ in the summer (before September 15, 2004) correspond to the lowest VOC / NO_x ratios on those days, about 3.2-5.2 (see Figures 29, 35 and 36). Because the estimate of the ratios of the incremental VOC and the incremental NO_x due to a hydrogen supply pathway is about 0.1-0.3, the new VOC / NO_x ratios inputted to the model would

decrease. That is, the VOC / NO_x ratio goes down and the peak ozone concentration goes up accordingly. In other words, VOC, NO_x, and peak O_3 concentrations are all positive increases. Therefore, ozone formation is generally in the NO_x limited regime (limited by NO_x) in the summer in that part of Sacramento. On the contrary, most of the largest negative peaks in $\%\Delta O_3(\max)$ in the fall (after September 15, 2004) correspond to the lowest VOC / NO_x ratios, approximately 1.1-1.5 (see Figures 29, 35 and 36). Similarly, the new VOC / NO_x ratios input to the model would decrease. That is, the VOC / NO_x ratio goes down and the resulting peak ozone concentration also goes down. Put another way, both VOC and NO_x increase, but peak O_3 concentrations decrease. Therefore, ozone formation mostly is limited by VOC in the fall. In summary, the ozone production ridge line on the isopleth corresponds to a VOC / NO_x ratio between 1.5 and 5.2 in Sacramento, which is slightly lower than a typical EKMA value. That could be in part because the Sacramento region is not a closed smog chamber, station 5 is not always where peak ozone formation occurs due to the variability of wind speed and direction, and the statistical model parameters are only estimates.

3.5. Conclusions

In this chapter, we assumed two sets of hydrogen vehicle market penetrations of 9% and 20%, respectively, and considered the following three hypothetical natural gas-to-hydrogen pathways: onsite hydrogen production, central hydrogen production with gaseous hydrogen pipeline delivery, and central hydrogen production with liquid hydrogen truck delivery. Prior to estimating changes in ozone air quality due to each

hydrogen pathway, lifecycle emission inventories and optimized spatial layouts of hydrogen infrastructure were determined.

Atmospheric ozone formation is complicated. In this research, a region-specific linear regression model was developed to link the peak ozone concentrations to ambient meteorological conditions and the early morning ambient VOC and NO_x as the precursors to ozone formation. The regression model and data were limited to the Sacramento region and the time period from July 3, 2004 to October 26, 2004. The model shows that increases in precursor concentrations do not necessarily increase the peak ozone concentration, and may even cause it to decrease. The results indicate that, in Sacramento, ozone formation is generally limited by NO_x in the summer and is mostly limited by VOC in the fall. The ozone production ridge line on the isopleth corresponds to a VOC / NO_x ratio between 1.5 and 5.2 in Sacramento, which is slightly lower than a typical value observed in a closed smog chamber or empirical kinetic modeling approach (EKMA) diagrams. The ozone monitoring station used in the regression analysis is also not always an accurate indicator of peak ozone formation due to the variability of wind speed and direction.

Compared to the background initial VOC and NO_x (the 3-hour averages, 6:00-9:00am), truck pathways have the greatest impact on both VOC and NO_x pollution, the onsite pathways have the smallest impact, and the pipeline pathways are between them. Since the current light duty fleet is held constant and additional hydrogen cars are added to the fleet, the incremental VOC and NO_x pollution resulting from lifecycle emissions of all

hydrogen pathways is a positive quantity. At the 9% market penetration, the truck pathway caused additional VOC (or NO_x) up to around 0.05% (or 1%) of current pollution level in 2004. At the 20% market penetration, the truck pathway caused additional VOC (or NO_x) up to around 0.1% (or 2%) of the current pollution level.

All the hydrogen pathways would result in very small (either negative or positive) changes in ozone air quality. In some cases worse ozone air quality (mostly in July, August, and September) resulted and ozone increments increased in the following order: onsite pathway < pipeline pathway < truck pathway. In some cases better ozone air quality was predicted to result (mostly in October), and the truck and onsite pathways had a greater impact than the pipeline pathway. The truck pathway tended to lead to a much wider fluctuation in degradation or improvement of ozone air quality: percentage changes in peak ozone concentrations are approximately -0.01% to 0.04% for the 9% market penetration scenario, and approximately -0.03% to 0.1% for the 20% market penetration scenario. Note that the 20% onsite pathway occasionally resulted in a decrease of around -0.1% of background ozone pollution. So the positive and negative limits of changes in ozone pollution would be around one thousandth of current pollution levels. Compared to the current ambient pollution level, the truck pathways (and therefore the onsite and pipeline pathways) are unlikely to cause a serious ozone problem for market penetration levels of hydrogen fuel cell vehicles in the 9-20% range.

The quantified ozone concentrations can be used to estimate agricultural losses and human health damages. Based on the predicted changes in ozone pollution (and the other

criteria pollutants), dose-response functions, and demographic data in Sacramento, social costs associated with hydrogen supply pathways can be estimated. This is useful for urban planners and policy makers.

4. Investigating Contributions of Gasoline Pathways to Urban Air Pollution Using Travel Demand Model Data

4.1. Introduction

The current petroleum-fueled transportation system emits significant amounts of criteria pollutants and, as a result, accounts for a major fraction of urban air pollution in the U.S. For example, on-road motor vehicles contribute 30.6-38.5% of volatile organic compounds (VOCs), 34.3-62.2% of nitrogen oxides (NO_x), and 4.4-5.7% of particulates (PM_{10}) to annual ambient concentrations in Sacramento, California for 2005 (Wang et al., 2007). Due to close proximity to people and their daily lives, current mobile emission sources are likely to cause human health damage and, thus, result in a significant social cost (ExternE, 1998; McCubbin and Delucchi, 1996; Delucchi and McCubbin, 2004).

For simplicity, in this study we use the term “gasoline pathway” to refer to the petroleum-based fuel pathway, including both gasoline and diesel transportation fuels. Compared to hydrogen pathways which emit all criteria pollutants upstream of vehicle operation, downstream vehicle operation plays an important role for a gasoline or diesel pathway. Therefore, our focus is on estimating contributions of gasoline vehicle operations to urban air pollution, although we also consider the other gasoline pathway steps like gasoline-delivery truck emissions.

As gasoline vehicle technology is evolving, we consider various types of current and advanced gasoline vehicles. For gasoline pathways examined in the chapter, the 2005 light duty (LD) fleet is used as the reference, which corresponds to current transportation technology. To reflect the improvements in vehicle/fuel technologies and standards over time, we use the predicted 2025 light duty fleet composition as representative of advanced or evolved gasoline vehicles in the near future.

In this dissertation, the overall goal is to compare hydrogen to gasoline or diesel in terms of the resulting impacts on urban air quality (see next chapter). To do so, in this chapter, we are addressing the following two prerequisite research questions.

(1) What would be the impacts of gasoline fleet operations (and gasoline pathways) on urban air quality, using traditional 4-step travel demand data and grid-level emission inventories? and

(2) How do current and advanced gasoline vehicle pathways compare, in terms of the resulting impacts on urban air quality, from a lifecycle analysis perspective?

Ambient concentrations of pollutants are correlated with emissions, but the contribution to ambient air quality of on-road mobile sources is not necessarily equal to their contribution to regional emissions. This is true for several reasons such as the distribution of other pollution sources and regional topology, as well as meteorology. The complexity of spatial and temporal distributions of vehicle emissions/activities and the mobility of

vehicles make it very hard to quantify the proportions of ambient air pollutant concentrations attributable to on-road mobile sources. To obtain specific results, it is useful to base the analysis on a particular geographic area, and this study chooses the Sacramento metropolitan area as the setting. Using the dataset of a travel demand model, regional vehicle emissions are estimated and disaggregated into hourly, gridded inventories with a 1×1 km resolution. Transportation-related concentrations of primary pollutants are then predicted using a Gaussian dispersion model. Finally, concentration contributions of light duty vehicles to urban air pollution are estimated on a regional scale.

In summary, in this chapter we investigate contributions of various current and advanced gasoline/diesel pathways to urban air pollution using travel demand model data. We examine four gasoline pathway scenarios, and the ground-level concentrations for four primary pollutants, i.e., carbon monoxide (CO), NO_x, VOC, and PM_{10}, are estimated in order to compare air quality impacts between gasoline pathway scenarios (in this chapter) and hydrogen pathway scenarios (in Chapter 2). This study provides the basis for quantitatively comparing hydrogen and gasoline pathways in terms of their impacts on ambient air pollutant concentrations.

4.2. Overview of gasoline pathway scenarios

We consider gasoline and diesel transportation fuels produced from petroleum and used in internal combustion engine (ICE) light duty vehicles. For simplicity, we use the term “gasoline pathway” to refer to the petroleum-based fuel pathway, including both gasoline

and diesel, as about 1.5% of LD vehicles were running on diesel in Sacramento in 2005 (EMFAC2007, 2007).

From a lifecycle analysis (LCA) perspective, a petroleum-based fuel gives rise to a series of emissions throughout the entire fuel pathway (see Figure 37). Figure 37 also presents the concept of integrated gasoline or diesel pathways; the dashed line area shows the portion of the lifecycle system that is included in this analysis.

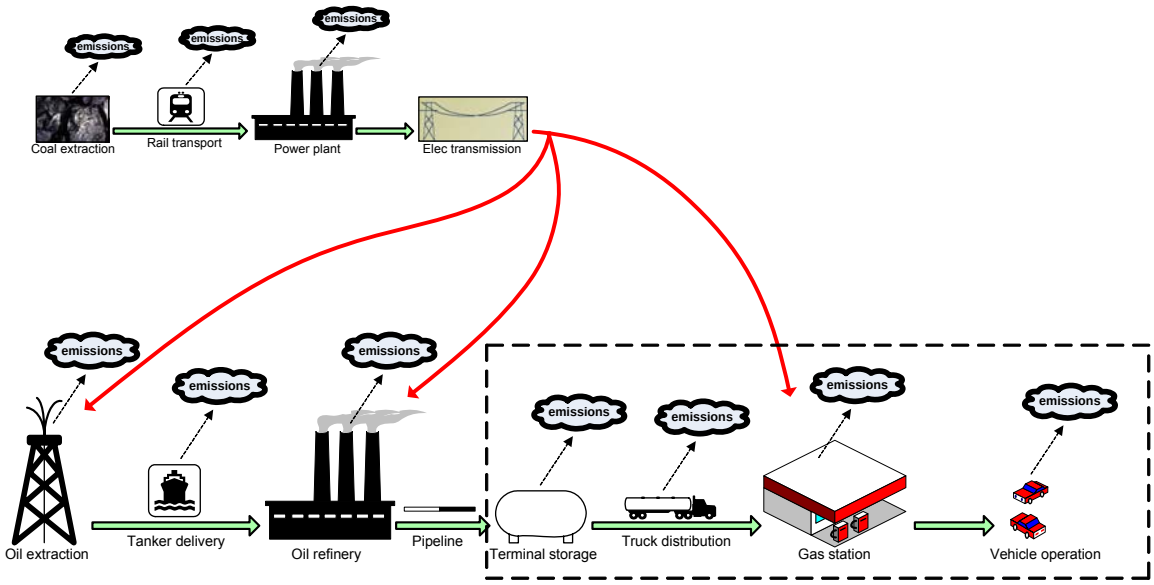

Figure 37. Integrated gasoline or diesel pathways

Although the portion of the fuel cycle upstream of the vehicle accounts for some fraction of emissions and causes air quality degradation accordingly, the vehicle operation stage, in general, plays a more important role in causing emissions and air pollution for a conventional petroleum-powered transportation system, both today and in the near future. On one hand, gasoline vehicles generate large amounts of tailpipe emissions. For

example, running the GREET model generates results indicating that, for the year 2005 in California, the passenger car operation accounts for 81% of total energy use, 63% of VOC emissions, 98% of CO emissions, 54% of NO_x emissions, and 44% of PM_{10} emissions, from well to wheels (WTW), including all sources of emissions from the entire fuel pathway (GREET1.7, 2006). On the other hand, the upstream emission sources like oil extraction sites and refineries are so far away from Sacramento that they cannot have a significant impact on urban air quality in Sacramento. In addition, vehicles have a much closer proximity to urbanized, populated areas, and they result in greater human exposure to pollutants than most major stationary sources such as power plants. This study mostly includes emissions from the vehicle operation stage of a gasoline fleet.

The air quality impacts of a gasoline pathway, however, would be underestimated if we included the vehicle operation stage only, as this ignores upstream activities, especially emissions from oil refineries, gasoline/diesel distribution, refueling stations, and sub-pathways like the electricity pathway. This could be a concern: as gasoline cars get cleaner, the upstream emissions matter more. Although refineries serving the Sacramento metropolitan area are located outside the local airshed (in Richmond or Martinez, southwest of Sacramento by about 70 miles), emissions from these oil refineries would occur and there would be a real effect on people living near the refineries. Some minor emissions, such as VOC, still occur at service stations although vapor recovery is required by law (CARB, 2008b; TCEQ, 2008; DECC, 2007). Analogous to truck delivery in the hydrogen pathways (see results in Table 9), gasoline delivery via truck or pipeline could have a non-negligible effect on air pollution. For simplicity, this study considers

the two important gasoline pathway steps: vehicle operation and gasoline delivery via diesel trucks. Inclusion of point sources like refueling stations would only make the hydrogen pathway scenarios even more favorable by comparison; thus, this approach is a conservative one that gives the "benefit of the doubt" to conventional fuels.

4.3. Gasoline fleets considered

The term “light duty vehicles” refers to passenger cars (including sedans, wagons, etc.) and light trucks of 8500 lbs or less (including pickup trucks, vans, and SUVs) (Davis and Diegel, 2007). A light duty fleet includes vehicle classes 1-4 (see Table 14), which are part of a total of 13 vehicle classes defined in the EMFAC2007 model (see Table 15) (CARB, 2007a). Although the four LD vehicle classes of interest have different emission rates and, thus, an inequality of emissions still exists among the four vehicle types, it is not as serious for a LD fleet as for the total regional fleet which includes light duty vehicles as well as heavy trucks and buses (Wang, 2007). In this sense, the fleet average emission rates for a light duty fleet could be more meaningful than that for a regional total fleet (see Section 4.4.3 for IRS4 applications).

Table 14. Light duty (LD) vehicle classes included in the gasoline pathway[1]

Vehicle class	Fuel type	Description	Weight class (lbs)
1	All[2]	Passenger Cars	All
2	All[2]	Light-Duty Trucks	0-3750
3	Gas, Diesel	Light-Duty Trucks	3751-5750
4	Gas, Diesel	Medium-Duty Trucks	5751-8500

[1] This table is selected and edited from the EMFAC2007 User’s Guide; note that EMFAC2007 includes 13 vehicle classes (CARB, 2007a).
[2] Includes gasoline, diesel, and electric.

The following four light duty (LD) fleets running on roads in the six-county Sacramento metropolitan area are investigated in the study. Vehicle miles traveled (VMT) for each of four vehicle classes and the fleet compositions including vehicle classes and populations, are extracted from EMFAC2007 using its default assumptions. The EMFAC dataset for a past or current year, say 2005, provides a reference for comparative analyses. However, the EMFAC model simulation results for a future year, say 2025, provide a reasonable projected future vehicle composition in the region. We choose these four cases spanning a range of possibilities for gasoline LD vehicles: from current technology to advanced gasoline technology. This allows us to estimate the air quality impacts resulting from cleaner, newer gasoline vehicles. Based on these four cases, we can compare air quality impacts of current and advanced gasoline pathways.

(1) The year 2025 in-use light duty vehicle fleet. This fleet is a projected in-use mixture of both old and new LD vehicles whose model years range from 1981-2025, spanning 45 years. This represents a projected composite in-use LD vehicle fleet on the road in 2025 and reflects the comprehensive historical progress in gasoline/diesel vehicle technologies and standards in California. This fleet has a population of 2,091,542;

(2) The year 2025 new light duty vehicle fleet. This fleet is composed of new LD vehicles only with the model year 2025. This represents a projected most advanced LD vehicle fleet on the road in 2025 and reflects the state of the art of progress in evolving gasoline/diesel vehicle technologies and standards in 2025 such as ultra low emission

vehicle (ULEV) and partial zero emission vehicle (PZEV) mandates in California. This fleet has a population of 127,802;

(3) The year 2005 in-use light duty vehicle fleet. This in-use fleet is composed of LD vehicles whose model years range from 1965-2005, spanning 41 years, as EMFAC2007 only includes vehicles with model years starting in 1965. This represents a composite in-use LD vehicle fleet in 2005, and we use this fleet to serve as a reference for comparison with the 2025 fleets. This fleet has a population of 1,510,255; and

(4) The year 2005 new light duty vehicle fleet. This fleet is composed of new LD vehicles only with the model year 2005. This represents the current LD vehicle fleet on the road in 2005 and reflects the state of the art of gasoline/diesel vehicle technologies and standards in 2005 in California. Similarly, this fleet also serves as a reference, since our focus is on comparing hydrogen pathways to gasoline pathways with future gasoline vehicles, say, in 2025. This fleet has a population of 95,874.

Note, however, that the emission and energy use performance of a gasoline or diesel fueled vehicle deteriorates with age and mileage, so vehicle turnover plays an important role in reducing tailpipe and evaporative emissions. Considering the U.S. average vehicle lifetime is about 14 years (Davis and Diegel, 2007), the fleet turnover of advanced gasoline or diesel vehicles will, to some extent, limit the rate reducing emissions.

4.4. Methodology

4.4.1. Overview of methodology

Figure 38 illustrates the Sacramento metropolitan area, the modeling domain of this study. This region comprises six counties, namely, Sacramento, Yolo, Sutter, Yuba, Placer, and El Dorado (SACMET2005, 2005), and it corresponds to the Sacramento Area Council of Governments (SACOG), a metropolitan planning organization (MPO).

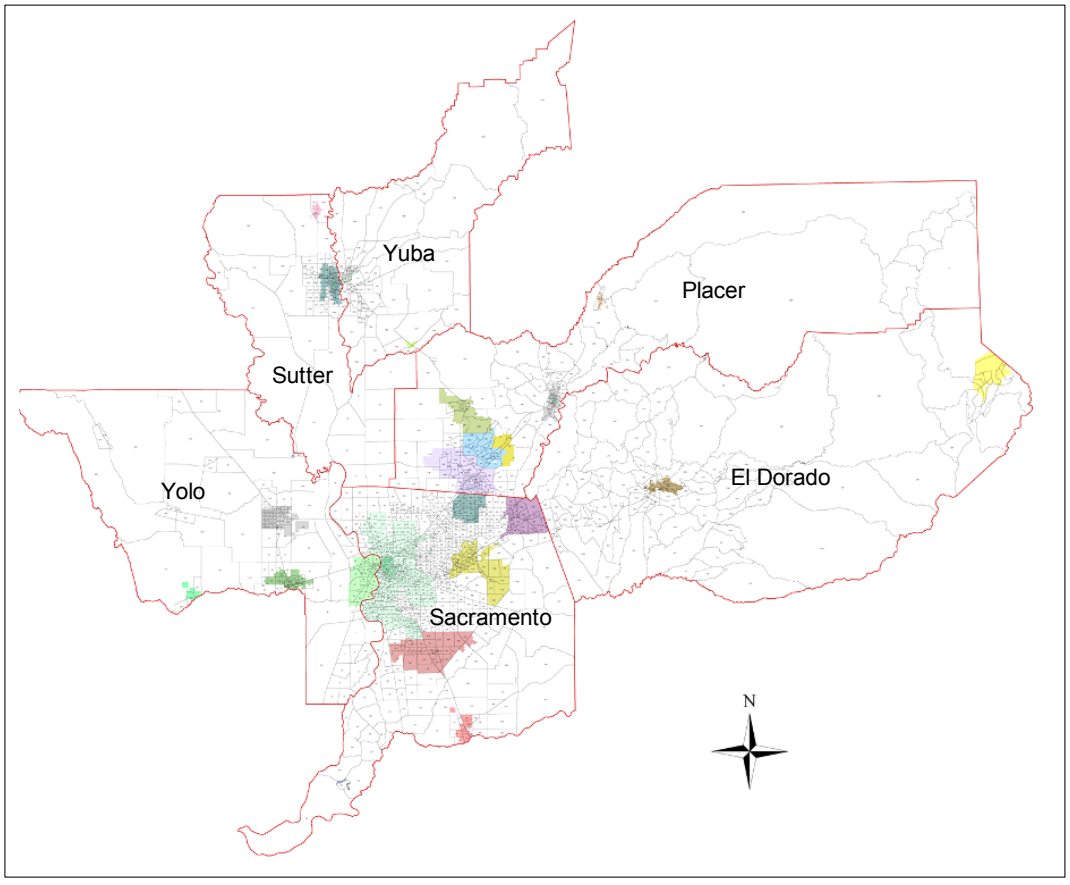

Figure 38. Sacramento metropolitan area and traffic analysis zones (TAZs)

A flow chart of the methodology and modeling sequence in the study is presented in Figure 39. We first run the California mobile emissions model, EMFAC2007, to derive emission rates for all vehicle classes in the region. For each vehicle class, annual average emission rates for the 6-county region are approximated by Sacramento County summer emissions.[2] Next, we use two intermediate models (i.e., CONVIRS4 and IRS4) to aggregate emission factors across all vehicle types. (Note that in IRS4 we could define a "fleet" of interest for useful applications; e.g., in this study we define and investigate light duty fleets, and do not analyze the regional total fleets which also include heavy duty vehicles besides light duty vehicles.) This produces a fleet averaged emission factor, which is applied throughout the region. Meanwhile, we employ data on the regional transportation networks and activities from a travel forecasting model, SACMET2005, which gives spatially detailed traffic flows for each road link, for several multi-hour time periods for a typical weekday. Thus, we combine SACMET traffic flow data with emission rate data to estimate spatially specific emissions. We run an hourly, gridded emission inventory model, DTIM4.02, to assign regional total emissions to predefined grid cells at a 1×1 km resolution, to address the spatial difference, which is important to subsequent atmospheric dispersion models.[3] Then, using the Typical Meteorological Year (TMY2) conditions and the grid-level area source emissions (i.e., DTIM model outcomes) as inputs to a Gaussian dispersion model, ISCST3, the air pollutant concentrations associated with the regional on-road motor vehicles are estimated.

[2] This is justified as emission factors in the region do not vary much over the year, and most of the traffic takes place in Sacramento County.

[3] This methodology could be useful for secondary air pollution models such as the Urban Airshed Model (UAM) for ozone formation in other studies.

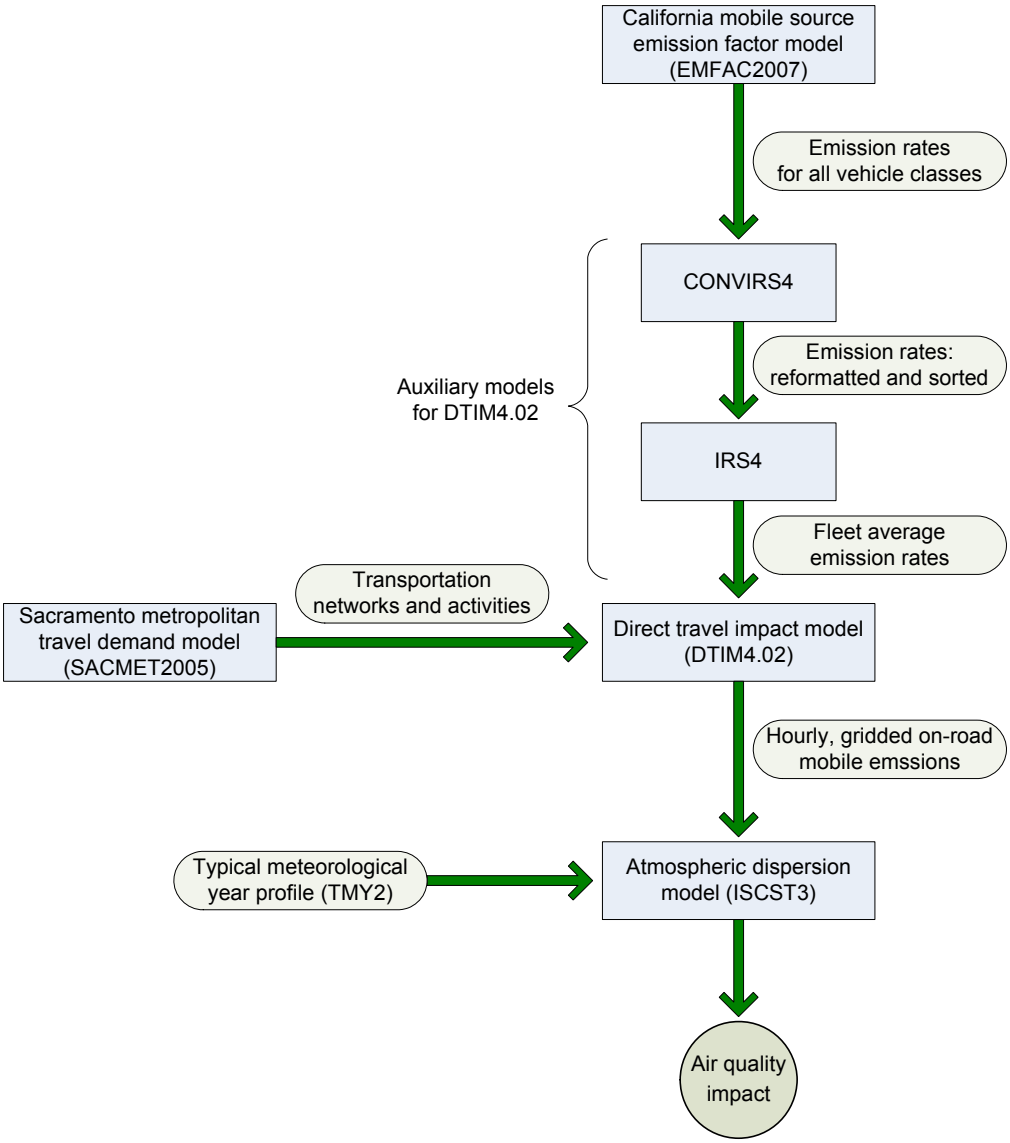

Figure 39. Methodological framework and model running sequence for gasoline fleet scenarios

4.4.2. The EMFAC model

EMFAC2007 (version 2.3, released in November 2006) is the latest version of the California mobile source emissions model. It is an officially approved regulatory model, which calculates emission inventories for motor vehicles operating on roads in California

by combining vehicle emission rates with local specific vehicle activity data (CARB, 2007a; EMFAC2007, 2007). The most common application is to generate emission factors for on-road motor vehicles, including predefined 13 vehicle classes (shown in Table 15), at a county, air basin, or state level. Also, EMFAC is capable of estimating regional total emissions by running its BURDEN module.

Table 15. Vehicle classes modeled in the EMFAC2007 model[1]

Vehicle class[3]	Fuel type	Description	Weight class (lbs)	Abbr.
1	All[2]	Passenger Cars	All	LDA
2	All[2]	Light-Duty Trucks	0-3750	LDT1
3	Gas, Diesel	Light-Duty Trucks	3751-5750	LDT2
4	Gas, Diesel	Medium-Duty Trucks	5751-8500	MDV
5	Gas, Diesel	Light-Heavy-Duty	8501-10000	LHDT1
6	Gas, Diesel	Light-Heavy-Duty	10001-14000	LHDT2
7	Gas, Diesel	Medium-Heavy-Duty	14001-33000	MHDT
8	Gas, Diesel	Heavy-Heavy-Duty	33001-60000	HHDT
9	Gas, Diesel	Other Buses	All	OB
10	Diesel	Urban Buses	All	UB
11	Gas	Motorcycles	All	MCY
12	Gas, Diesel	School Buses	All	SBUS
13	Gas, Diesel	Motor Homes	All	MH

[1] This table is selected and edited from the EMFAC2007 User's Guide (CARB, 2007a).
[2] Includes gasoline, diesel, and electric.
[3] In this study, we focus on "light duty vehicles", including the first four vehicle classes in Table 15 (with shading).

The EMFAC model has multiple applications in the study. As stated earlier, we extract LD fleet data for both 2005 and 2025 from EMFAC; e.g., the regional total vehicle population (or VMT) is derived by adding up the six county-level data. We also run

EMFAC2007 to provide emission rates for the predefined four light duty vehicle classes (see Table 14), i.e., the first four vehicle classes in Table 15.

The following types of air pollutants and their associated emission processes are considered in the research (see Table 16): CO, NO_x, total organic gases (TOGs) or VOC, and PM_{10}. Note that we only calculate direct vehicular emissions; e.g., we do not account for re-entrainment of PM_{10} and particulates from tire wear and brake wear. For TOG or VOC, we account for both vehicle tailpipe exhaust and evaporative emissions; for the other pollutants, we just consider direct tailpipe emissions. Only primary emissions that are directly released from emission sources are included and no secondary atmospheric formation, such as secondary particulate matter and ozone (O_3), is considered in this study.

Table 16. Vehicle emission processes and activities

Pollutant	Emission processes and sources
CO	Running exhaust, idle exhaust, starting exhaust
NO_x	Running exhaust, idle exhaust, starting exhaust
TOG or VOC	Running exhaust, idle exhaust, starting exhaust, diurnal, hot soak, running loss, resting loss
PM_{10}	Running exhaust, idle exhaust, starting exhaust

The EMFAC model can provide emission estimates for both summer and winter scenarios, but the seasonal variations in the daily vehicle emissions appear insignificant in the Sacramento region, based on results generated by running EMFAC for summer and winter, respectively. For simplicity, a typical daily emission inventory is developed based

on the summer 2005 or 2025 vehicle emission rates for Sacramento County; i.e., we use the summer vehicle emission rates to represent the annual (including summer and winter) average emission rates.

4.4.3. The CONVIRS and IRS models

Two auxiliary models, namely CONVIRS4 and IRS4, are used to develop an estimate of the fleet average emission rates. These two programs further process the emission rate output from EMFAC and prepare for inputs to DTIM. CONVIRS4 must be run prior to IRS4 because it provides valid input into IRS4.

The application of CONVIRS4 reformats and sorts the emission rates generated by EMFAC, and then IRS4 calculates fleet average emission rates. In this step, vehicle class weights are used to define the fleet of interest, and weighted average emission rates for the specific fleet are derived. The vehicle class weights are usually the proportions of total VMT by each vehicle class in the fleet, and they should be supplied to IRS in the order that EMFAC vehicle classes have (Caltrans, 2001), shown in Table 15. Note, however, that we are only interested in the LD fleets, so the vehicle class weights for those other than classes 1-4 are zero. This produces a new fleet averaged emission factor, which is applied throughout the region.

4.4.4. The SACMET model

We employ data on the regional transportation networks and activities from the Sacramento Metropolitan travel demand model (SACMET2005), which gives spatially detailed traffic flows for each road link, for several multi-hour time periods for a typical weekday (DKS Associates, 2002).

SACMET is a travel forecasting model specifically developed for SACOG. The SACMET model applies a traditional four-step travel demand modeling procedure, namely trip generation, trip distribution, mode choice, and trip assignment (DKS Associates, 2002). Vehicle trips and loaded networks are estimated based on the regional travel demand. The term “loaded networks” refers to the base networks with assigned forecasted traffic (e.g., vehicle volume and speed). Figure 40 depicts the transportation network links (i.e., roadways or roadway segments, including centroid connectors) for 2005 (SACMET2005, 2005).

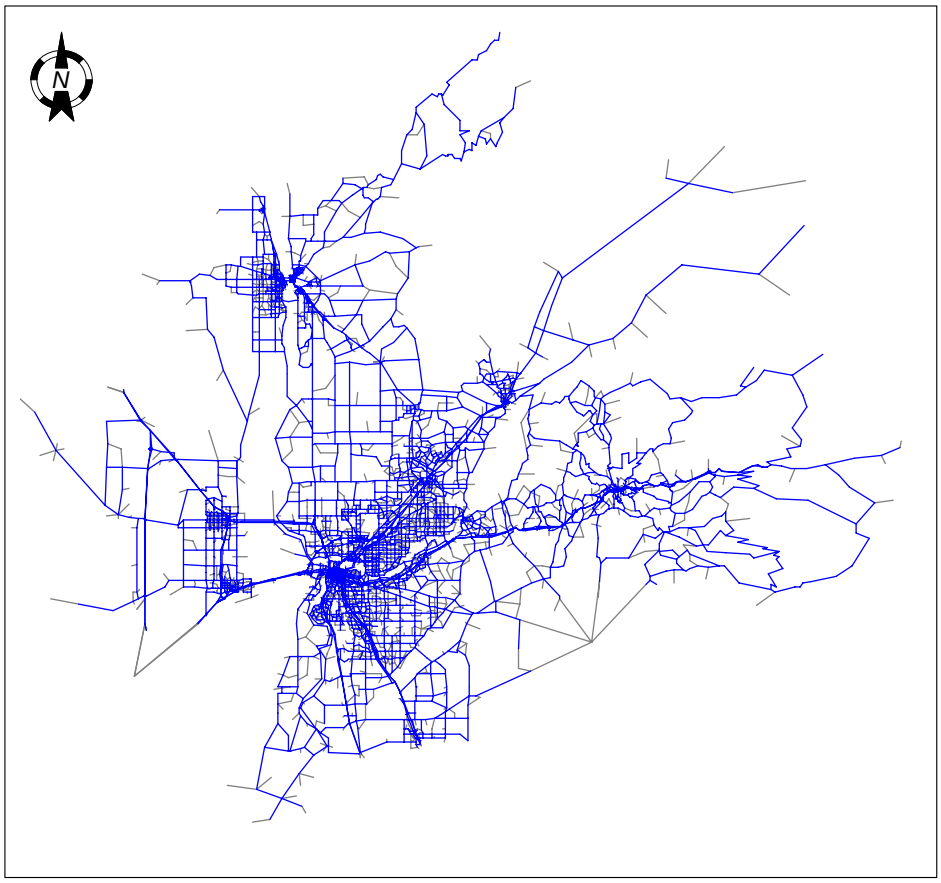

Figure 40. Transportation networks in the Sacramento metropolitan area

The modeling domain, composed of six counties, is divided into traffic analysis zones (TAZs) for the purpose of transportation planning rather than mobile emissions control (see minor zones in Figure 38). Generally, a TAZ is not the same as a census tract, and a zone centroid is usually predefined to reflect travel between TAZs in a travel demand forecasting model (Niemeier et al., 2004). In SACOG's database, TAZs are aggregated into some regional analysis districts (RADs). Table 17 presents the regional network modeling profile (SACMET2005, 2005).

Table 17. The SACMET2005 network modeling profile

Modeled parameter	Number
Counties	6
Regional analysis districts (RADs)	73
Traffic analysis zones (TAZs)	1,398
Roadway links (including centroid connectors)	20,049
Link nodes (including centroids)	8,886

SACMET2005 generates traffic data such as traffic volume, congested speed, and travel time for four time periods, based on roadway link attributes (e.g., road capacity, free-flow speed, link length, and number of lanes). The four modeled time periods are AM peak (6am-9am, 3 hours), midday (9am-3pm, 6 hours), PM peak (3pm-6pm, 3 hours), and evening (6pm-6am, 12 hours). The model results are multi-hour aggregate data for a weekday, which requires the subsequent hourly, gridded emission inventory model to disaggregate them into hourly data for each grid cell.

4.4.5. The DTIM model

The next step in the modeling procedure is to apply the Direct Travel Impact Model (DTIM4.02), maintained by the California Department of Transportation, to generate hourly, gridded emission inventories to address temporal and spatial distributions of motor vehicle emissions (Caltrans, 2001). Vehicle emission processes in DTIM are the same as those in the EMFAC scenario (see Table 16).

4.4.5.1. Temporal distribution of emissions

As stated earlier, SACMET only provides four time-period aggregate traffic data. DTIM is capable of disaggregating time-period emissions into hourly emissions by using the data file of the region-specific vehicle starts/parks/stables distributions.

4.4.5.2. Spatial distribution of emissions

The following three types of vehicle trips/activities, associated with emissions, are considered in DTIM in order to derive grid-level emissions.

(1) Links. Links refer to roadways or roadway segments, including centroid connectors. Stabilized running exhaust emissions occur regardless of an interzonal or intrazonal vehicle trip. Running emissions are estimated at the link level and assigned to the grid cell that the link belongs to;

(2) Trip-ends. Trip-ends incur starts/parks emissions; and

(3) Intrazonal. Intrazonal vehicle trips refer to the travel within a traffic analysis zone (TAZ). Intrazonal trips incur running/starts/parks emissions.

4.4.5.3. Determining the size of grid cells

Both trip-end and intrazonal emissions are assigned to the grid cell where the TAZ centroid is located. A zone centroid is not designated for emission purposes and emission activities do not necessarily occur at a TAZ centroid (Niemeier and Zheng, 2004), so it is

somewhat biased when the size of grid cells and the size of TAZs are not comparable. However, it is currently the common method for estimating gridded vehicle emissions. In the Sacramento region, 99% of TAZs are larger than the 1×1 km grid cell resolution (Niemeier and Zheng, 2004). Generally, it would be ideal that TAZs and grid cells have a comparable size. Although the TAZs in suburban and rural areas are much larger in size than those TAZs in urban areas and central business districts (CBDs), we expect that the urban TAZs contribute more to urban air pollution. In addition, most, if not all, TAZs do not have a regular geometric shape and, thus, the length of one side is not necessarily larger than 1 km although the area of this zone is possibly much larger than 1 km^2. Therefore, the 1×1 km grid cells determined in this study are appropriate in terms of spatial resolution, and emissions at the grid level could be reasonably generated.

In summary, we divide the modeling domain into 48400 (=220×220) grid cells at a 1×1 km resolution, according to the Universal Transverse Mercator (UTM) coordinate system. The regional total emissions are assigned, by running DTIM, to each grid cell by hour based on the actual transportation networks and activities derived from SACMET. These grid cells are treated as an area source of pollution.

4.4.6. The ISC model (revisited)

Following DTIM, an air quality model is run. The Industrial Source Complex Short Term (ISCST3) model, maintained by U.S. EPA, is a steady state Gaussian plume dispersion model (ISCST3, 2006; U.S. EPA, 1995). It works directly for point, area, volume, and open pit sources of pollution, and by approximation to a series of long, thin area sources

or volume sources, a line source of pollution can be simulated as well (U.S. EPA, 1995). It also can be used to assess air pollution from a variety of sources simultaneously.

Nine air quality monitoring stations located in urban Sacramento are chosen to serve as the dispersion model pollution receptors (see Figure 41). They are the same sites as for the hydrogen pathway scenarios (see Figures 7 and 8 in Chapter 2).

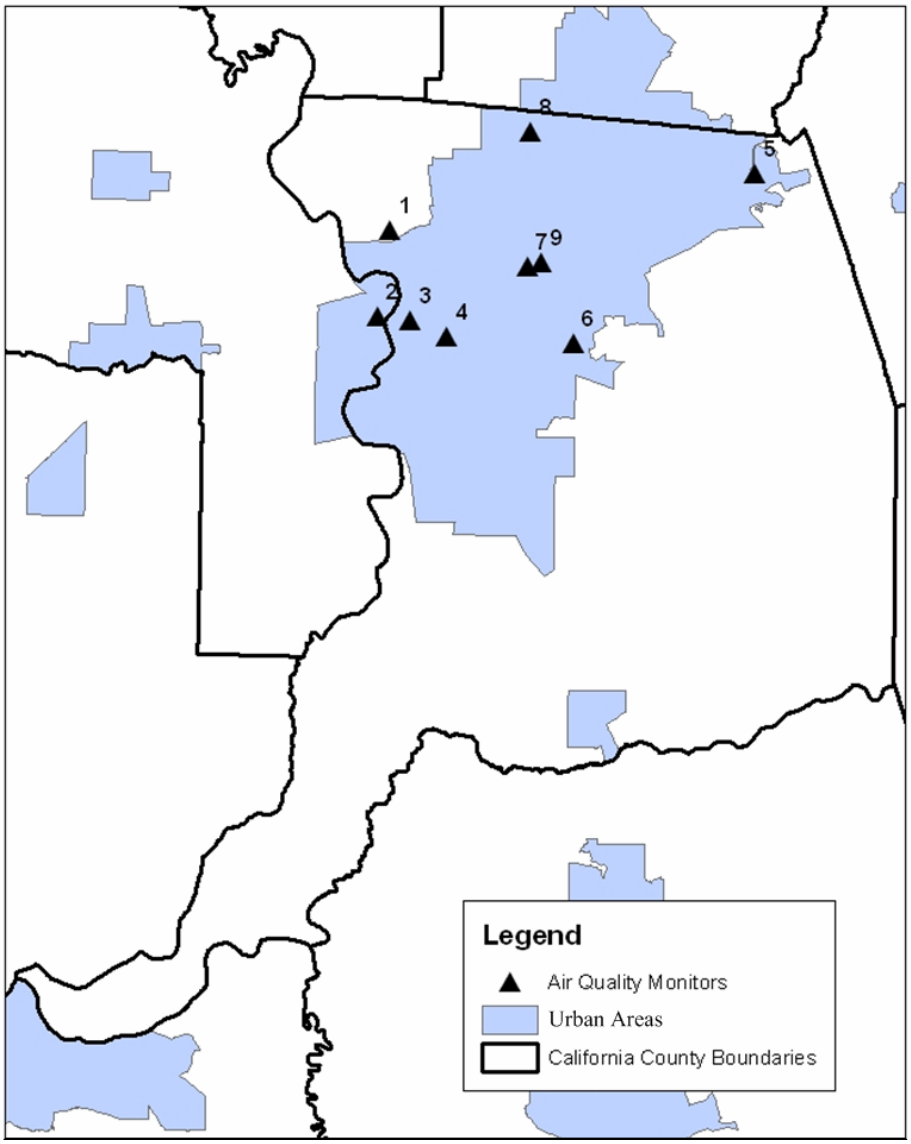

Figure 41. Nine air quality monitoring stations (or pollution receptors) in urban Sacramento

In this study, grid-based emissions from on-road light duty vehicles are treated as an area source of pollution. Specifically, the running emissions from links are assigned to the grid cells that the links belong to since we know the coordinates of link nodes and other location information from SACMET. Usually, the running emissions account for the majority of the total grid cell emissions. The trip-end (starts/parks) emissions are assigned to the TAZ-centroid grid cells, as discussed above. The intrazonal (within zones) emissions, including running, starts, and parks, are assigned to the TAZ-centroid grid cells as well.

Finally, the 24 hour-of-day dynamic (time-varying) emission rates of the grid-cell area sources are input to the ISCST3 model. Note that only physical transport applies to this study and no chemical transformation such as ozone formation or secondary PM is considered. Thus, with typical meteorological factor inputs described below, the impacts of on-road mobile sources on urban air quality can be estimated.

4.4.7. The TMY2 dataset (revisited)

The Typical Meteorological Year (TMY2) dataset, developed by the National Renewable Energy Laboratory (NREL), consists of months selected from 30 years (from 1961 through 1990) to form a hypothetical complete year, so it is the statistically most typical meteorological condition rather than the worst case conditions (TMY2, 2006). TMY2 provides the following hourly inputs to ISC: hour of day, wind direction, wind speed,

ground-level ambient temperature, atmospheric stability class, rural mixing height, and urban mixing height.

We use Sacramento County TMY2 data to represent the whole region; i.e., throughout all the six counties, the meteorological elements are assumed to be uniformly the same as those typically experienced in Sacramento County. Because TMY2 is not necessarily identical to the actual meteorological conditions in 2005 (even for Sacramento County), it may cause an error when we compare the predicted air pollutant concentrations with the measured concentrations at air quality monitoring stations. However, the predictive results associated with TMY2 conditions are more statistically meaningful, particularly for a future year, say 2025; in addition, the annual average pollution derived in this study may, at least to some extent, average out the random and non-typical effects throughout the year.

4.5. Results and discussion

4.5.1. Air pollutant concentrations caused by gasoline fleet operations

Air pollutant emissions depend on the magnitude of vehicle miles traveled (VMT). In our modeling, we use the SACMET2005 model for traffic flow data, which is appropriate for 2005 traffic conditions only. SACMET2005 gives us an estimate of the total number of vehicle miles traveled by all vehicle types including light duty vehicles plus heavy duty vehicles and buses. But in our gasoline fleet scenarios, we want to model LD vehicle traffic flows in different years: 2005 and 2025. First we define a base VMT level, VMT_0,

which is equal to the number of vehicle miles traveled by all vehicle types in 2005; then, we use a scaling method to account for the VMT variability for various gasoline fleet scenarios.

Recall that the levels of travel activity represented by the regional travel demand model SACMET2005 are used in the methodology (see the flow chart in Figure 39). This means when we use our methodology for various fleet scenarios, the regional transportation activity level (say VMT) for 2005 is always used by default; put another way, the procedure automatically keeps VMT and traffic flows unchanged for any fleet mix modeled. To make the framework broadly applicable, the scaled concentration is defined as

$$C_1 = \frac{C_0}{VMT_0} \times VMT_1 \quad , \qquad \text{(Equation 8)}$$

where

C_0 refers to the concentration output from our model running sequence (see Figure 39), shown in Table 18. It represents the predicted annual mean concentration corresponding to the travel activity level VMT_0;

C_1 refers to the concentration resulting from the entire LD fleet defined of interest, shown in Table 19;

$VMT_0 = 57{,}203{,}211$ miles/day, which refers to vehicle miles traveled (VMT) for all the vehicles, composed of vehicle classes 1-13 defined in EMFAC2007, operating in the Sacramento metropolitan area in 2005. The number is obtained from EMFAC2007 by adding up VMT for six individual counties. This level of traffic flow and activity corresponds to that represented by the travel demand model SACMET2005 and it is a very important metric, as our estimation is built upon the framework of the SACMET2005 model; and

VMT_1 refers to VMT for a light duty fleet, composed of vehicle classes 1-4 only, projected or observed to be operating in the Sacramento metropolitan area in 2025 or 2005; e.g., $VMT_1 = 66{,}498{,}532$ miles/day for the in-use LD fleet in 2025. The numbers are obtained from EMFAC2007 by adding up LD vehicle VMT for six individual counties.

Table 18. Annual mean concentrations (i.e., C_0) resulting from gasoline fleet operations, given VMT_0=57,203,211 miles/day

Pollutant	In-use 2025 LD fleet	New 2025 LD fleet	In-use 2005 LD fleet	New 2005 LD fleet
CO (μg/m^3)	25.61546	8.38921	126.83858	15.64285
NO_x (μg/m^3)	1.14844	0.26821	6.24944	0.76226
VOC[1] (μg/m^3)	2.31277	0.35974	9.31742	0.61128
TOG (μg/m^3)	2.51388	0.39102	10.12763	0.66443
PM_{10} (μg/m^3)	0.22434	0.03592	0.19333	0.03903

[1] The VOC concentrations are obtained by scaling the TOG data, as TOG is used in a series of transportation models like EMFAC2007. VOC accounts for 92% of the mass fraction of TOG on an on-road mobile source basis, derived from on-road mobile emissions data for the Sacramento metropolitan area (CARB, 2007b). For non-mobile sources, this mass fraction does not necessarily hold.

Table 19, based on Table 18, presents estimated annual mean incremental concentrations to ambient pollution resulting from the entire LD fleet scenarios, averaged over nine receptors. Although vehicle population and total VMT are projected to increase significantly from 2005 to 2025, the in-use 2025 LD fleet results in much less pollution, with the exception of PM_{10}, than the in-use 2005 LD fleet, which reflects the improvements in vehicle/fuel technologies and standards.

Table 19. Estimated annual mean concentrations (i.e., C_1) resulting from the entire LD fleet operations

	In-use 2025 LD fleet[2]	New 2025 LD fleet	In-use 2005 LD fleet	New 2005 LD fleet	Sac. 2005 ambient measurement[3]
# of gas. vehicles	2,091,542	127,802	1,510,255	95,874	
VMT (mile/day)	66,498,532	6,750,272	51,124,896	5,083,584	
CO (μg/m^3)	29.77789	0.98997	113.36093	1.39016	601
NO_x (μg/m^3)	1.33506	0.03165	5.58538	0.06774	43.2 (NO_2)
VOC[1] (μg/m^3)	2.68858	0.04245	8.32737	0.05432	36.9
TOG (μg/m^3)	2.92237	0.04614	9.05149	0.05905	N.A.
PM_{10} (μg/m^3)	0.26079	0.00424	0.17279	0.00347	20.4

[1] The VOC concentrations are obtained from the TOG data scaled by a factor of 92%.
[2] For example, the resulting concentration for the entire 2025 in-use LD fleet, shown in column 2 of Table 19, is derived using Equation 8 as follows: $C_1 = \frac{C_0}{VMT_0} \times VMT_1 = \frac{C_0}{57203211} \times 66498532 = C_0 \times 1.162$; that is, the concentration results shown in columns 2 are equal to those outputs from our model running sequence scaled by 1.162. The same approach applies to the other fleets, listed in columns 3-5 of Table 19.
[3] The Sacramento 2005 ambient measurements are calculated based on data from U.S. EPA's Air Quality System (AQS).

For intuitive comparison purposes, the Sacramento annual average ambient measurements for 2005 are listed in the last column of Table 19. The in-use 2005 LD fleet, with a population of 1,510,255 (which is part of the regional total vehicle population of 1,688,877 in 2005), contributes about 19% of CO, 13% of NO_x, 23% of

VOC, and 1% of PM_{10} to measured ambient concentrations for 2005. Note that, due to the limitations of the methodology (see Figure 39), these results tend to be underestimated because the corresponding emissions are underestimated compared to EMFAC2007-based emission calculations (Wang et al., 2007). In contrast, on-road motor vehicles (including light duty vehicles as well as heavy duty vehicles and buses) contribute 30.6-38.5% of VOC, 34.3-62.2% of NO_x, and 4.4-5.7% of PM_{10} to annual ambient concentrations in Sacramento, California for 2005 (Wang et al., 2007).

It is interesting to note that we may also consider our 2025 gasoline fleet scenarios as representing a hypothetical year 2005 with a year 2025 fleet mix (either new or in-use), i.e., what 2005 would have looked like if the future fleet mix had been realized early.

To allow better comparison with hydrogen pathways, the concentrations in Table 19 are now further scaled to make the number of LD vehicles for each gasoline fleet scenario equal to the number of FCVs served by a hydrogen pathway (see Table 20). Table 20 presents estimated annual mean incremental concentrations to ambient pollution due to gasoline fleet operations, averaged over nine receptors. Given the same number of vehicles or on a per vehicle basis, the in-use 2025 LD fleet results in much less pollution, with the exception of PM_{10}, than the in-use 2005 LD fleet, which certainly reflects the improvements in vehicle/fuel technologies driven by evolving emissions standards in California. Similarly, for a scenario year, the new LD vehicle fleet would lead to much lower concentrations than the in-use fleet, assuming the same size population. On one hand, that reflects progress in technologies and standards in California. On the other hand,

the energy consumption and emission performance of a conventional vehicle deteriorates as vehicle age or mileage increases; thus, it appears that fleet turnover is conducive to reducing vehicle emissions and improving urban air quality. In fact, light duty vehicles last a long time and the U.S. average vehicle lifetime is about 14 years (Davis and Diegel, 2007). The relatively slow turnover rate somewhat limits the potential effect of changes in vehicle technology on reductions in energy consumption and tailpipe emissions (Farrell and Sperling, 2007).

On a per vehicle basis, a new vehicle of the model year 2025 is cleaner than a new vehicle of the model year 2005 in terms of the resulting concentrations of all four pollutants: CO, NO_x, VOC, and PM_{10}. However, PM_{10} concentrations for the model year 2025 vehicle are just slightly lower than for the model year 2005 vehicle, which might be because reducing particulate matter from a LD vehicle is either making slow progress or not a research and development (R&D) focus due to relatively small amounts of emissions by mass, as vehicular particulates are mainly from heavy duty vehicles like heavy trucks or buses which are not considered in the study. It is interesting to note that the in-use 2025 LD fleet, on a per vehicle basis, tends to cause greater PM_{10} pollution level than the in-use 2005 LD fleet, as is discussed in more detail later in Section 4.5.3.

Table 20. Estimated annual mean concentrations due to gasoline fleet operations

Pollutant	# of gasoline vehicles[1]	In-use 2025 LD fleet ($\mu g/m^3$)	New 2025 LD fleet ($\mu g/m^3$)	In-use 2005 LD fleet ($\mu g/m^3$)	New 2005 LD fleet ($\mu g/m^3$)	Sac. 2005 ambient measurement ($\mu g/m^3$)[2]
CO	111,400	1.58603	0.86292	8.36177	1.61529	601
	278,600	3.96651	2.15807	20.91193	4.03967	
NO_x	111,400	0.07111	0.02759	0.41199	0.07871	43.2 (NO_2)
	278,600	0.17783	0.06900	1.03035	0.19685	
VOC	111,400	0.14320	0.03700	0.61425	0.06312	36.9
	278,600	0.35813	0.09254	1.53617	0.15786	
PM_{10}	111,400	0.01389	0.00369	0.01275	0.00403	20.4
	278,600	0.03474	0.00924	0.03188	0.01008	

[1] To compare with the hydrogen scenarios examined in previous chapters, we consider two levels of gasoline vehicle populations: 111,400 gasoline vehicles (compared to the hydrogen scenario with 111,400 FCVs which equal 10% of the total LD fleet in urbanized Sacramento in the year 2000) and 278,600 gasoline vehicles (compared to the hydrogen scenario with 278,600 FCVs which equal 25% of the urbanized Sacramento gasoline fleet in 2000). For more details on hydrogen vehicle population scenarios, see Section 2.2.2.

[2] For comparison purposes, the Sacramento annual average ambient measurements for 2005 are listed in the last column. The Sacramento 2005 ambient measurements are calculated based on data from U.S. EPA's Air Quality System (AQS).

4.5.2. The treatment of gasoline-delivery trucks

Major oil refineries serving the Sacramento metropolitan area are located outside the local airshed and, instead, in Richmond or Martinez. Gasoline comes to Sacramento via pipeline, stored in terminals near urban Sacramento, and is then trucked from there to refueling stations. If we assume the terminals are about as far away from the stations on average as the central hydrogen plant, we could simply scale the liquid hydrogen (LH2) truck-based concentrations to estimate the air quality impact of diesel trucks delivering gasoline. The treatment of gasoline-delivery trucks would have a minor effect on ambient

air quality, as the gasoline fleet operation stage usually accounts for the majority of emissions and concentrations of air pollutants.

Recall the truck emissions estimated for the LH2 truck pathway in Chapter 2. Those are for LH2 delivery trucks going to each of 27 (or 66) stations. To do this scaling, we need to know how many deliveries would be made by gasoline-delivery trucks versus LH2 trucks to fuel the same number of vehicles at these stations.

For the heavy heavy-duty trucks (HHDTs) delivering gasoline, the cargo payload = 25 tons (1 ton = 907 kg) and the carried gasoline density = 2.8 kg/gal, extracted from the GREET model (GREET1.7, 2006), so the delivered gasoline is about 8000 gal per truck. We employ the in-use HHDT delivery trucks for the scenario years 2005 and 2025, respectively, and their per-vehicle emission factors are derived from the EMFAC2007 model, at the Sacramento County aggregate level, shown in Table 21 (EMFAC2007, 2007).

Table 21. Sacramento County aggregate emission factors for in-use HHDT delivery trucks

	2005		2025	
	ton/day	g/mile	ton/day	g/mile
CO	14.95	18.374	2.18	2.423
NO_x	16.91	20.782	3.88	4.313
TOG	1.94	2.384	0.43	0.478
VOC[1]	N.A.	2.194	N.A.	0.440
PM_{10}	0.65	0.799	0.1	0.111
VMT (mile/day)	738,000		816,000	

[1] The VOC emissions are obtained from the TOG data scaled by a factor of 92%.
[2] The aggregate per-vehicle per-mile emission factors are derived as follows. For example, in 2005, the in-use delivery truck CO emission factor $= \frac{14.95\ ton/day}{738000\ mile/day} \times \frac{907 \times 1000\ g}{ton} = 18.374\ g/mile$.

To estimate the air quality impact of emissions from gasoline-delivery trucks, the LH2 truck-based concentrations are scaled by using both truck emission factors (for 2025 and 2005, respectively) and needed delivery trips of gasoline vs. LH2. Table 22 shows estimated annual mean concentrations due to gasoline-delivery truck emissions for each fleet scenario. For the treatment of gasoline-delivery trucks, we make a simplifying assumption that, on average, a gasoline vehicle (either new or in-use) served by the delivery trucks has the same annual VMT level as a hydrogen FCV; this assumption does not affect the results much, as the air quality influence of truck delivery is relatively small.

Table 22. Estimated annual mean incremental concentrations due to gasoline-delivery truck emissions

Pollutant	# of gasoline vehicles served	In-use 2025 LD fleet ($\mu g/m^3$)	New 2025 LD fleet ($\mu g/m^3$)	In-use 2005 LD fleet ($\mu g/m^3$)	New 2005 LD fleet ($\mu g/m^3$)
CO	111,400	0.00155	0.00152	0.01216	0.01209
	278,600	0.00548	0.00536	0.04293	0.04269
NO_x	111,400	0.00276	0.00270	0.01376	0.01368
	278,600	0.00975	0.00954	0.04858	0.04831
VOC	111,400	0.00028	0.00028	0.00145	0.00145
	278,600	0.00099	0.00097	0.00512	0.00510
PM_{10}	111,400	0.00007	0.00007	0.00053	0.00053
	278,600	0.00025	0.00025	0.00187	0.00185
Average fuel economy (MPG)[1]		18.3	18.7	17.7	17.8
Delivery trips of gasoline vs. LH2[2]		1.23	1.20	1.27	1.26

[1] The fuel economy is for the LD fleet, and averaged over four vehicle classes (see Table 14), extracted from EMFAC2007 (EMFAC2007, 2007).

[2] We now explain how to calculate the needed delivery trips of gasoline vs. LH2, to fuel the same number of vehicles. For example, the fuel economy of the FCV is 3.28 (= 60/18.3) times that of the gasoline vehicle in the 2025 in-use fleet on average, so we need 0.305 (= 1/3.28) times the amount of energy delivered for the hydrogen case. The LH2 truck carries 3000 kg of H_2, and the gasoline truck 8000 gallons of gasoline. Since 1 kg of H_2 = 1 gallon of gasoline on a lower heating value (LHV) energy basis, the gasoline truck carries 8000/3000 times as much energy as the LH2 truck. But we only need 0.305 times the energy delivered with hydrogen compared to gasoline. So the gasoline-delivery trucks make 1.23 ($= \frac{3000\ kgH_2}{8000\ gal} \times \frac{60\ mpkgH_2}{18.3\ mpg} = \frac{3000}{8000}/0.305$) times as many deliveries as the LH2 trucks, assuming that a hydrogen FCV and a gasoline vehicle have the same annual VMT level.

4.5.3. Further discussion on aggregate vehicle emission trends

For the in-use fleet scenarios, the 2025 fleets would make dramatic progress in environmental impacts, with the exception of PM_{10}, over the 2005 in-use fleets (see Tables 18 and 20). Actually, the PM_{10} pollution resulting from in-use 2025 fleets would be slightly worse than that caused by in-use 2005 fleets.

Based on the EMFAC dataset, the level of CO, TOG, and NO_x emissions from a LD vehicle would steadily decrease from 2005 to 2025, on a per–mile-traveled basis. This is consistent with common sense and explains the improvements over time in air quality for the in-use fleet scenarios. Figure 42 shows the trends in in-use LD fleet averaged per-vehicle emissions per mile traveled for aggregate CO, TOG, or NO_x emissions (including emissions from vehicle running, idle, starting, etc., shown in Table 16).

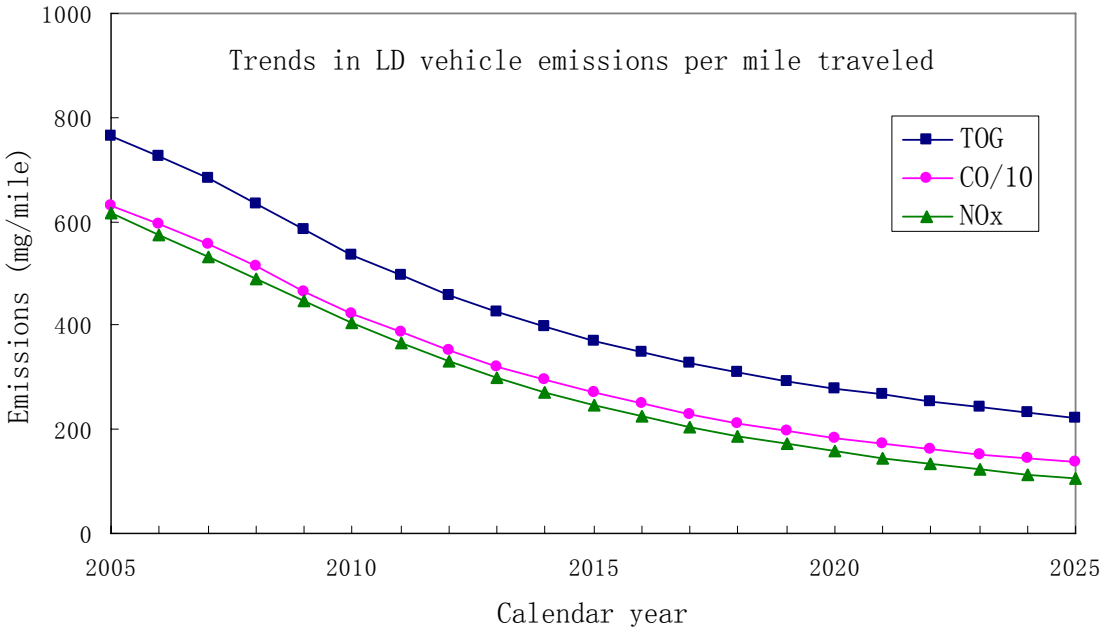

Figure 42. Trends in in-use LD fleet averaged per-vehicle emissions per mile traveled (aggregate emissions of CO, TOG, or NO_x)

Figure 43 presents the trends in aggregate PM_{10} emissions from an in-use LD fleet average vehicle on a per-mile-traveled basis (including emissions from vehicle running, idle, and starting, shown in Table 16), using data extracted from EMFAC. Different from the other three emissions, in-use vehicular PM_{10} emissions would gradually, though not smoothly, increase from 2005 to 2025, on a per-mile-traveled basis.

We know from Tables 18 and 20 that there is little improvement in new vehicle PM_{10} emissions and ambient concentrations for 2025 over 2005. One possible reason to explain the trends of PM_{10} increase (see Figure 43) for an in-use average LD vehicle is that the vehicle fleet compositions change over time and the average LD vehicle is getting bigger in size; e.g., trucks and SUVs are increasingly popular in the U.S. Usually, light duty vehicles such as passenger cars are not an important emitter of particulates; therefore, it is also possible that LD vehicle PM_{10} emissions cannot be calculated accurately by mobile emission inventory models due to the small magnitude of emissions, e.g., in the range 14.7-17.6 mg/mile spanning 2005-2025. This is not inconsistent with the results in Tables 18 and 20 that PM_{10} concentrations caused by in-use LD fleet scenarios are very close for 2025 and 2005.

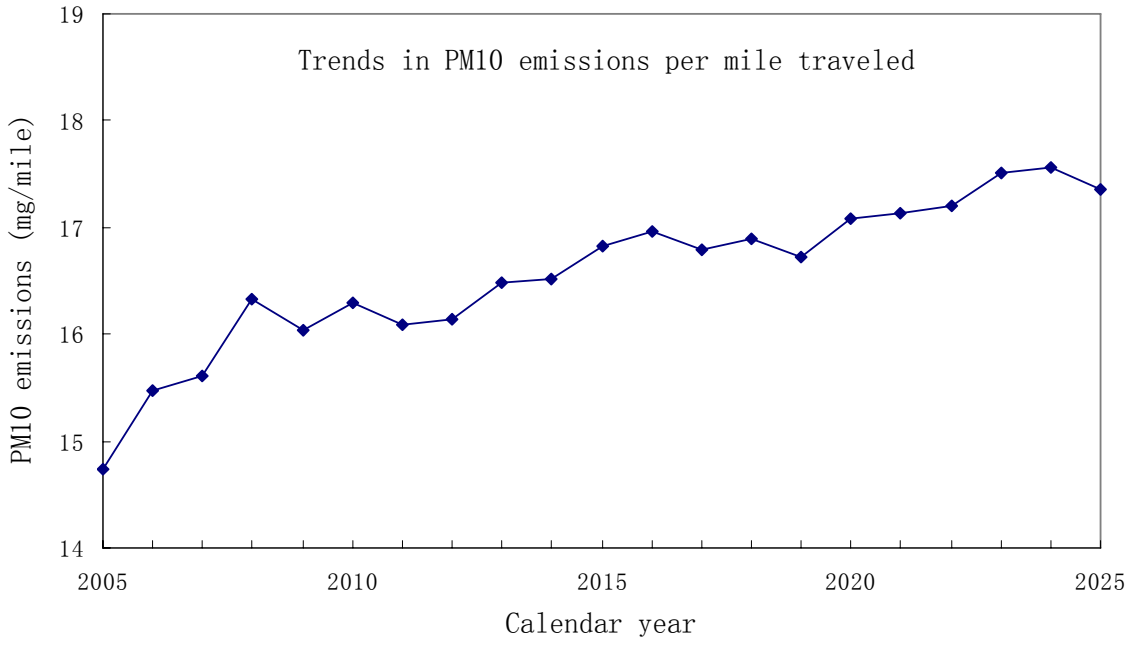

Figure 43. Trends in in-use LD fleet averaged per-vehicle emissions per mile traveled (aggregate emissions of PM_{10})

4.6. Conclusions

In this study, we investigated contributions of gasoline pathways to urban air pollution using travel demand model data. The two most important components of a gasoline pathway were included: gasoline fleet operations and gasoline delivery via diesel trucks.

Four important mobile-source pollutants, namely CO, NO_x, VOC, and PM_{10}, were considered in the study. Only physical transport of primary pollutants was considered, and no chemical transformation such as ozone formation or secondary PM was examined. In this sense, the pollution contributions of transportation sources are more important than investigated here, especially with respect to urban photochemical smog formation.

We developed gasoline/diesel pathway scenarios for the calendar years 2005 and 2025, respectively. The 2005 light duty (LD) fleet is used as the baseline, which corresponds to current transportation technology. To reflect the improvements in vehicle/fuel technologies and standards over time, we use the 2025 light duty fleet as representative of advanced or evolved gasoline vehicles in the near future.

We sequentially applied a series of models to identify the contributions of on-road light duty vehicles to urban air pollution. The modeling procedure started from the California approved regulatory emissions model (EMFAC) and Sacramento Metropolitan travel demand model (SACMET) to the gridded emission inventory model (DTIM) and air quality dispersion model (ISCST3). The modeling domain was divided into 220×220 grid

cells at a 1×1 km resolution, and the regional total emissions were disaggregated and allocated to each grid cell by hour. Then, air pollutant concentrations associated with the regional on-road light duty vehicles were estimated by using the dispersion model.

The in-use light duty vehicles in 2005 contribute about 19% of CO, 13% of NO_x, 23% of VOC, and 1% of PM_{10} to measured ambient concentrations. Note that, due to the limitations of the methodology, these results tend to be underestimated because the corresponding emissions are underestimated compared to EMFAC2007-based emission calculations (Wang et al., 2007). In contrast, on-road motor vehicles (including light duty vehicles as well as heavy duty vehicles and buses) contribute 30.6-38.5% of VOC, 34.3-62.2% of NO_x, and 4.4-5.7% of PM_{10} to annual ambient concentrations in Sacramento, California for 2005 (Wang et al., 2007).

There is scope to greatly reduce the air quality impact of gasoline vehicles, with newer, cleaner gasoline technologies. Given the same number of vehicles or on a per vehicle basis, the in-use 2025 conventional LD fleet scenario results in much less pollution, with the exception of PM_{10}, than the in-use 2005 conventional LD fleet scenario, which certainly reflects the expected improvements in vehicle/fuel technologies driven by evolving emissions standards in California. Similarly, for a scenario year, the new LD vehicle fleet (i.e., containing only vehicles of the latest model year) would lead to much lower concentrations than the in-use fleet, assuming the same population size. On a per vehicle basis, a new vehicle of the model year 2025 is cleaner than a new vehicle of the model year 2005 in terms of the resulting ambient concentrations of all four pollutants.

These results indicate that the fleet turnover plays an important role in reducing vehicular emissions and improving air quality.

However, PM_{10} concentrations from the model year 2025 vehicle are just slightly lower than those from the model year 2005 vehicle, which might be because reducing particulate matter from a LD vehicle is either making slow progress or not a focus of research and development (R&D) due to the relatively small magnitude of emissions by mass, as vehicular particulates are mainly from heavy duty vehicles like heavy trucks or buses or roadway re-entrainment which are not considered in the study.

In future work, we suggest developing an efficient and consistent approach to improve the quality of both regional and gridded emissions estimation. To be more precise, we also suggest collecting more data on gasoline pathway components other than those considered here.

5. Comparing Air Quality Impacts of Hydrogen and Gasoline Supply Pathways

5.1. Introduction

In this chapter we compare the lifecycle air quality impacts of hydrogen and gasoline supply pathways mostly based on results generated in Chapters 2 (for hydrogen pathways) and 4 (for gasoline pathways). Only primary pollutants that are directly emitted from emission sources are included, and no secondary atmospheric formation like secondary particulates or ozone (O_3) is considered in this comparison. In this chapter, we are addressing the following research question.

(1) How does hydrogen compare to conventional or advanced gasoline (or diesel) in terms of the resulting impacts on urban air pollutant concentrations, from a lifecycle analysis perspective?

Compared to hydrogen pathways which emit all criteria pollutants upstream of vehicle operation, the downstream vehicle activities play an important role for a gasoline pathway. Therefore, our focus is on estimating contributions of gasoline fleet operations to urban air pollution for a gasoline pathway, although we also consider some other pathway steps like gasoline-delivery truck emissions.

We examine changes in ambient air quality due to various hydrogen pathways compared to current and advanced gasoline/diesel pathways. We explore three hydrogen supply pathways and four gasoline pathways in this study, and compare the same size fleets of each type (either hydrogen fuel cell vehicles or gasoline vehicles). (Refer forward to Section 5.3 for the four examined gasoline scenarios for the fleet mix for gasoline vehicles.) To compare air quality impacts among the seven scenarios, we estimate the ground-level concentrations for four pollutants, i.e., carbon monoxide (CO), nitrogen oxides (NO_x, referring to both NO and NO_2), volatile organic compounds (VOCs, also called non-methane organic carbon, or NMOC), and particulate matter (PM_{10}). This chapter quantitatively compares hydrogen and gasoline pathways in terms of ambient air pollutant concentrations.

5.2. Summary of hydrogen supply pathway scenarios

This section is described in more detail in Chapter 2. For the purposes of convenient comparison, we here provide a brief summary of key information on hydrogen supply pathway scenarios.

From a lifecycle analysis (LCA) perspective, we estimate regional air quality impacts for the following three hypothetical hydrogen production and delivery pathways, based on steam methane reforming (SMR) of natural gas, which is currently the most common approach for producing hydrogen.

(1) The onsite pathway. This refers to onsite hydrogen production at the refueling station;

(2) The pipeline pathway. This refers to centralized hydrogen production with gaseous hydrogen pipeline delivery; and

(3) The truck pathway. This refers to centralized hydrogen production with liquid hydrogen truck delivery.

To link hydrogen pathways to ambient air quality in urban Sacramento, California, we develop a methodological framework for the hydrogen pathway scenarios, as shown in Figure 44.

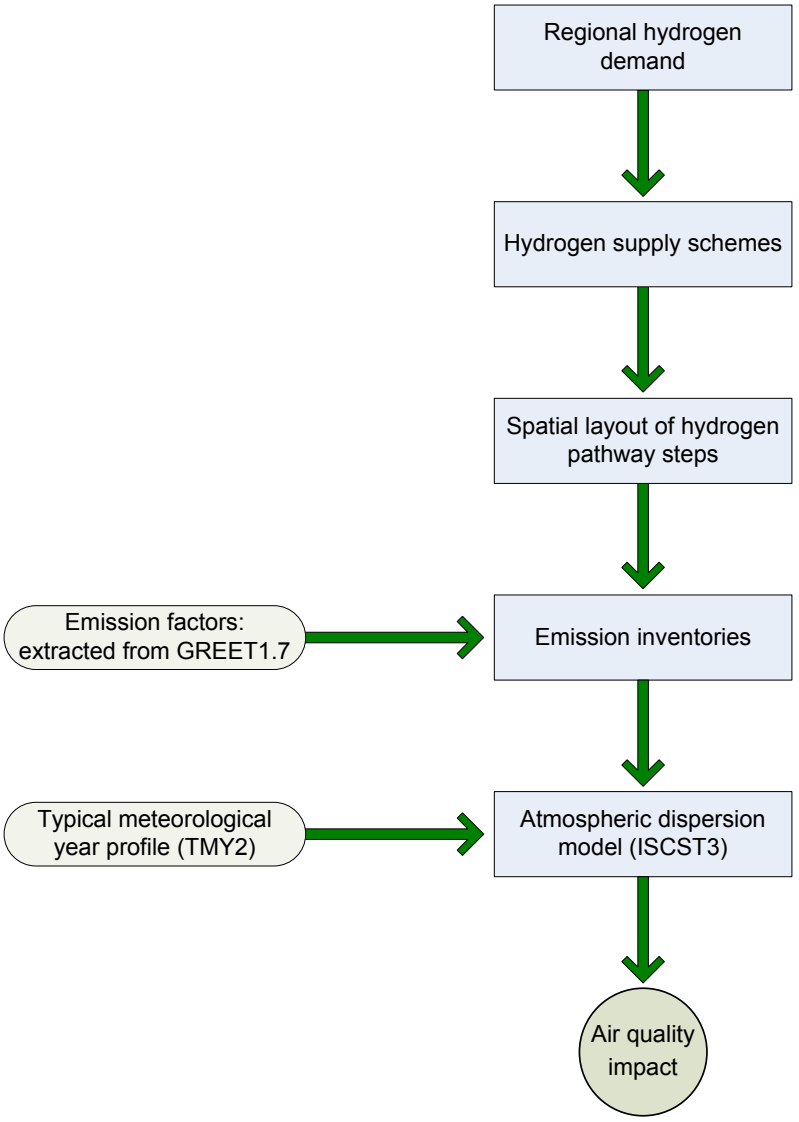

Figure 44. Methodological framework for hydrogen pathway scenarios (same as Figure 2)

There are two fleet fraction scenarios, as shown in Table 23. We use the urbanized Sacramento conventional light duty (LD) fleet in 2000 as the baseline. Holding the year 2000 gasoline fleet constant, we consider two scenarios where a number of hydrogen fuel cell vehicles (FCVs) are added to this fleet (i.e., in this study, we are not replacing gasoline vehicles with hydrogen FCVs). Thus, the total vehicle population is the sum of the year 2000 gasoline light duty fleet and added hydrogen vehicles. In scenario 1, we

add a number of hydrogen FCVs equal to 10% of the total conventional LD fleet in urbanized Sacramento in the year 2000. In scenario 2, we add a number of FCVs equal to 25% of the urbanized Sacramento LD fleet in the year 2000.

Table 23. Hydrogen pathway scenarios and hydrogen demand/supply assumptions

	Scenario 1	Scenario 2
LD gasoline vehicle population (2000)[1]	1.114 million	
Number of added hydrogen FCVs	111,400	278,600
Hydrogen FCV market penetration	9%	20%
Hydrogen fuel demand	78,000 kg/day	195,000 kg/day
Number of hydrogen stations	27	66
Fuel economy of the hydrogen FCV	60 miles/kg hydrogen	
Vehicle miles traveled (VMT)	15,000 miles/year/vehicle	
Hydrogen consumption	0.7 kg/day/vehicle	
Hydrogen station size	3,000 kg/day	
Liquid truck capacity	3,000 kg liquid hydrogen	

[1] The 2000 LD gasoline vehicle population in urbanized Sacramento is calculated as follows: Sacramento city population × LD vehicle ownership = 1.393 million × 0.8 vehicles/person = 1.114 million.

We consider only physical transport of conserved pollutants in this chapter. Thus, the background ambient pollution levels do not influence the results for the incremental ambient concentrations due to hydrogen vehicles. The two market penetration scenarios can be presented in the following way. Scenario 1 explores the resulting concentrations of pollutants from a hydrogen system with 111,400 fuel cell vehicles, a number equal to 10% of the total LD gasoline fleet in urbanized Sacramento in the year 2000. Similarly, scenario 2 explores the resulting concentrations of pollutants from 278,600 fuel cell vehicles, a number equal to 25% of the total gasoline fleet in urbanized Sacramento in

2000. Note that year 2000 was a somewhat arbitrary choice (based on available information at the time of analysis conducted) to provide a real-world basis for the number of hydrogen vehicles to analyze.

The lifecycle emissions associated with each hydrogen pathway are used to determine the impact on urban air quality within the Sacramento region. Figure 45 presents the lifecycle of one of the three integrated natural gas-to-hydrogen pathways considered, and the portion of the lifecycle system included in this analysis is enclosed by the dashed line.

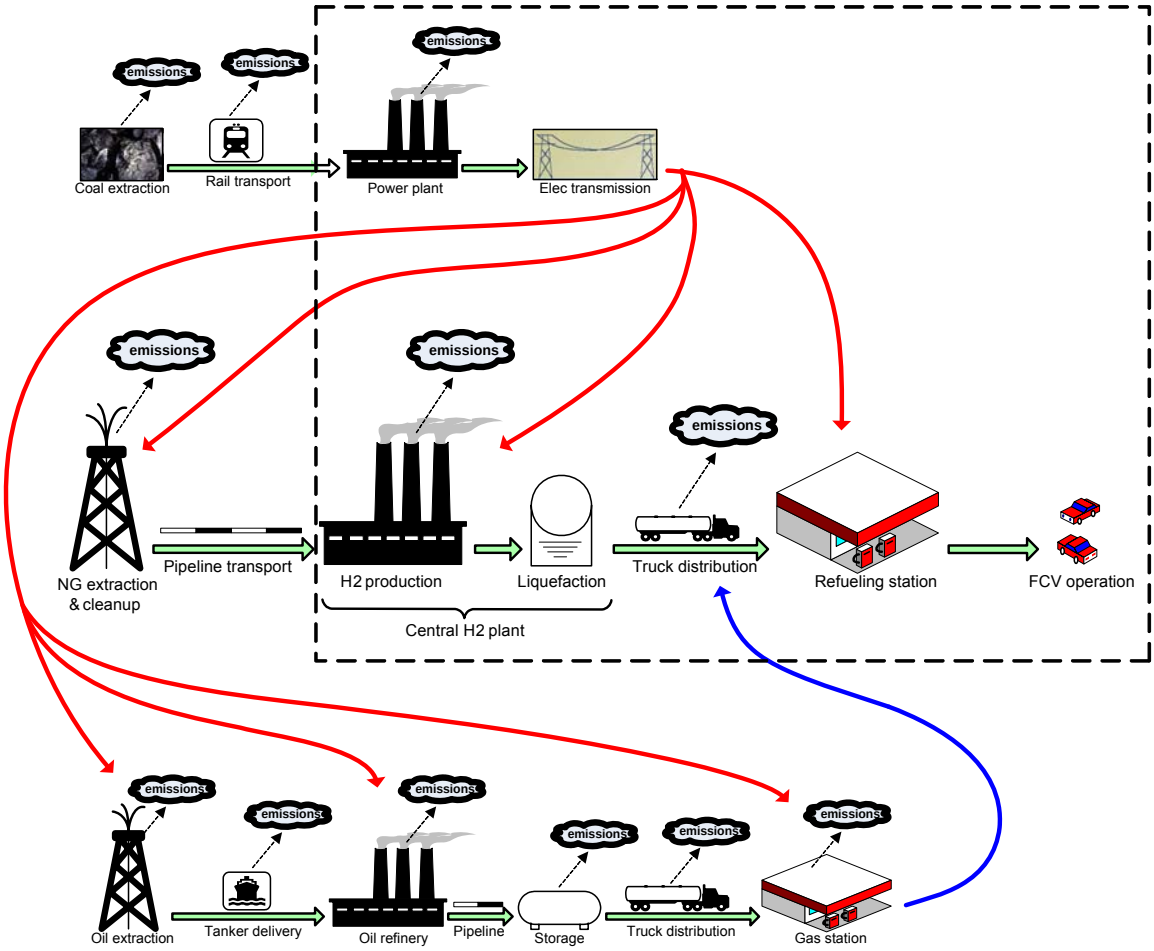

Figure 45. The example of integrated NG-to-H2 pathways (Same as Figure 6)

5.3. Summary of gasoline supply pathway scenarios

See Chapter 4 for more details regarding the gasoline supply pathway scenarios. The brief summary in this section is for the purposes of convenient comparison.

As a point of comparison, we consider gasoline and diesel transportation fuels produced from petroleum and used in internal combustion engine (ICE) vehicles. For simplicity, we use the term “gasoline pathway” to refer to the petroleum-based fuel pathway, including both gasoline and diesel. Figure 46 presents the concept of integrated gasoline pathways; the dashed line area shows the portion of the lifecycle system that is included in this analysis.

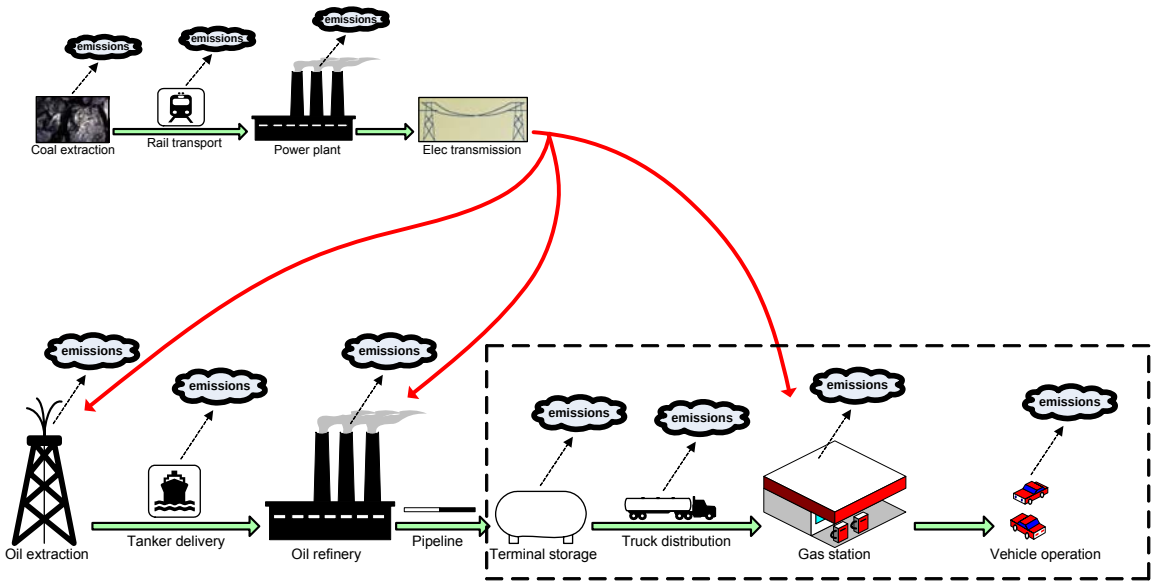

Figure 46. Integrated gasoline or diesel pathways (same as Figure 37)

The vehicle operation stage, in general, plays the most important role in generating emissions and air pollution for a conventional petroleum-based transportation system. In this study, emphasis is given to the two important gasoline pathway steps: vehicle operation and gasoline delivery via diesel trucks.

The following four light duty gasoline fleets are considered in the study. Refer back to Section 4.3 for more details regarding the choice of years.

(1) The year 2025 in-use light duty vehicle fleet. This fleet is a projected in-use mixture of both old and new LD vehicles whose model years range from 1981-2025, spanning 45 years;

(2) The year 2025 new light duty vehicle fleet. This fleet is composed of new LD vehicles only with the model year 2025;

(3) The year 2005 in-use light duty vehicle fleet. This in-use fleet is composed of LD vehicles whose model years range from 1965-2005, spanning 41 years; and

(4) The year 2005 new light duty vehicle fleet. This fleet is composed of new LD vehicles only with the model year 2005.

Applied to the study are a number of models including the regulatory emissions model, transportation planning model, gridded emission inventory model, and air quality model.

The methodology is presented in a flow chart showing the model running sequence (see Figure 47).

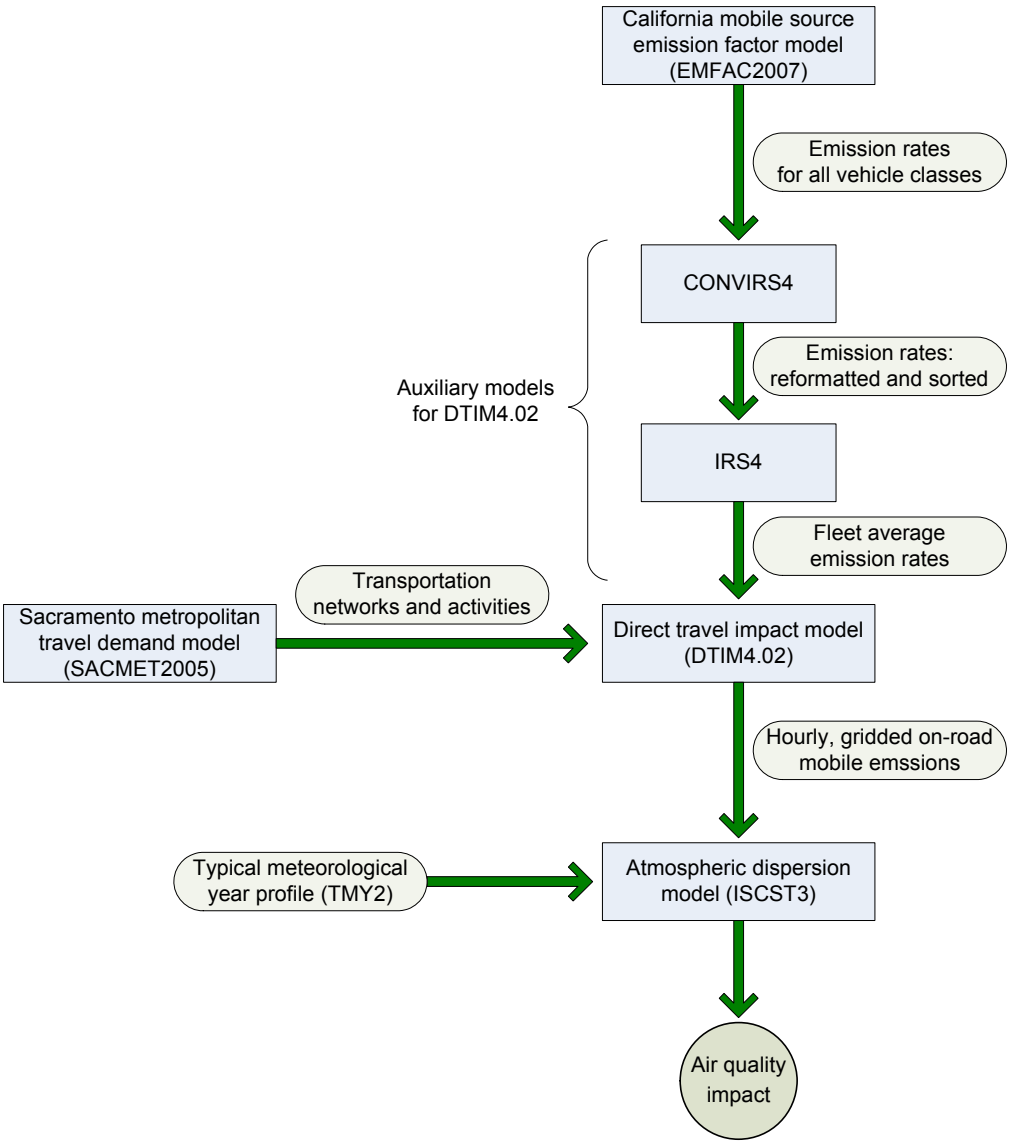

Figure 47. Methodological framework and model running sequence for gasoline fleet scenarios (same as Figure 39)

5.4. Results and discussion

5.4.1. Incremental concentrations caused by hydrogen pathways

To better represent a typical pollution level caused by a hydrogen pathway, we estimate the annual mean incremental concentrations to ambient pollution due to hydrogen pathways, averaged over nine receptors (shown in Table 24), although environmental impacts vary with receptor site.

Table 24. Estimated annual mean incremental concentrations due to hydrogen pathway scenarios (part of Table 9)

Pollutant	# of FCVs served	Onsite pathway ($\mu g/m^3$)	Pipeline pathway ($\mu g/m^3$)	Truck pathway ($\mu g/m^3$)	Sac. 2000 ambient measurement ($\mu g/m^3$)
CO	111,400	0.00473	0.00316	0.00848	639.69
	278,600	0.01423	0.00791	0.02331	
NO_x	111,400	0.00654	0.00896	0.01909	56.64 (NO_2)
	278,600	0.01967	0.02241	0.05394	
VOC	111,400	0.00043	0.00043	0.00114	74.80 (NMOC)
	278,600	0.00126	0.00107	0.00326	
PM_{10}	111,400	0.00046	0.00048	0.00087	22.45
	278,600	0.00146	0.00119	0.00229	

All the three hydrogen pathways would result in negligible pollution levels, compared to ambient measurements. The incremental pollution differs across hydrogen pathways. Among the hydrogen supply options, it is found that the central SMR with pipeline delivery systems is the lowest-pollution option available provided the plant is located to avoid transport of pollutants into the city via prevailing winds. The onsite hydrogen pathway is comparable to the central hydrogen pathway with pipeline delivery systems in

terms of the resulting air pollution. Both pathways are very clean. The truck pathway also results in relatively low pollution levels, but higher than concentrations resulting from the other two hydrogen pathways. Most emissions for the truck pathway are from diesel trucks that deliver the liquid hydrogen and from the electricity used to liquefy the product hydrogen.

The onsite pathway and the pipeline pathway result in very similar ground-level pollution levels, especially in terms of VOC and PM_{10}. However, the onsite pathway leads to more CO and less NO_x pollution than the pipeline pathway. This makes sense because CO tends to be released from incomplete combustion or partial oxidation, which is more likely to occur at a small-scale onsite station, whereas NO_x tends to be released from large stationary sources resulting from combustion, e.g., to produce the high temperature steam which is used in the central hydrogen plant or in a power plant.

5.4.2. Air pollutant concentrations caused by gasoline pathways

The air quality impact of gasoline pathways is the sum of the resulting concentrations due to both the gasoline fleet operation and gasoline-delivery trucks. The estimation approaches and discussions are presented in more detail in Chapter 4.

5.4.2.1. Air pollutant concentrations caused by gasoline fleet operations

Table 25 presents estimated annual mean incremental concentrations to ambient pollution due to gasoline fleet operations, averaged over nine receptors.

Table 25. Estimated annual mean concentrations due to gasoline fleet operations (same as Table 20)

Pollutant	# of gasoline vehicles	In-use 2025 LD fleet ($\mu g/m^3$)	New 2025 LD fleet ($\mu g/m^3$)	In-use 2005 LD fleet ($\mu g/m^3$)	New 2005 LD fleet ($\mu g/m^3$)	Sac. 2005 ambient measurement ($\mu g/m^3$)
CO	111,400	1.58603	0.86292	8.36177	1.61529	601
	278,600	3.96651	2.15807	20.91193	4.03967	
NO_x	111,400	0.07111	0.02759	0.41199	0.07871	43.2 (NO_2)
	278,600	0.17783	0.06900	1.03035	0.19685	
VOC	111,400	0.14320	0.03700	0.61425	0.06312	36.9
	278,600	0.35813	0.09254	1.53617	0.15786	
PM_{10}	111,400	0.01389	0.00369	0.01275	0.00403	20.4
	278,600	0.03474	0.00924	0.03188	0.01008	

5.4.2.2. Air pollutant concentrations caused by gasoline-delivery trucks

To estimate the air quality impact of emissions from gasoline-delivery trucks, the liquid hydrogen (LH2) truck-based concentrations are scaled by using both truck emission factors (for 2025 and 2005, respectively) and needed delivery trips of gasoline vs. LH2, assuming the gasoline storage terminals are about as far away from the refueling stations on average as the central hydrogen plant. Table 26 presents estimated annual mean concentrations due to gasoline-delivery truck emissions for each fleet scenario.

Table 26. Estimated annual mean incremental concentrations due to gasoline-delivery truck emissions (part of Table 22)

Pollutant	# of gasoline vehicles served	In-use 2025 LD fleet (μg/m^3)	New 2025 LD fleet (μg/m^3)	In-use 2005 LD fleet (μg/m^3)	New 2005 LD fleet (μg/m^3)
CO	111,400	0.00155	0.00152	0.01216	0.01209
	278,600	0.00548	0.00536	0.04293	0.04269
NO_x	111,400	0.00276	0.00270	0.01376	0.01368
	278,600	0.00975	0.00954	0.04858	0.04831
VOC	111,400	0.00028	0.00028	0.00145	0.00145
	278,600	0.00099	0.00097	0.00512	0.00510
PM_{10}	111,400	0.00007	0.00007	0.00053	0.00053
	278,600	0.00025	0.00025	0.00187	0.00185

5.4.2.3. Summary

To sum up, Table 27 presents absolute concentrations caused by gasoline pathways, including both pathway components: gasoline vehicle operations and diesel-truck deliveries. We will compare these comprehensive pollution results to those caused by hydrogen pathways in later sections.

Table 27. Estimated annual mean concentrations due to gasoline pathways

Pollutant	# of gasoline vehicles	In-use 2025 LD fleet (μg/m^3)	New 2025 LD fleet (μg/m^3)	In-use 2005 LD fleet (μg/m^3)	New 2005 LD fleet (μg/m^3)	Sac. 2005 ambient measurement (μg/m^3)
CO	111,400	1.58759	0.86444	8.37393	1.62738	601
	278,600	3.97199	2.16343	20.95487	4.08236	
NO_x	111,400	0.07387	0.03029	0.42575	0.09240	43.2 (NO_2)
	278,600	0.18759	0.07854	1.07893	0.24516	
VOC	111,400	0.14348	0.03728	0.61570	0.06457	36.9
	278,600	0.35912	0.09351	1.54129	0.16295	
PM_{10}	111,400	0.01396	0.00376	0.01327	0.00456	20.4
	278,600	0.03499	0.00949	0.03374	0.01193	

5.4.3. Comparison between hydrogen and gasoline pathways

For intuitive comparison purposes, we use the metric “concentration ratio”, which is defined as the incremental ambient concentration resulting from a hydrogen or gasoline pathway relative to that resulting from the hydrogen pathway with pipeline delivery systems. Keep in mind that we are comparing the incremental concentrations, not including any ambient background of CO, VOC, etc. Table 28 summarizes the comparison of concentration ratios relative to the centralized/pipeline hydrogen pathway, for three hydrogen pathway scenarios and four light duty gasoline pathway scenarios, assuming the same size vehicle population. Considering that the VMT level is sometimes used to measure the level of mobility service, Table 29 also summarizes the comparison of concentration ratios relative to the centralized/pipeline hydrogen pathway, assuming the same size VMT level. Tables 27 and 28 present similar patterns of concentration ratios, so we only focus on the results corresponding to the same size population (see Table 28).

Table 28. Comparison of concentration ratios relative to the hydrogen pipeline pathway (assuming the same size vehicle population)

Pollutant	# of FCVs or gas. vehicles	Hydrogen pathways			Gasoline pathways			
		Onsite pathway	Pipeline pathway	Truck pathway	In-use 2025 LD fleet	New 2025 LD fleet	In-use 2005 LD fleet	New 2005 LD fleet
CO	111,400	1.5	1.0	2.7	502	273	2646	514
	278,600	1.8	1.0	2.9	502	273	2648	516
NO_x	111,400	0.7	1.0	2.1	8.2	3.4	47	10.3
	278,600	0.9	1.0	2.4	8.4	3.5	48	10.9
VOC	111,400	1.0	1.0	2.7	336	87	1443	151
	278,600	1.2	1.0	3.1	337	88	1446	153
PM_{10}	111,400	1.0	1.0	1.8	29	7.9	28	9.5
	278,600	1.2	1.0	1.9	29	8.0	28	10.0

Table 29. Comparison of concentration ratios relative to the hydrogen pipeline pathway (assuming the same size VMT)

Pollutant	Total VMT (mile/day)[1]	Hydrogen pathways			Gasoline pathways			
		Onsite pathway	Pipeline pathway	Truck pathway	In-use 2025 LD fleet	New 2025 LD fleet	In-use 2005 LD fleet	New 2005 LD fleet
CO	4,578,082	1.5	1.0	2.7	648	213	3212	399
	11,449,315	1.8	1.0	2.9	649	213	3214	401
NO_x	4,578,082	0.7	1.0	2.1	10.6	2.7	57	8.3
	11,449,315	0.9	1.0	2.4	10.7	2.8	58	9.0
VOC	4,578,082	1.0	1.0	2.7	434	68	1751	118
	11,449,315	1.2	1.0	3.1	435	68	1755	120
PM_{10}	4,578,082	1.0	1.0	1.8	38	6.2	33	7.6
	11,449,315	1.2	1.0	1.9	38	6.2	34	8.1

[1] The numbers of total VMT are based on hydrogen scenarios. Assuming the annual VMT of the hydrogen FCV = 15,000 miles/year, so scenario 1 with 111,400 FCVs has the total VMT = 15,000 miles/year ×111,400 / (365 days/year) = 4,578,082 miles/day. Similarly, scenario 2 with 278,600 FCVs has the total VMT = 11,449,315 miles/day.

Using the centralized/pipeline hydrogen pathway with the same vehicle population as the reference scenario, an onsite hydrogen pathway has an almost comparable impact on air quality, especially for VOC and PM_{10} concentrations. The onsite pathway would lead to slightly greater CO and lower NO_x concentrations, as CO tends to be released from incomplete combustion or partial oxidation that more often occurs at a small-scale onsite station, whereas NO_x tends to be released from large-scale stationary sources related to high temperature processes, such as those found at a central hydrogen or power plant. The centralized hydrogen production with liquid hydrogen truck delivery pathway is the least clean option among these three means of hydrogen supply, for the four pollutant types examined in the study. The extra energy consumed to liquefy hydrogen plays an important role, and the diesel-truck delivery also matters in terms of emissions and air pollution.

As today's representative of LD fleet operations, the current gasoline pathways with the model year 2005 vehicles would lead to 510 times greater CO, 150 times greater VOC, 10 times greater PM_{10}, and 10 times greater NO_x concentrations than those caused by the centralized/pipeline hydrogen pathway (see Table 28), assuming the same number of vehicles.

As the least polluting representative of LD gasoline/diesel fleet operations, the gasoline pathways with the future evolved model year 2025 vehicles (advanced gasoline vehicles) would lead to 273 times greater CO, 88 times greater VOC, 8 times greater PM_{10}, and 3.5 times greater NO_x concentrations than those caused by the centralized/pipeline hydrogen

pathway (see Table 28), assuming the same number of vehicles. The resulting CO and VOC pollution is even about two orders of magnitude greater than that of the truck pathway, the least clean hydrogen scenario. Again, the 2025 new fleet pathways cause significantly higher PM_{10} and NO_x pollution than any of the hydrogen pathways. Considering that we have not included emissions from the gasoline terminal storage and refueling stations, it is safe to say that hydrogen pathways with year 2005 production technology would be less polluting than any LD fleet (both in-use and new) scenarios examined in the study, from a lifecycle analysis perspective.

In short, introducing any hydrogen pathway analyzed in the study would improve urban air quality compared to the 2005 in-use scenario in the following order of concentration ratio: CO (greatest concentration ratio) > VOC >> NO_x > PM_{10} (lowest concentration ratio).

For the new fleet scenarios, the model year 2025 vehicles, at the aggregate level, would be dramatically more advanced than the model year 2005 vehicles in terms of environmental impacts (to be accurate, with the exception of slight improvement for PM_{10}). Thus, it is not necessary to compare hydrogen pathways with the 2005 new fleets (and, hence, the in-use fleets in 2005 or 2025), as we have done this for the 2025 new fleets. That is, the superiority of the hydrogen pathway scenarios, compared to the best-case gasoline scenario of all-new 2025 vehicles, is even more pronounced when comparing to gasoline scenarios involving older vehicles. This reflects the trends in new vehicle emission performance in California: new vehicles are getting cleaner over time.

Also, due to the methodological limitation of directly using travel demand model data (from SACMET), transportation-related air pollution tends to be underestimated (Wang et al., 2007), meaning that the improvement offered by hydrogen pathways is even greater than shown here.

Note, again, that hydrogen demand is estimated based on the population in urbanized Sacramento, whereas the SACMET2005 modeling domain is made up of six counties. This is not an inconsistency, as hydrogen vehicles could be running everywhere in the six-county region; they are zero emission vehicles (ZEVs), so we do not need the precise locations of hydrogen FCVs.

5.5. Conclusions

We have examined the potential regional air quality impacts of hydrogen transportation fuel from a lifecycle analysis (LCA) perspective, including impacts from fuel production, fuel delivery, and vehicle use. Four ground-level pollutants, namely CO, NO_x, VOC, and PM_{10}, were considered. We assumed two population levels of hydrogen fuel cell vehicles in Sacramento, California. Three hypothetical natural gas-to-hydrogen pathways were examined: onsite hydrogen production via small-scale steam methane reforming (SMR), central SMR production with gaseous hydrogen pipeline delivery, and central SMR production with liquid hydrogen truck delivery.

For comparison purposes, we have also developed gasoline/diesel pathway scenarios for the calendar years 2005 and 2025, respectively. The 2005 light duty (LD) fleet is used as the baseline, which corresponds to current transportation technology. To reflect the improvements in vehicle/fuel technologies and standards over time, we use the 2025 light duty fleet as representative of advanced gasoline vehicles in the near future. We sequentially applied a series of models to estimate the contributions of light duty fleets to urban air pollution using travel forecasting model data. The modeling procedure started from the California regulatory emissions model (EMFAC) and Sacramento Metropolitan travel demand model (SACMET) to the hourly, gridded emission inventory model (DTIM) and air quality dispersion model (ISC).

Using the centralized/pipeline hydrogen pathway with the same vehicle population as the reference scenario, an onsite hydrogen production pathway has an almost comparable impact on air quality, especially for VOC and PM_{10} concentrations; the onsite pathway would lead to slightly greater CO and lower NO_x concentrations. Centralized hydrogen production with liquid hydrogen truck delivery is the least clean option among these three means of hydrogen supply, for the four pollutants examined in the study.

All the gasoline pathways studied yield much higher pollutant concentrations than the hydrogen pathways. As today's representative of LD fleet operations, the current gasoline pathways with the model year 2005 vehicles would lead to 510 times greater CO, 150 times greater VOC, 10 times greater PM_{10}, and 10 times greater NO_x concentrations than those caused by the centralized/pipeline hydrogen pathway, assuming the same number

of vehicles. As the least polluting representative of LD gasoline/diesel fleet operations, the gasoline pathways with the future evolved model year 2025 vehicles (advanced gasoline vehicles) would lead to 273 times greater CO, 88 times greater VOC, 8 times greater PM_{10}, and 3.5 times greater NO_x concentrations than those caused by the centralized/pipeline hydrogen pathway, assuming the same number of vehicles. In short, in a future year like 2025, hydrogen pathways with year 2005 production technology would be less polluting than any LD fleet (both in-use and new) scenarios examined in the study, from a lifecycle analysis perspective.

There is scope to greatly reduce the air quality impact of gasoline vehicles, with newer, cleaner gasoline technologies. Given the same number of vehicles, the in-use 2025 conventional LD fleet scenario results in much less pollution, with the exception of PM_{10}, than the in-use 2005 conventional LD fleet scenario, which certainly reflects the expected improvements in vehicle/fuel technologies and standards in California. Similarly, for a scenario year, the new LD vehicle fleet (i.e., containing only vehicles of the latest model year) would lead to much lower concentrations than the in-use fleet, assuming the same population size. On a per vehicle basis, a new vehicle of the model year 2025 is cleaner than a new vehicle of the model year 2005 in terms of resulting ambient concentrations of all four pollutants: CO, NO_x, VOC, and PM_{10}. However, the contribution of primary PM_{10} to ambient concentrations from the model year 2025 vehicle are just slightly lower than those from the model year 2005 vehicle.

This study quantitatively compares hydrogen and gasoline pathways in terms of the resulting air pollution. The results show that advanced gasoline vehicle technologies are a promising route to reducing air quality impacts of light duty vehicles; however, with hydrogen vehicle technologies, the contribution of light duty vehicles to ambient air pollutant concentrations could be reduced even further, to near-zero.

6. Summary and Conclusions

One of the key motivations for hydrogen as an alternative transportation fuel is its potential to help improve urban air quality. Although hydrogen fuel cell vehicles (FCVs) emit no tailpipe emissions, hydrogen must be produced from other sources and delivered to users. These steps can generate air pollutant emissions. Thus, the entire lifecycle from well to wheels must be considered in an assessment of hydrogen's air quality impacts.

In this dissertation, we have examined the potential regional air quality impacts of hydrogen transportation fuel from a lifecycle analysis (LCA) perspective, including impacts from fuel production, delivery, and vehicle use. The analysis is a case study for a specific region, Sacramento, California. Three natural gas-based hydrogen supply pathways are considered: onsite production via small-scale steam methane reformer (SMR), large-scale centralized SMR with liquid hydrogen truck delivery, and large-scale centralized SMR with gaseous hydrogen pipeline delivery. The lifecycle emission inventories and optimized spatial layouts of hydrogen infrastructure are determined, considering geographic factors.

We use the urbanized Sacramento conventional light duty (LD) fleet in 2000 as the baseline. We consider two levels of market penetration where 9% or 20% of the light duty fleet are hydrogen fuel cell vehicles. We keep the number of year 2000 gasoline vehicles constant in both scenarios, and add hydrogen vehicles and hydrogen fuel supply systems to the region. Thus, the total vehicle population is the sum of the year 2000 light

duty gasoline fleet and the added hydrogen vehicles. This allows us to estimate the incremental impact of hydrogen energy systems on ambient pollution levels in the Sacramento area, without the complexities of simultaneously reducing the number of gasoline vehicles.

Because we consider only physical transport of primary pollutants, the assumed background ambient pollution levels do not influence the results for the incremental ambient concentrations due to hydrogen vehicles. Therefore, the two market penetration scenarios (9% and 20%) can be presented in the following way. The 9% scenario explores the resulting concentrations of pollutants from a hydrogen system with 111,400 fuel cell vehicles, a number equal to 10% of the total LD gasoline fleet in urbanized Sacramento in the year 2000. Similarly, The 20% scenario explores the resulting concentrations of pollutants from 278,600 fuel cell vehicles, a number equal to 25% of the total gasoline fleet in urbanized Sacramento in 2000.

In the first project, we consider five directly-emitted primary pollutants, i.e., carbon monoxide (CO), nitrogen oxides (NO_x, referring to both NO and NO_2), volatile organic compounds (VOCs), particulate matter (PM_{10}), and sulfur oxides (SO_x, referring roughly to SO_2 here). Using data for typical meteorological conditions, we run an atmospheric dispersion model, ISCST3, to estimate the incremental pollutant concentrations resulting from each of the three hydrogen pathways.

The pollution levels associated with each of the hydrogen scenarios are dependent upon the location of emitters and receptors, regional meteorological conditions, and geographic factors. The spatial layout of pathway steps therefore plays an important role in determining ambient pollution levels at air quality monitoring stations. We find that all the hydrogen pathways considered are associated with extremely low pollution levels relative to current ambient air concentrations of NO_x, CO, VOC, particulates, and SO_x. For the hydrogen scenarios studied, the results are typically less than 0.1% of the current ambient pollution.

Using the centralized/pipeline hydrogen pathway with the same vehicle population as the reference scenario, an onsite hydrogen production pathway has an almost comparable impact on air quality, especially for VOC and PM_{10} concentrations; the onsite pathway would lead to slightly greater CO and lower NO_x concentrations. Centralized hydrogen production with liquid truck delivery has a greater impact on air quality relative to the other pathways due to emissions associated with diesel trucks and electricity consumption to liquefy hydrogen. Again, compared to ambient concentrations, all the three hydrogen pathways result in negligible air pollution in the region.

Emission source contributions to air pollutant concentrations vary by pathway. In general, there are the following three important emission sources: hydrogen production facility, electric power plants, and delivery trucks. For the pipeline or onsite pathway, hydrogen production accounts for the largest share of pollution. For the electricity-intensive liquid hydrogen truck pathway, emissions from diesel-truck delivery and electric generation at

power plants are much more important than hydrogen plant emissions in terms of the resulting pollution.

In the second project, we estimate the potential impacts of the above hydrogen pathways on secondary ozone (O_3) formation. A region-specific linear regression model was developed to link the peak ozone concentrations to ambient meteorological conditions and the early morning ambient VOC and NO_x as the precursors to ozone formation. The regression model and data were limited to the Sacramento region and the time period from July 3, 2004 to October 26, 2004. The model shows that increases in precursor concentrations do not necessarily increase the peak ozone concentration, and may even cause it to decrease.

Since the current light duty fleet is held constant and additional hydrogen cars are added to the fleet, the incremental VOC and NO_x pollution resulting from lifecycle emissions of all hydrogen pathways is a positive quantity. All the hydrogen pathways would result in very small (either negative or positive) changes in ozone air quality. In some cases worse ozone air quality (mostly in July, August, and September) occurred, and peak ozone concentration increments increased in the following order: onsite pathway < pipeline pathway < truck pathway. In some cases better ozone air quality was predicted to occur (mostly in October), and the truck and onsite pathways had a greater impact than the pipeline pathway. The truck pathway tended to lead to a much wider fluctuation in degradation or improvement of ozone air quality: percentage changes in peak ozone concentrations were approximately -0.01% to 0.04% for the 9% market penetration, and

approximately -0.03% to 0.1% for the 20% market penetration. Note that the 20% onsite pathway occasionally resulted in a decrease of around -0.1% of background ozone pollution. So the positive and negative limits of changes in ozone pollution would be around one thousandth of current pollution levels. Compared to the current ambient pollution level, the truck pathways (and therefore the onsite and pipeline pathways) are unlikely to cause a serious ozone problem for market penetrations of hydrogen FCVs in the 9-20% range.

In the third project, we developed gasoline pathway scenarios, for comparison purposes, for the calendar years 2005 and 2025 in Sacramento, California for four pollutants: CO, NO_x, VOC, and PM_{10}. For simplicity, we use the term “gasoline pathway” to refer to the petroleum-based fuel pathway, including both gasoline and diesel transportation fuels.

The 2005 light duty fleet is used as the baseline, which corresponds to current transportation technology. To reflect the improvements in vehicle/fuel technologies, we use the 2025 light duty fleet as representative of advanced gasoline vehicles in the near future. We sequentially applied a series of models to estimate the contributions of light duty fleet operations to urban air pollution. The modeling procedure started from the California regulatory emissions model (EMFAC) and Sacramento Metropolitan travel demand model (SACMET) to the hourly, gridded emission inventory model (DTIM) and air quality dispersion model (ISCST3).

Still, using the centralized/pipeline hydrogen pathway with the same vehicle population as the reference scenario, all the gasoline pathways studied yield much higher pollutant concentrations than the hydrogen pathways. As today's representative of LD fleet operations, the current gasoline pathways with the model year 2005 vehicles would lead to 510 times greater CO, 150 times greater VOC, 10 times greater PM_{10}, and 10 times greater NO_x concentrations than those caused by the centralized/pipeline hydrogen pathway, assuming the same number of vehicles. As the least polluting representative of LD gasoline/diesel fleet operations, the gasoline pathways with the future evolved model year 2025 vehicles (advanced gasoline vehicles) would lead to 273 times greater CO, 88 times greater VOC, 8 times greater PM_{10}, and 3.5 times greater NO_x concentrations than those caused by the centralized/pipeline hydrogen pathway, assuming the same number of vehicles. In short, in a future year like 2025, hydrogen pathways with year 2005 production technology would be less polluting from a lifecycle analysis perspective than any LD fleet (both in-use and new) scenarios examined in the study.

There is scope to greatly reduce the air quality impact of gasoline vehicles, with newer, cleaner gasoline technologies. Given the same number of vehicles or on a per vehicle basis, the in-use 2025 conventional LD fleet scenario results in much less pollution, with the exception of PM_{10}, than the in-use 2005 conventional LD fleet scenario, which certainly reflects the expected improvements in vehicle/fuel technologies driven by evolving emissions standards in California. Similarly, for a scenario year, the new LD vehicle fleet (i.e., containing only vehicles of the latest model year) would lead to much lower concentrations than the in-use fleet, assuming the same population size. On a per

vehicle basis, a new vehicle of the model year 2025 is cleaner than a new vehicle of the model year 2005 in terms of the resulting ambient concentrations of all four pollutants: CO, NO_x, VOC, and PM_{10}. These results indicate that the fleet turnover plays an important role in reducing vehicular emissions and improving air quality.

In conclusion, this dissertation quantitatively compares hydrogen and gasoline pathways in terms of the resulting air pollution, from a lifecycle emissions perspective. The results show that advanced gasoline vehicle technologies are a promising route to reducing air quality impacts of light duty vehicles; however, with hydrogen vehicle technologies, the contribution of light duty vehicles to ambient air pollutant concentrations could be reduced even further, to near-zero. Moreover, adding a significant number of hydrogen fuel cell vehicles to the region would have a very small impact on ozone pollution as well; in fact, it does not necessarily increase the peak ozone concentration, and may even cause it to decrease in some cases. The findings have meaningful implications for policy makers as they evaluate hydrogen, a promising alternative transportation fuel.

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