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Determining Air Quality and Greenhouse Gas Impacts of Hydrogen Infrastructure and Fuel Cell Vehicles

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Adoption of hydrogen infrastructure and hydrogen fuel cell vehicles (HFCVs) to replace gasoline internal combustion engine (ICE) vehicles has been proposed as a strategy to reduce criteria pollutant and greenhouse gas (GHG) emissions from the transportation sector and transition to fuel independence. However, it is uncertain (1) to what degree the reduction in criteria pollutants will impact urban air quality, and (2) how the reductions in pollutant emissions and concomitant urban air quality impacts compare to ultralow emission gasoline-powered vehicles projected for a future year (e.g., 2060). To address these questions, the present study introduces a “spatially and temporally resolved energy and environment tool” (STREET) to characterize the pollutant and GHG emissions associated with a comprehensive hydrogen supply infrastructure and HFCVs at a high level of geographic and temporal resolution. To demonstrate the utility of STREET, two spatially and temporally resolved scenarios for hydrogen infrastructure are evaluated in a prototypical urban airshed (the South Coast Air Basin of California) using geographic information systems (GIS) data. The well-to-wheels (WTW) GHG emissions are quantified and the air quality is established using a detailed atmospheric chemistry and transport model followed by a comparison to a future gasoline scenario comprised of advanced ICE vehicles. One hydrogen scenario includes more renewable primary energy sources for hydrogen generation and the other includes more fossil fuel sources. The two scenarios encompass a variety of hydrogen generation, distribution, and fueling strategies. GHG emissions reductions range from 61 to 68% for both hydrogen scenarios in parallel with substantial improvements in urban air quality (e.g., reductions of 10 ppb in peak 8-h-averaged ozone and 6 μg/m³ in 24-h-averaged particulate matter concentrations, particularly in regions of the airshed where concentrations are highest for the gasoline scenario).

1. Introduction

Future transport of people and goods will be constrained by limits on criteria pollutant emissions, scarcity of hydrocarbon fossil fuel resources, and greenhouse gas (GHG) regulation (1). The use of hydrogen in combination with fuel cells for vehicle power is proposed as a new paradigm to meet future transportation demands in the face of these challenges (2).

Although studies widely agree that the implementation of hydrogen infrastructure will reduce air pollutant emissions from the transportation sector (3–7), the extent to which air quality in an urban airshed will be affected by these reductions is a more complex matter than quantifying emissions. Wang, et al. (2007), (2008) broadly applied an empirical air quality formula to emissions reductions from hydrogen infrastructure, but neglected to simulate detailed atmospheric chemistry and transport mechanisms that lead to the formation of secondary pollutants (6, 8). Understanding air quality requires detailed and extensive modeling efforts to account for atmospheric chemistry, transport, deposition, meteorological conditions, regional geography, and other physical phenomena that affect the balance of tropospheric chemical species (9). Jacobson, et al. (2005) made an important contribution by simulating the air pollution effects of replacing gasoline vehicles with hydrogen fuel cell vehicles (HFCVs) (4). The Jacobson study considered a coarse resolution over the whole U.S. and scenarios developed on the basis of discrete production and delivery strategies for the provision of hydrogen fuel, that is, one hydrogen production and delivery method at a time. Current interest in reducing greenhouse gases and improving urban air quality coupled with readiness to demonstrate and deploy alternative transportation fuel infrastructure and vehicle technologies has created a need for the capability to simulate the environmental impacts of potentially realistic energy scenarios that are simulated at a higher resolution and with the option of integrating and comparing a variety of fuel, energy, and vehicle strategies. For the first time, this study compares the air quality impacts of fully integrated hydrogen infrastructure scenarios in an urban airshed by introducing a spatially and temporally resolved scenario development and analysis methodology. The methodology—referred to as the spatially and temporally resolved energy and environment tool (STREET)—characterizes the pollutant and GHG emissions associated with a comprehensive hydrogen supply infrastructure and HFCV deployment at a high level of geographic and temporal resolution. The methodology then follows with detailed simulations of atmospheric chemistry and transport in the prototypical urban airshed to produce tropospheric ozone and particulate matter (PM) signatures. This study is also the first to quantify GHG emissions from fully integrated hydrogen infrastructure scenarios designed with high geographic and temporal resolution. Previous efforts to model GHG impacts of hydrogen infrastructure deployment have included only discrete production and delivery strategies for the provision hydrogen fuel at a regional or national scale (3–8).
The South Coast Air Basin of California (SoCAB) is chosen as the urban region of interest in this study for several reasons: it ranks among the worst in the United States with respect to air quality (10), it is the most extensively studied airshed, and it serves as a test bed for hydrogen infrastructure and HFCV deployment (11). Detailed hydrogen infrastructure scenarios are designed by using geographic information systems (GIS) data (12, 13) to allocate spatially hydrogen infrastructure sufficient to service the SoCAB in the year 2060. Other factors required to characterize fully a hydrogen infrastructure scenario in 2060 (market expectations, emissions and operating characteristics of hydrogen infrastructure devices, energy requirements, etc.) have been established previously on the basis of measurements from technology performance, archival publications, reports, and input from experts (3). Hydrogen scenarios are analyzed with respect to criteria pollutant and GHG emissions using the preferred combination assessment (PCA) methodology (3), which integrates several hydrogen technologies to assess the performance of the hydrogen supply chain on a life cycle basis. GHG emissions are quantified and compared to conventional vehicles. The University of California, Irvine: California Institute of Technology (UCI-CIT) atmospheric chemistry and transport model is then used to analyze air quality in the South Coast Air Basin (SoCAB) of California based on the spatially and temporally resolved primary pollutant emissions that are generated from the PCA model. These two capabilities, PCA integrated with the UCI-CIT airshed model, form the methodology “STREET.”

2. STREET Methodology

2.1. Scenario Development. The first step in applying STREET is to establish infrastructure and vehicle scenarios for the future years selected for analysis. In the present case, two hydrogen infrastructure scenarios are developed for 2060 with the assumption that HFCV comprise 75% of the passenger vehicle fleet. 2060 is chosen for the purpose of analyzing the greatest potential impacts produced by a high penetration of HFCV, which is not expected to occur until 2060 (14). While both scenarios integrate a mix of hydrogen feedstocks, generation technologies, distribution and fueling technologies, one relies more heavily on renewable energy sources for hydrogen generation (Scenario HR) and the other more heavily on fossil fuel energy sources (Scenario HF). The approach of designing two scenarios for comparison is used to address the uncertainty related to technology choices throughout the evolution of hydrogen infrastructure in future years. Table 1 presents the HFCV population, fuel demand, and technology allocated to the generation, distribution, and dispensing of hydrogen in each scenario. Note that the majority of the fuel requirements in both scenarios are produced in the airshed. Only coal-based and large renew-

<table>
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<th>Table 1. Hydrogen Infrastructure Scenarios Implemented for the South Coast Air Basin (SoCAB) of California in the Year 2060.a</th>
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<td><strong>H₂ generated from more renewable sources</strong></td>
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<td>140 bar gaseous fueling</td>
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<td>350 bar gaseous fueling</td>
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a HFCV comprise 75% of passenger vehicles and various hydrogen generation, distribution, and fueling technologies are used to meet hydrogen demand. Scenario-HR includes more renewable primary energy sources for hydrogen generation, whereas Scenario-HF includes more fossil fuel sources. Hydrogen distribution depends upon generation technologies, trucking and pipeline infrastructure. Represents 75% of expected passenger vehicle kilometers traveled (VKT) in the SoCAB, year 2060 (ref-EMFAC). Coal-fired integrated gasification combined cycle plant with carbon capture and storage cogenerating hydrogen and electricity. Petroleum coke-fired integrated gasification combined cycle plant cogenerating hydrogen and electricity. Electrolysis is powered by mostly large renewable wind and solar facilities outside the air shed and some nuclear facility. Cogeners hydrogen, electrical power, and heat using a low emissions high temperature fuel cell. Electrolysis is powered by photovoltaic electricity.
able-based hydrogen generation facilities are located outside the airshed. A gasoline vehicle scenario (Scenario G) for 2060 serves as the basis for comparison. All nonpassenger vehicle emissions for 2060 are derived from estimates made by the South Coast Air Quality Management District (SCAQMD) to demonstrate attainment with ozone standards in the SoCAB by the year 2023 (15). The resulting emissions inventory is applied to the year 2060 therein assuming that emissions will not exceed those estimated to achieve attainment in 2023. Conventional passenger vehicle emissions projected for the year 2060 are extrapolated based upon California Air Resources Board (CARB) projections of a future passenger vehicle fleet and associated emissions. The projection accounts for the gradual retirement of old vehicles and introduction of new vehicles compliant with the Low Emission Vehicle II (LEV II) Standards, adopted by the California Air Resources Board through the year 2010 (16). As a result, 2060 gasoline vehicle emissions are projected to be 70% lower than 2008 levels.

2.2. Spatial and Temporal Allocation of Infrastructure. Figure 1 illustrates the spatial allocation of hydrogen infrastructure developed in this study. To locate hydrogen fueling stations, GIS data are extracted from a database of existing retail gasoline stations providing spatial information. High HFCV efficiency and improved information technology on-board passenger vehicles allows for the introduction of fewer hydrogen fueling stations in 2060 compared to gasoline stations in 2008. Sites for hydrogen fueling stations are determined by randomly removing a portion of current gasoline stations while keeping the relative local density of future hydrogen stations consistent with today’s gasoline stations.

Sites for distributed and centralized hydrogen generation facilities are selected by applying screening criteria that are unique to the type of facility. Screening criteria include proximity to existing fueling stations, GIS land use allocation (12), location of existing steam methane reforming (SMR) and power generation facilities, proximity to infrastructure such as gas pipelines and roads, and location of wind and solar resources. Several hydrogen generation technologies, such as petroleum coke plants, coal integrated gasification combined cycle (IGCC) plants, and high temperature fuel cells cogenerate hydrogen and electric power. The latter is absorbed by the SoCAB’s projected growth in electrical power demand in the SoCAB between now and 2060. Hydrogen is transmitted and distributed as a pressurized gas via pipeline or as a cryogenic liquid by truck. Also accounted for is the energy required for pressurization or liquefaction of the hydrogen. Remote pipelines transporting hydrogen generated outside the SoCAB to the urban region are sited on existing pipelines corridors (18) and are sized according to the output of each generation facility. An urban pipeline network located at existing pipeline corridors distributes hydrogen from central facilities to hydrogen fueling stations throughout the developed regions of the SoCAB. Likely truck delivery routes for hydrogen are determined with “closest facility” algorithms (19). Emissions from improved diesel combustion engine trucks are spatially and temporally allocated accordingly.
2.3. Air Quality Model. Spatially and temporally resolved air pollution emissions are developed to serve as input to the UCI-CIT atmospheric chemistry and transport model used to analyze air quality in the SoCAB. The UCI-CIT model includes the CalTech Atmospheric Chemistry Mechanism (CACM) (20–22), which is intended for use in three-dimensional urban/regional atmospheric models with O₃ formation and secondary organic aerosol (SOA) production. Solution of the atmospheric chemistry is coupled in a set of dynamic atmospheric transport equations with state-of-the-art solvers in an Eulerian frame of reference with 5 x 5 km horizontal resolution (23, 24). The horizontal resolution requires the high level of detail in the spatial distribution of emissions that are produced by each of the hydrogen scenarios. Vertical resolution is in five variable height cells up to 1100 m using terrain following coordinates.

Meteorological conditions are from the Southern California Air Quality Study (SCAQS); a comprehensive campaign of atmospheric measurements that occurred in the SoCAB during August 27–29, 1987. Resulting data have been utilized widely to validate air quality models (25–27). Zeldin et al. found that August 28, 1987 is representative of the meteorological conditions in the SoCAB, making it suitable for modeling an air quality episode (28). In addition, the August 27–28, 1987 episode is statistically within the top 10% of severe ozone-forming meteorological conditions. The SCAQS August 27–29, 1987 episode is characterized by a weak onshore pressure gradient and warming temperatures aloft, a sea breeze during the day, and a weak land–mountain breeze at night. The presence of a well-defined diurnal inversion layer at the top of neutral and unstable layers near the surface, along with a slightly stable nocturnal boundary layer, facilitates the accumulation of pollutants throughout the SoCAB.

Simulations are conducted using the aforementioned episode twice to create a six-day time period. The first three days of simulation are used to dissipate the effects of initial conditions, as three days has been found sufficient for the South Coast Air Basin of California (29). Air quality results are based upon the ground-level concentrations obtained in the sixth day of simulation, represented by the meteorological conditions of August 29 of the episode.

3. Results and Discussion

3.1. Air Quality Impacts of Hydrogen Infrastructure Scenarios. High penetration of HFCV and hydrogen infrastructure substantially improves air quality in the SoCAB with respect to ground-level concentrations of both peak ozone and PM of 2.5 µm or smaller (PM₁.₅), as shown in Figure 2. Concentrations of ozone and PM₁.₅ are modeled for a typical ozone episode for 2060 using the gasoline vehicles emissions case (Scenario G). Reductions in stationary and mobile source emissions expected to occur between now and the year 2060 are accounted for in Scenario G. The use of HFCV by comparison would further reduce pollutant emissions. Consequently, when pollutant formation in hydrogen infrastructure Scenarios HR and HF is compared to that of the gasoline case, significant improvements in peak 8-h ozone and 24-h PM₁.₅ are observed. With respect to peak ozone, the most dramatic reductions (10 ppb) occur in the northeastern region of the SoCAB. Figure 2(a) shows that peak Scenario G ozone concentrations of 120 ppb occur in the same region of the airshed. Similarly the largest reductions in PM₁.₅ (6 µg/m³) occur where peak Scenario G concentrations are observed, in the midwest region of the SoCAB, just northeast of Riverside. In both Scenarios HR and HF the improvement in air quality and ability to identify localized improvements, is attributed to differences in the quantity and distribution (both spatial and temporal) of air pollution emissions associated with hydrogen infrastructure and HFCV use compared to conventional passenger vehicles.

Absolute and localized decreases in 8-h peak ozone and 24-h PM₁.₅ are observed throughout the entire SoCAB in Scenario HR, presented in Figure 2(b). In this scenario, emissions are introduced into the basin by hydrogen generation and distribution activities. But due to the displacement of emissions from conventional passenger vehicles, petroleum refineries, and gasoline delivery trucks, emissions levels are generally reduced throughout the SoCAB relative to Scenario G. From the spatial and temporal resolution of the hydrogen infrastructure scenarios, it becomes apparent that significant reductions in total daytime emissions occur locally in regions of the airshed critical to secondary pollutant formation. Decreases in absolute peak ozone and PM₁.₅ are also observed in Scenario HF, presented in Figure 2(c). However, spatial and temporal distribution produces the observation that in contrast to Scenario HR, Scenario HF produces localized increases in 8-h peak ozone and 24-h PM₁.₅. This difference is attributed to the operation of two petroleum coke hydrogen generation facilities in Scenario HF and the associated spatial and temporal distribution of emissions. Importantly, the introduction of petroleum coke power generation facilities with carbon sequestration was recently under consideration in the SoCAB (30). Compared to Scenario G, Scenario HF localized emissions increase for most of the day near the two petroleum coke facilities in Scenario HF. In particular, the increase in localized NOₓ emissions generates ozone in Scenario HF. NOₓ reacts with oxidized products of volatile organic compounds (VOCs) leading to pockets of increased ozone concentrations east of Los Angeles, downwind from the petroleum coke facilities. Ozone concentrations also increase along the coast due to air recirculation. Nighttime sea breezes blow NOₓ emissions offshore and low NOₓ and ozone concentrations over the ocean provide a more efficient medium for ozone production compared to areas with high NOₓ. Despite this increase, ozone concentrations remain low in the coastal areas overall.

Localized increases in PM₁.₅ northeast of Long Beach and in the northeastern region of the SoCAB that are observed in Scenario HF are linked to NOₓ and SOₓ emissions from the petroleum coke facilities. NOₓ and SOₓ are converted to nitric and sulfuric acid, respectively, which react with ammonia to produce PM. With regards to prevailing daytime winds, the two distinct pockets of increased PM₁.₅ occur downwind of the petroleum coke facilities. SOₓ emissions, which aerosolize in less time relative to NOₓ, react earlier. Therefore, SOₓ accounts for the pocket of increased PM₁.₅ northeast of Long Beach and closer to the petroleum coke facilities in Figure 2(c). NOₓ emissions, which react more slowly than SOₓ emissions and produce PM, account for the pocket of increased PM₁.₅ in the northeastern region of the SoCAB. Regions of peak Scenario G concentrations of 8-h ozone and 24-h PM₁.₅ show significant improvements in Scenario HF despite petroleum coke plants operating in the SoCAB. Where local pollutant concentrations are highest in Scenario G, Scenario HF reductions of ozone and PM₁.₅ are comparable to those of Scenario HR (10 ppb and 6 µg/m³, respectively).

3.2. GHG Impacts of Hydrogen Infrastructure Scenarios. The implementation of hydrogen infrastructure scenarios HR and HF leads to substantial reductions in well-to-wheels (WTW) GHG emissions from the SoCAB passenger vehicle fleet compared to Scenario G. The extent to which reductions occur is shown in Figure 3(a). Scenario HR leads to a 63% reduction in GHG emissions from SoCAB passenger vehicles and Scenario HF a 59% reduction when they are compared to Scenario G. Figure 3(a) also shows that a majority of the
GHG emissions associated with passenger vehicles in Scenarios HR and HF come from the remaining 25% of gasoline vehicles of which the total fleet is comprised: 68% in Scenario HR and 61% in Scenario HF. Passenger vehicles currently account for 28.6% of California’s total GHG emissions and are projected to account for 27.0% in 2020 (31) suggesting that HFCV deployment can play a significant role in California’s overall GHG reduction goals. The reductions in GHG emissions from Scenarios HR and HF relative to conventional vehicles are attributed to efficiency advantages of HFCV over gasoline ICE vehicles, reduced GHG intensity of hydrogen generation strategies compared to WTW gasoline combustion, and carbon capture from coal IGCC facilities that cogenerate hydrogen and electricity in both Scenarios HR and HF. It is valuable to mention that this study does not consider carbon sequestration for SMR and petroleum coke.
IGCC facilities. The inclusion of carbon capture in these hydrogen generation technologies can further reduce GHG emissions associated with hydrogen infrastructure and HFCV.

3.4. Discussion of Potential Feedback Effects of Decreases in Pollution Concentrations. It is important to recognize potential feedback effects resulting from notable decreases in pollutant concentrations and GHG emissions even though they are not modeled in this study. For instance, ozone formation increases with increasing temperatures implying that reducing radiative forcing could lead to further reductions in ozone formation above those that are presented (29). Reductions in O₃ and other GHG observed in the scenarios above would contribute to reductions in atmospheric radiative forcing and thus dampen the increases in temperature projected throughout the decades spanning to the year 2060. Unger et al. 2009 estimated that ozone linked to on-road vehicles is responsible for nearly a fourth of the total radiative forcing from the transportation sector (32). Hence, reducing ozone concentrations could help reduce local temperatures, in turn further reducing ozone formation.

The feedback effect from PM is more complex than ozone. Reduction of PM decreases light scattering which increases the yield of photochemical reactions, such as ozone formation. In addition, reduction of sulfate and nitrate aerosol would reduce the aerosol indirect effect that leads to atmospheric cooling. However, reduction of black carbon would reduce the atmospheric radiative forcing, which could compensate for the increased forcing from reducing sulfate and nitrate PM. Finally, changes in PM could impact precipitation thus affecting pollutant removal. However, the analysis presented in this study is based upon typical summer ozone and PM episode conditions under which effects on cloud condensation are not relevant.

3.5. General Observations and Conclusions. Projecting the air quality implications of hydrogen infrastructure in urban areas requires spatial and temporal characterization of the emissions fields followed by detailed atmospheric chemistry and transport computations. Results obtained in this study for the SoCAB establish that (1) a significant adoption of hydrogen infrastructure with HFCV in the year 2060 will substantially improve urban air quality in the SoCAB concomitant with a reduction in WTW GHG emissions from the passenger vehicle fleet, and (2) a renewable energy emphasis (Scenario HR) on hydrogen infrastructure deployment produces localized air quality benefits that surpass those of a hydrogen infrastructure scenario with more fossil fuel use (Scenario HF). The spatial and temporal emissions fields in both Scenario HR and HF lead to significant reductions in ozone and particulate matter in general, and especially where the poorest local air quality is experienced. This is important given that high correlations between long-term exposure to atmospheric aerosols and human health have been detected in population-based studies for several decades (33). A recent study by Pope et al. (2009) even quantifies the effects of aerosols on human lifespan. It is suggested, for example, that a decrease of 10 µg per cubic meter in the concentration of fine particulate corresponds to an increase in life expectancy of 0.61 years (34). In the SoCAB, ozone and particulate matter are the major concern for compliance with federal and state standards, and therefore of greatest concern to human health. Both scenarios reduce GHG emissions associated with passenger vehicles by more than 59% with just a 75% penetration of HFCV. Overall, Scenario HR leads to a greater reduction in GHG than Scenario HF.

Findings suggest that, compared to projections of remarkably improved ICE and hybrid ICE vehicles, hydrogen infrastructure and HFCV deployment will substantially improve air quality in an urban airshed and reduce GHG emissions from passenger vehicles, even when fossil fuels are a significant source of hydrogen. While these results agree with previous hydrogen infrastructure studies in a general sense (e.g., GHG are reduced, air quality is improved) (3–8), they provide an unprecedented level of detail and insight from a planning perspective. The coupling of spatially and temporally resolved hydrogen scenarios with the UCI-CIT air quality model provides an understanding of how HFCV can effect localized pollution within an urban air basin as well as how these effects can change depending upon spatial allocation of hydrogen infrastructure and temporal distribution of emissions from the infrastructure. Furthermore, the capability to simulate integrated hydrogen infrastructure scenarios provides insight into the degree to which variations in a diverse hydrogen production and distribution portfolio may affect overall environmental benefits.

The methodology presented, the spatially and temporally resolved energy and environment tool (STREET), integrates the PCA methodology (3) with spatial and temporal infrastructure design and a robust air quality simulation model and thereby provides a capability to assess quantitatively the impacts (e.g., GHG, criteria pollutants, energy intensity,
water resources, costs, urban air quality) for future year scenarios proposed for fuels and power plants associated with both mobile and stationary sources. The utility of STREET as a planning tool is herein demonstrated through application to future year hydrogen infrastructure scenarios with high temporal and spatial resolution. Today, business decisions are being made by industry involved in mobile sources (e.g., automobile, truck, locomotive, shipping, aircraft), industry involved in fuels (e.g., gasoline, diesel, natural gas, oil, Jet-A and JP-8, hydrogen, biofuels), and industry involved in electric power generation (from manufacturing to utilities to the burgeoning distributed generation) predicated on a paradigm change to lower carbon, enhanced air quality, and a protection of water resources. STREET allows for business scenarios to be vetted, modified, and refined to meet these goals. In parallel, the political process at both the Federal and state levels is invoking policy to address this future, and empowering governmental agencies with the responsibility to implement, monitor, enforce, and regulate. STREET allows the impacts associated with proposed policy to be assessed quantitatively for future year scenarios prior to enactment.

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Supporting Information Available

Details regarding the development of spatial and temporal resolution for hydrogen infrastructure scenarios are provided. Emissions factors for hydrogen technologies operating within the urban airshed are provided with references and the standard deviation is calculated to account for variations in values from different literature sources. Also provided is a summary of the total emissions and change in emissions associated with each scenario and finally, the speciation of emissions assumed for steam methane reforming, petroleum refining, heavy duty trucks, and passenger automobiles. This material is available free of charge via the Internet at http://pubs.acs.org.

Literature Cited

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