Impact of a future H$_2$-based road transportation sector on the composition and chemistry of the atmosphere – Part 1: Tropospheric composition and air quality

D. Wang$^1$, W. Jia$^1$, S. C. Olsen$^1$, D. J. Wuebbles$^1$, M. K. Dubey$^2$, and A. A. Rockett$^3$

$^1$Department of Atmospheric Sciences, University of Illinois at Urbana-Champaign, Urbana, IL, USA
$^2$Earth Systems Observations, Los Alamos National Lab, Los Alamos, NM, USA
$^3$Department of Materials Science and Engineering, University of Illinois at Urbana-Champaign, Urbana, IL, USA

Correspondence to: D. J. Wuebbles (wuebbles@atmos.uiuc.edu)

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Abstract. Vehicles burning fossil fuel emit a number of substances that change the composition and chemistry of the atmosphere, and contribute to global air and water pollution and climate change. For example, nitrogen oxides and volatile organic compounds (VOCs) emitted as byproducts of fossil fuel combustion are key precursors to ground-level ozone and aerosol formation. In addition, on-road vehicles are major CO$_2$ emitters. In order to tackle these problems, molecular hydrogen (H$_2$) has been proposed as an energy carrier to substitute for fossil fuels in the future. However, before implementing any such strategy it is crucial to evaluate its potential impacts on air quality and climate. Here, we evaluate the impact of a future (2050) H$_2$-based road transportation sector on tropospheric chemistry and air quality for several possible growth and technology adoption scenarios. The growth scenarios are based on the high and low emissions Intergovernmental Panel on Climate Change Special Report on Emissions Scenarios, A1FI and B1, respectively. The technological adoption scenarios include H$_2$ fuel cell and H$_2$ internal combustion engine options. The impacts are evaluated with the Community Atmospheric Model Chemistry global chemistry transport model (CAM-Chem). Higher resolution simulations focusing on the contiguous United States are also carried out with the Community Multiscale Air Quality Modeling System (CMAQ) regional chemistry transport model. For all scenarios future air quality improves with the adoption of a H$_2$-based road transportation sector; however, the magnitude and type of improvement depend on the scenario. Model results show that the adoption of H$_2$ fuel cells would decrease tropospheric burdens of ozone (7 %), CO (14 %), NO$_x$ (16 %), soot (17 %), sulfate aerosol (4 %), and ammonium nitrate aerosol (12 %) in the A1FI scenario, and would decrease those of ozone (5 %), CO (4 %), NO$_x$ (11 %), soot (7 %), sulfate aerosol (4 %), and ammonium nitrate aerosol (9 %) in the B1 scenario. The adoption of H$_2$ internal combustion engines would decrease tropospheric burdens of ozone (1 %), CO (18 %), soot (17 %), and sulfate aerosol (3 %) in the A1FI scenario, and would decrease those of ozone (1 %), CO (7 %), soot (7 %), and sulfate aerosol (3 %) in the B1 scenario. In the future, people residing in the contiguous United States could expect to experience significantly fewer days of elevated levels of pollution if a H$_2$ fuel cell road transportation sector were to be adopted. Health benefits of transitioning to a H$_2$ economy for citizens in developing nations, like China and India, will be much more dramatic, particularly in megacities with severe, intensifying air-quality problems.

1 Introduction

Fossil fuel combustion in on-road vehicles emits a number of pollutants directly into the surrounding air. The exhaust air contains nitrogen oxides (NO$_x$ = NO + NO$_2$), carbon monoxide (CO), volatile organic compounds (VOCs), and particulate matter. These byproducts can have adverse impacts on human health. For example, sustained high levels of NO$_2$ have an adverse impact on the respiratory system.
as elevated CO concentrations degrade the ability of blood to transport oxygen (e.g., WHO/Europe, 2003). In addition, NOx, CO and VOCs are precursors to the formation of ground-level ozone (O3), which adversely impacts human health and ecosystems in a number of ways. Particulate matter, which raises concern of physiological hazard, is either directly released in the form of soot (black carbon) or formed indirectly from gaseous pollutants through a series of photochemical and physical processes (sulfate and secondary organics). In the United States, on-road vehicles rank first in NOx and CO emissions and second in VOCs emissions among all source sectors (US EPA, 2009). In the future the on-road vehicle fleet is expected to become an increasingly more important contributor to ambient air pollution.

Molecular hydrogen (H2) has been proposed as an alternative energy carrier for on-road vehicles since it has higher energy efficiency and much cleaner oxidation products. Since H2 fuel cells emit only water vapor (H2O), tailpipe emissions of NOx, CO, VOCs, and soot associated with fossil fuel combustion are eliminated. In addition to being oxidized in fuel cells, H2 can also be burned in internal combustion engines. In this paper, the impact of both the H2 fuel cell and internal combustion engine options will be investigated. Vehicles powered by H2 internal combustion engines emit no CO, VOCs, or soot but do emit NOx. Additionally, in each of these scenarios some H2 is emitted due to leakage during its production, distribution, and storage. Given the reductions in tailpipe pollutant emissions, it is reasonably foreseeable that transitioning the world’s road traffic from fossil fuel powered vehicles to H2 powered vehicles could substantially improve air quality and climate, provided that the H2 is produced by non-polluting methods, e.g., wind, solar, nuclear power, hydroelectricity or, promisingly, photocatalytic and photoelectrochemical dissociation of water (Abe, 2010) and microbial reverse-electrodialysis electrolysis cells (Kim and Logan, 2011). Even if H2 is generated conventionally from fossil fuel, it would be easier to control the pollutant emissions from H2 production since they would be emitted from relatively few locations and the produced CO2 could be sequestered. This paper aims at quantitative evaluation of the potential environmental impacts of such a transition.

Tropospheric H2 concentrations are currently around 530 ppbv (Novelli et al., 1999), making it second to CH4 as the most abundant oxidizable gas in the atmosphere (Constant et al., 2009). The primary sources of H2 are fossil fuel and biofuel combustion, biomass burning, ocean emissions, and photolysis of formaldehyde. Bottom-up inventories estimate its source as ∼75 Tg H2 yr−1 (Ehhalt and Rohrer, 2009). Its largest sink (∼80 %) is microbial uptake in soils, with oxidation by OH in the atmosphere making up the remainder of its loss (Constant et al., 2009). Its uptake by microbes in soil, despite its importance, is not well understood and is subject to large uncertainties. Observations show a correlation between the H2 and CO deposition velocities (Conrad and Seiler, 1985; Liebl and Seiler, 1976; Yonemura et al., 1999, 2000). Recently there have been attempts to examine microbial H2 uptake by different soil types under a variety of conditions (e.g., Smith-Downey et al., 2008; Constant et al., 2008). It is known that the H2 uptake depends on soil moisture and temperature, among a number of factors. However, there still remains relatively large uncertainty in variation among ecosystems, the controlling factors, and how to map these to global ecosystems.

H2’s total atmospheric lifetime is estimated to be 1 to 2 yr (Constant et al., 2009; Ehhalt and Rohrer, 2009; Novelli et al., 1999; Price et al., 2007) and its lifetime with respect to OH loss is ∼8 yr (Ehhalt and Rohrer, 2009). The latitudinal gradient of H2 concentrations is unique from many other trace gases, with ∼3 % higher average concentrations in the Southern Hemisphere (SH) than in the Northern Hemisphere (NH). This is thought to be due to the greater soil microbial uptake over land in the NH (Ehhalt and Rohrer, 2009). Since there are no clear long-term trends (Constant et al., 2009; Ehhalt and Rohrer, 2009), a conversion to a H2-based road transportation sector could elevate atmospheric H2 concentrations to values not previously experienced.

Initial studies on the environmental impacts of a H2 economy have focused mainly on the possible adverse effects of increased H2 on stratospheric ozone. Using a two-dimensional model, Tromp et al. (2003) found that in an assumed world with a H2-based energy source with a perhaps unrealistically large H2 leakage rate (∼12 %), the surface H2 concentration of 2.3 ppmv would lead to 3–8 % polar spring stratospheric ozone loss due to a decrease in lower stratosphere temperature. Later studies found a much smaller impact (less than 1 %) on stratospheric ozone (Warwick et al., 2004), and instead focused on the relatively large improvements in tropospheric air quality due to the reduction in ozone precursor emissions.

A 3-D modeling study by Schultz et al. (2003) suggested a H2 fuel cell road traffic fleet in ca. 2000 would reduce human emissions of NOx and CO by half, resulting in 3 %, 6 %, 5 % and 29 % decrease in tropospheric CO, OH, O3 and NOx, respectively; tropospheric H2 burden would increase by 28 %, assuming a 3 % leakage rate. Warwick et al. (2004) reported an estimated 2.2 % decrease in tropospheric O3 in a H2 economy based on a 2-D modeling study. Jacobson et al. (2005) compared the environmental impact of a 1999 hydrogen on-road vehicle fleet in the US by means of steam reforming of natural gas, wind electrolysis, and coal gasification; they found the wind and natural gas options to have the best desired environmental improvement.

Although H2 is not a direct greenhouse gas and its utilization could provide opportunities to offset CO2 emissions, concern has arisen due to its indirect greenhouse potential (e.g., Prather, 2003). Through its reaction with OH, increases in H2 concentration could decrease the oxidizing capacity of the troposphere and hence increase the CH4 lifetime. CH4 is an ozone precursor and a long-lived greenhouse gas that is 25 times more powerful than CO2 in terms of integrated...
radiative forcing over a 100 yr time horizon. Currently its radiative forcing is second only to that of CO₂ (IPCC, 2007). Derwent et al. (2006) estimated that the climate impact of 1 % leakage of H₂ is 0.6 % of that of the replaced CO₂.

The impact of the reductions in ozone precursor emissions may be significant on the oxidizing capacity of the troposphere as well. Taking this effect into account, the decrease in tropospheric OH abundance ranges from 0 % to 12 %, and the increase in CH₄ lifetime ranges from 0 % to 26 %, depending on the specific scenario assumed (Jacobson, 2008; Schultz et al., 2003; Warwick et al., 2004).

All of these previous studies were based on the then current (ca. 2000) background atmosphere, focused on a conversion of the then current infrastructure, and assumed an immediate transition to hydrogen technology. The transition to hydrogen-based energy delivery will, in real-world practice, require the development of a massive industrial H₂ production capacity and substantial changes to the energy delivery infrastructure, and there are still a number of technological barriers yet to overcome. Considering the time needed to make the current infrastructure H₂-compatible, we evaluate the impacts of a transition occurring in the future that allows a reasonable amount of time for the infrastructure changes to occur. Thus, we assume that in the coming decades road transportation will transform to H₂ powered such that the transition will be complete by 2050. That is, all on-road vehicles operating in 2050 will be powered by H₂.

In this study we assess the impact on the composition and chemistry of the troposphere in 2050 due to 100 % conversion of the road transportation system from fossil fuel to H₂. To accomplish this we develop 2050 emissions scenarios for continued use of fossil fuel and for a H₂-based transportation sector. The emissions scenarios encompass several possible growth and technology adoption paths. The growth scenarios are based on the high and low emitting Intergovernmental Panel on Climate Change (IPCC) Special Report on Emissions Scenarios (SRES), A1FI and B1, respectively. The technological adoption scenarios include H₂ fuel cell and H₂ internal combustion engine options. The impacts of these emissions are evaluated using the Community Atmosphere Model with Chemistry (CAM-Chem) three-dimensional global climate-chemistry model. In addition, we evaluate the air quality impact over the contiguous United States using the Community Multiscale Air Quality Modeling System (CMAQ) regional air quality model. The development of the emissions scenarios is discussed in Sect. 2, the models are described in Sect. 3, and the results are presented in Sect. 4.

2 Emission scenarios

Emissions of a given species are often estimated based on emissions factors, which relate the emissions of a given pollutant to an activity and to the level of the activity. Thus, projections of future emissions are based on projections of the change in the activity level and the changes in the emissions factors. Commonly, the activity factors include population, energy usage, and GDP. Since there are large uncertainties in projecting the change in each of these activities over time, there are also relatively large uncertainties in the emissions estimates based on them. To bracket the possible evolution pathways of the future emissions, we develop scenarios based on the highest (A1FI) and lowest (B1) IPCC SRES emissions scenarios (IPCC, 2000). In the A1FI scenario, the world is assumed to evolve with rapid economic growth and rely intensively on fossil fuel; in the B1 scenario, the world’s economic structures are projected to become more service and information intensive. For each of these two IPCC growth scenarios (A1FI and B1), three emission scenarios with different technologies were developed. (1) In this future baseline (BL) scenario, the reference scenario for 2050, fossil fuel powered vehicles remain the dominant mode of road transportation. (2) In the H₂ fuel cell (H₂-FC) scenario, energy demands of the road transportation sector are met by H₂ fuel cell technology. In H₂-FC scenario, H₂ emissions due to leakage as well as reductions in combustion-related emissions of H₂, NOₓ, VOCs, CO, SO₂ and soot are included. (3) In the H₂ internal combustion engine H₂-ICE scenario, energy demands of the road transportation sector are met by H₂ using internal combustion technology. In H₂-ICE scenario, H₂ emissions due to leakage as well as reductions in combustion-related emissions of H₂, VOCs, CO, SO₂ and soot are included, as in H₂-FC scenario; however, unlike H₂-FC scenario, there are no reductions in NOₓ emissions because NOₓ is still a byproduct of internal combustion process regardless of the type of fuel. For each IPCC scenario, an additional scenario, baseline + H₂ (BL + H₂), was developed. In this additional scenario, H₂ emissions leakage into the atmosphere was included but all other emissions remain as described above. This H₂ emissions leakage scenario, though not realistic, is designed as a sensitivity study in order to evaluate and compare the impact of H₂ emission alone versus the impact of the reduction in ozone precursor emissions on tropospheric composition and chemistry.

The future baseline (BL) emissions are calculated by scaling the current fossil fuel combustion emissions by regionally dependent growth scale factors from the applicable IPCC scenario. The included species are NOₓ, CO, SO₂ and several NMVOCs (non-methane VOCs), namely, CH₂O, CH₃OH, CH₂CHO, C₂H₄, C₂H₆, C₂H₂OH, C₃H₆, C₃H₈, CH₃COCH₃ (acetone), C₄H₈O (methyl ethyl ketone), C₄H₁₀, C₅H₈ (isoprene), C₁₀H₁₆, BIGALK (lumped species for alkanes with four or more carbon atoms), BIGENE (lumped species for aromatics) and TOLUENE (lumped species for aromatic compounds). Emissions of these species are prepared from the Precursors of Ozone and their Effects in the Troposphere (POET) emissions dataset, which is based on version 3 of the Emission.
For the global model simulations in this study we use the Community Atmosphere Model with Chemistry (CAM-Chem) three-dimensional climate-chemistry model (Lamarque et al., 2005; Pfister et al., 2008). CAM-Chem’s chemistry module is based on the chemical component of the Model for OZone And Related chemical Tracers (MOZART). MOZART was described and extensively evaluated by Horowitz et al. (2003), and has been employed in a number of studies (e.g., Huang et al., 2008; Lin et al., 2008a, b; Tie et al., 2005). It contains a comprehensive tropospheric gas phase and aerosol chemical mechanism and includes 119 species and 300 reactions. CAM-Chem uses a bulk aerosol method and includes organic carbon, black carbon, sulfate, and ammonium nitrate aerosols. Based on the
observed correlation between H\textsubscript{2} and CO deposition velocities (Conrad and Seiler, 1985; Liebl and Seiler, 1976; Yonemura et al., 1999, 2000), we adopt a H\textsubscript{2} deposition velocity equal to twice the CO deposition velocity.

For this study, the global atmosphere is divided into grids with a horizontal resolution of 1.9\degree latitude by 2.5\degree longitude, and 26 vertical layers extending from the surface to 3-millibar level (∼40 km altitude). The meteorology is prescribed by climate model output data from Community Climate System Model (CCSM), which corresponds to the climate state of the mid-1990s. We choose to use this meteorology since our focus in this study is on air quality and photochemistry-influenced impacts due to changes in emissions. Previous studies (e.g., Lin et al., 2008a; Wu et al., 2008) have shown that future climate changes will likely have less effect on air quality than the future changes in emissions. The model is integrated with a time step of 30 minute sub-time step. Detailed descriptions of the mechanisms, algorithms, and implementation are available in the CMAQ scientific documentation (Byun and Schere, 2006). Boundary conditions for the CMAQ model simulations are taken from the corresponding global tropospheric model simulations. The CMAQ simulations are performed for the BL, H\textsubscript{2}-FC, and H\textsubscript{2}-ICE scenarios for each IPCC scenario.

### 4 Model results and discussion

Overall, H\textsubscript{2} concentrations increase substantially in all H\textsubscript{2} scenarios and air quality improves with the adoption of a H\textsubscript{2}-based road transportation sector; however, the magnitude and quality of improvement depends on the scenario. While the changes in H\textsubscript{2} concentrations are sensitive to uncertainties in the assumed H\textsubscript{2} leakage rate of 2.5 %, the changes in air quality, e.g., O\textsubscript{3}, are driven by changes in ozone precursor emissions that are not related to the assumed H\textsubscript{2} leakage rate. Each relevant species is discussed in detail below.
4.1 Molecular hydrogen

In all of the scenarios examined in this study, average tropospheric H$_2$ concentrations are greater in 2050 than they are in 2000. In the A1FI baseline scenario the annual average global mean concentration is 839 ppbv (Table 1). The H$_2$ lifetime in the A1FI 2050 baseline scenario is essentially unchanged from its value of 2.5 yr in the year 2000 simulated atmosphere. In this scenario the NH emissions are strong enough to offset the stronger soil sink in this hemisphere, leading to a reverse in the inter-hemispheric gradient, i.e., higher NH mean H$_2$ concentration (~850 ppbv) than SH (~830 ppbv). Annual-mean ground-level H$_2$ mixing ratios range from 720 ppbv to more than 1 ppmv (Fig. 2a). Higher concentrations are located in highly urbanized and/or industrialized regions such as northeastern United States, California, Western Europe, Korea, Japan and eastern China, while lower concentrations appear over mid-latitude continents of the SH. The maximum/minimum pattern reflects the forcing of the H$_2$ emissions from human activities and the large H$_2$ microbial soil sink.

In the A1FI H$_2$ technology scenarios (BL+H$_2$, H$_2$-FC, and H$_2$-ICE), global mean H$_2$ concentrations increase substantially from the baseline scenario by ~40%, up to ~1170 ppbv (Table 1). The H$_2$ lifetime in the H$_2$-FC scenario is the same as in the baseline simulation (2.5 yr) but decreases slightly (by ~3%) in the H$_2$-ICE scenario. This increase is because the H$_2$ leakage emissions are much greater than the replaced fossil fuel H$_2$ emissions in the baseline scenario. In the H$_2$-FC scenario the south to north H$_2$ inter-hemispheric gradient is even larger than in the BL scenario (Fig. 2b) since H$_2$ emissions increased more in the NH than in the SH due to the asymmetric intensity of road transportation activities between the hemispheres. The background surface H$_2$ mixing ratio increases by at least 40% in the NH and by at least 35% in the SH, reaching about 1.2 ppmv in the NH and 1.15 ppmv in the SH. The highest H$_2$ concentrations (greater than 1.6 ppmv) are in eastern China and Korea, corresponding to a greater than 75% increase from the baseline scenario. H$_2$ concentrations increase by approximately 50% in northeastern United States. The largest increase (~45%) in zonal-average H$_2$ concentration occurs in the boundary layer in the Northern Hemisphere mid-latitudes where road transportation activities are concentrated. The spatial distributions of the H$_2$ concentrations in the BL+H$_2$ and H$_2$-ICE scenarios (not shown) are nearly identical to those in the H$_2$-FC scenario. The geographical pattern of relative increase in surface H$_2$ concentrations in this scenario is similar to that calculated by Schultz et al. (2003), even though they assumed a leakage rate of 3% and their calculation is for the year 2000 atmosphere. However, the increase in eastern Asia is comparatively larger in this study because of the projected significant economic development in this region by 2050.

In the B1 baseline scenario the average tropospheric H$_2$ mixing ratio is 621 ppbv (Table 2), and the annual-mean ground-level H$_2$ concentration is ~600 ppbv (Fig. 2c). The H$_2$ lifetime is 2.5 yr, unchanged from the current day and A1FI baseline simulations. The inter-hemispheric gradient is of the same direction as in the current atmosphere, with higher background H$_2$ concentrations in the SH. Since human activities that emit H$_2$ are not as intensive as in the A1FI scenario, there are few regions bearing significantly higher than background value, except the northeastern United States, where H$_2$ concentrations are around 650 ppbv.

In the B1 H$_2$ scenarios (BL + H$_2$, H$_2$-FC, and H$_2$-ICE), the tropospheric H$_2$ burden increases by ~20%, corresponding to a mean tropospheric concentration of 740 ppbv. As for the A1FI simulations, the H$_2$ lifetime for the B1 H$_2$-FC simulations is unchanged at 2.5 yr while for the H$_2$-ICE scenario it decreases by ~3%. In the H$_2$-FC scenario (the other two are quite similar), surface H$_2$ concentrations in remote regions increase from the BL scenario by more than 20% in the NH and by more than 15% in the SH (Fig. 2d), leading to a slightly higher H$_2$ concentration in the NH, a reverse in the inter-hemispheric gradient. Maximum H$_2$ mixing ratios (860 ppbv) occur in eastern Asia, corresponding to a more than 40% increase from the baseline scenario.
Fig. 2. CAM-Chem simulated 2050 annual-mean ground-level H\textsubscript{2} mixing ratios in the A1FI (a) and B1 (c) Baseline scenarios and the percent differences for the A1FI (b) and B1 (d) H\textsubscript{2}-FC scenarios. Note the different scales.

Table 2. Tropospheric mean mixing ratios or concentrations for the 2050 B1 baseline scenario and percent changes for the baseline + H\textsubscript{2} (BL+H\textsubscript{2}), hydrogen fuel cell (H\textsubscript{2}-FC), and hydrogen internal combustion engine (H\textsubscript{2}-ICE) scenarios from the CAM-Chem model simulations.

<table>
<thead>
<tr>
<th></th>
<th>Baseline</th>
<th>BL+H\textsubscript{2}</th>
<th>H\textsubscript{2}-FC</th>
<th>H\textsubscript{2}-ICE</th>
</tr>
</thead>
<tbody>
<tr>
<td>H\textsubscript{2}</td>
<td>621 ppbv</td>
<td>20 %</td>
<td>20 %</td>
<td>19 %</td>
</tr>
<tr>
<td>O\textsubscript{3}</td>
<td>37 ppbv</td>
<td>0 %</td>
<td>-5 %</td>
<td>-1 %</td>
</tr>
<tr>
<td>OH</td>
<td>10.1 \texttimes 10\textsuperscript{5} molecules cm\textsuperscript{-3}</td>
<td>-1 %</td>
<td>-4 %</td>
<td>3 %</td>
</tr>
<tr>
<td>CO</td>
<td>90 ppbv</td>
<td>0 %</td>
<td>-4 %</td>
<td>-7 %</td>
</tr>
<tr>
<td>NO\textsubscript{x}</td>
<td>60 pptv</td>
<td>0 %</td>
<td>-11 %</td>
<td>3 %</td>
</tr>
<tr>
<td>soot</td>
<td>0.014 µgC m\textsuperscript{-3}</td>
<td>0 %</td>
<td>-7 %</td>
<td>-7 %</td>
</tr>
<tr>
<td>SO\textsubscript{4}</td>
<td>0.33 µg SO\textsubscript{4} m\textsuperscript{-3}</td>
<td>0 %</td>
<td>-4 %</td>
<td>-3 %</td>
</tr>
<tr>
<td>NH\textsubbox{4}NO\textsubbox{3}</td>
<td>0.010 µg m\textsuperscript{-3}</td>
<td>0 %</td>
<td>-9 %</td>
<td>2 %</td>
</tr>
</tbody>
</table>

4.2 Ozone

The annual-averaged tropospheric ozone (averaged over the entire troposphere) concentration is 44 ppbv in the 2050 A1FI BL scenario, higher than the 2000 value of 39 ppbv (IPCC, 2001), due to increased precursor emissions. The summertime (June, July, and August) average surface ozone concentrations over land in the A1FI BL scenario are projected to be mostly above 40 ppbv (Fig. 3a). Summertime ozone concentrations are highest near densely populated areas, such as the eastern United States, California, the Middle East, and eastern China. In the A1FI H\textsubscript{2}-FC scenario, surface ozone mixing ratios decrease by more than 5 % around the world, with as much as 20 % decrease in Latin America and Southeast Asia (Fig. 3b). In heavily polluted areas surface ozone mixing ratios are about 10 % lower than those in the BL scenario. However, ozone increases in some localized populated areas in eastern China, Korea, South Africa, and London, UK. In these regions ozone production is VOC-limited. The VOC-limited regime is characterized by high background NO\textsubscript{x} concentrations with ozone production proportional to VOC concentrations but inversely proportional to NO\textsubscript{x} concentrations (e.g., Jacob, 1999). In this case the increase in O\textsubscript{3} production due to the decrease in NO\textsubscript{x} emissions wins over the decrease in O\textsubscript{3} production due to the decrease in VOC emissions causing an increase in O\textsubscript{3} in these regions for the H\textsubscript{2} fuel cell scenario. While the changes in ozone extend throughout the troposphere, the largest changes occur near the surface (Fig. 4). In the A1FI H\textsubscript{2}-FC scenario, zonally-averaged summertime ozone mixing ratios are at least 5 % lower than in the BL scenario in the lower troposphere (Fig. 4b). The maximum decrease (more than 10 %) appears in the boundary layer at the low latitudes of the NH (centered at around 15\degree N).

In the A1FI H\textsubscript{2}-ICE scenario, summertime surface ozone concentrations are virtually unchanged over most of the globe (Fig. 3c). The relatively modest decreases occur over limited regions, such as England and Korea, which tend to include the highly localized regions where ozone increased in the H\textsubscript{2} fuel cells scenario. The zonally-averaged ozone concentrations do not change significantly except in the lower troposphere of the northern middle latitudes, where ozone decreases by as much as 5 % (Fig. 4c).

In the 2050 B1 BL scenario, the average tropospheric ozone concentration is \sim 37 ppbv, slightly lower than the current value, due to slightly decreased precursor emissions
(except NOx, which slightly increases). The distribution pattern of the summertime surface ozone concentrations over the world is similar to that for the corresponding A1FI scenarios, but the concentrations are 10–20 ppbv lower than those in the A1FI BL scenario (Figs. 3d and 4d). In the B1 H2-FC scenario, summertime near surface ozone decreases significantly (by 10 % to 30 %) (Figs. 3e and 4e) along the coasts of the United State, making them considerably lower than current O3 concentrations. Ozone reductions in the B1 H2-ICE scenario are not as apparent (Figs. 3f and 4f), except in the same sensitive areas as in the A1FI H2-ICE scenario, where relative reductions are no more than 15 %.

The simulation results of the CMAQ model over the contiguous United States is consistent with the global model results in general; however, due to its finer resolution, CMAQ is capable of capturing some of the finer features of the ozone concentrations. For example, while CAM-Chem predicts that the highest summertime ozone concentrations appear in California and the Midwest, CMAQ predicts higher maximum daily 8 h average ozone concentrations near population centers and along the US west and east coasts with a plume extending off the east coast (Fig. 5a and d). Over the southwestern United States and parts of the East Coast, daily maximum 8 h average O3 concentrations at surface are projected to exceed the US EPA National Ambient Air Quality Standards (NAAQS) of 75 ppbv over much of the summertime for the A1FI BL scenario. In contrast, concentrations are in compliance with the NAAQS (below ∼ 56 ppbv) for the vast majority of the time for the B1 BL scenario.

Relatively large decreases in daily maximum 8 h O3 concentrations are noted with the adoption of the H2-FC scenarios. For both the A1FI and B1 scenarios the decreases are largest over the population centers (Fig. 5b and e). The decreases in summertime O3 concentrations are nearly 18 ppbv for A1FI and 12 ppbv for the B1 scenario. In stark contrast to these decreases for the H2-FC scenario, there is little decrease in daily maximum 8 h O3 concentrations for the H2-ICE scenario over the vast majority of the United States (Fig. 5e and f). There are some relatively small reductions over the Los Angeles and San Francisco regions of California and a few very localized spots in the Midwest but these...
Fig. 4. CAM-Chem simulated 2050 NH summer (June, July, and August) average zonal-mean O$_3$ mixing ratios in the A1FI (a) and B1 (d) baseline scenarios and percent differences for the H$_2$ fuel cell (H$_2$-FC) and H$_2$ internal combustion engine (H$_2$-ICE) scenarios (b, c, e, f). Note the different scales.

Fig. 5. July 2050 averages of the daily maximum 8 h-average O$_3$ concentrations simulated by CMAQ for the A1FI (top) and B1 scenarios (bottom). Each row contains the baseline scenarios (left: a and d), differences between the baseline and H$_2$ fuel cell (H$_2$-FC) scenarios (center: b and e), and differences between the baseline and H$_2$ internal combustion engine (H$_2$-ICE) scenarios (right: c and f). Data are averaged over the lowest model level (∼100 m). Note that the scales are different among panels.
reductions are much less significant than in the H$_2$-FC scenario.

In the A1FI H$_2$-FC scenario, the population of the United States would experience considerably fewer days above the 75 ppb limit than in the baseline scenario (Fig. 6). Here, we perform an analysis of time cumulated population exposure to levels of ambient ozone using population data from the 2000 US census (http://www.esri.com/data/download/census2000-tigerline/index.html) and assuming the population distribution does not change significantly by 2050. There is a large shift towards lower O$_3$ concentrations for the H$_2$-FC scenarios for both the A1FI and B1 scenarios (Fig. 6). In the A1FI scenario much of the large tail area above 75 ppbv is reduced and compliance with the ozone air quality standard is much more frequent (Fig. 6). While the improvements for the H$_2$-FC scenarios are quite impressive, the H$_2$-ICE scenario provides almost no noticeable improvement. The distributions are extremely similar except for a small reduction in the > 75 ppbv tail. In the B1 scenarios (Fig. 6) the standard is rarely exceeded in any scenario, but still there are substantial improvements with the adoption of a fuel cell powered road sector.

The scenarios examined in this study suggest that summertime O$_3$ over the contiguous United States is predominantly NO$_x$ controlled. This highlights the importance of the utilization of non-combustion technologies in which NO$_x$ emissions are avoided, e.g., fuel cells, towards improving O$_3$ air quality in a H$_2$-based road transportation sector.

4.3 The tropospheric oxidizing capacity (OH) and climate implication

As discussed in Sect. 1, utilization of H$_2$ raises concern over its potential impact on the oxidizing capacity of the troposphere. Our model simulations show that adoption of a H$_2$-based road transportation sector can either decrease or increase the tropospheric OH abundance (e.g., see Spivakovsky et al., 2000 for discussion of the climatology of tropospheric OH), depending on the technology option selected.

The tropospheric mean OH abundance changes from the 2000 modeled value of $9.7 \times 10^5$ molecules cm$^{-3}$ to $9.1 \times 10^5$ molecules cm$^{-3}$ in the 2050 A1FI baseline scenario (Table 1), mainly a result of the projected increase in CO emissions and CH$_4$ concentration in this scenario despite that the modeled tropospheric O$_3$ concentration increases. In contrast, the tropospheric mean OH abundance increases to $10.1 \times 10^5$ molecules cm$^{-3}$ in the B1 baseline scenario (Table 2) due to the projected decreased CO emissions in this scenario.

The sensitivity study of the BL + H$_2$ scenarios suggests that H$_2$ leakage emission itself from a H$_2$-based road transportation sector can have some impact on tropospheric OH abundance, assuming a leakage rate of 2.5%. The mean tropospheric OH abundance decreases by 2% and 1% in the A1FI and B1 scenarios, respectively, from the baseline scenarios (Tables 1 and 2). The reductions in OH abundance mainly occur in the tropical troposphere (Fig. 7a and d).

The combined effect of H$_2$ leakage emission and reductions in ozone precursor emissions in the A1FI and B1 H$_2$-FC scenarios is a 4% decrease in the mean tropospheric OH abundance (Tables 1 and 2). The largest OH abundance reduction is in the tropical boundary layer and extends to the middle troposphere (Fig. 7b and e). However, the OH abundance increases in the remote troposphere at high latitudes in the NH as a result of its decreased sink against CO, as CO concentrations decrease there.

In the H$_2$-ICE scenarios the tropospheric OH abundance would actually increase by 7% for the A1FI scenario and by 3% for the B1 scenario (Tables 1 and 2) despite the increased atmospheric H$_2$ concentrations, which would reduce OH abundance by 2%. This is likely because O$_3$ initiated OH production is not severely affected in these scenarios, while the OH sink through reactions with CO and other fossil fuel combustion byproducts is significantly reduced. The
tropospheric OH abundance increases by 10 % and by 3 % in the NH and in the SH, respectively, in the A1FI H\textsubscript{2}-ICE scenario (Fig. 7c), whereas it increases by 5 % and by 1 % in the NH and in the SH, respectively, in the B1 H\textsubscript{2}-ICE scenario (Fig. 7f). Evidently the effect of the H\textsubscript{2} economy on global OH is relatively small for the scenarios examined here and can be positive or negative with relatively minor contributions to atmospheric chemistry and climate forcing, as discussed next.

Impact on CH\textsubscript{4} lifetime and climate

Since the reaction with OH is the primary sink of CH\textsubscript{4}, changes in the OH abundance have direct impact on the atmospheric CH\textsubscript{4} lifetime. Here, the CH\textsubscript{4} lifetime was calculated by dividing the CH\textsubscript{4} tropospheric burden by its sink against OH. A companion run of the current atmosphere estimates CH\textsubscript{4} lifetime to be 8.7 yr, well within the 8~10 yr range summarized by IPCC (2007). The baseline CH\textsubscript{4} lifetime is 9.1 yr for the A1FI scenario and 8.3 yr for the B1 scenario. In the 2050 H\textsubscript{2}-FC scenarios, the CH\textsubscript{4} lifetimes increased by 7 % to 9.7 yr for the A1FI scenario and by 6 % to 8.8 yr in the B1 scenario. However, in the H\textsubscript{2}-ICE scenarios, the CH\textsubscript{4} lifetime decreased by 4 % to 8.7 yr in the A1FI scenario and by 2 % to 8.1 yr in the B1 scenario. These changes imply that transitioning to a H\textsubscript{2} fuel cell type road transportation sector may exacerbate climate warming due to CH\textsubscript{4} through its increased lifetime. However, the amount of warming due to CH\textsubscript{4} will depend on its actual atmospheric concentrations, which will depend on many factors, including how any transition to a H\textsubscript{2} economy occurs, and also any changes in other, e.g., biogenic, CH\textsubscript{4} emissions that might occur (not evaluated here).

It is interesting to investigate the combined climate effects of a H\textsubscript{2}-based road transportation sector in terms of both changes in CH\textsubscript{4} lifetime and changes in O\textsubscript{3} concentration. In the H\textsubscript{2}-FC scenarios, the decrease in tropospheric ozone concentrations, which implies a cooling effect, is accompanied by an increase in CH\textsubscript{4} lifetime, which implies a warming effect. The two effects tend to offset each other, but the effect is inhomogeneous because tropospheric ozone is short-lived and is thus not uniform, whereas CH\textsubscript{4} is long-lived and uniform. The changes in aerosol loadings, especially for soot
and sulfate, would further complicate the combined climate effect problem of the H$_2$-FC scenarios.

Whereas in the H$_2$-ICE scenarios, both tropospheric ozone concentration and CH$_4$ lifetime are decreased, implicating a negative radiative forcing. Taking soot reductions enhances this cooling effect while reductions of sulfate aerosols would counteract it. Again, it is interesting to investigate the climate effect of the H$_2$-ICE scenarios because of its inhomogeneity.

### 4.4 NO$_x$

The tropospheric NO$_x$ burden depends on the type of H$_2$ technology option selected. In the H$_2$-FC scenarios tropospheric NO$_x$ concentrations are reduced significantly, by 16% in the A1FI scenario and by 11% in the B1 scenario (Tables 1 and 2). In the H$_2$-ICE scenarios; however, tropospheric NO$_x$ concentrations increase by 10% in the A1FI scenario and by 3% in the B1 scenario.

Regions with intense human activities, such as eastern China, India, northeastern United States, and Western Europe, have high NO$_x$ concentrations in the baseline scenarios (Fig. 8a and d). These regions would benefit substantially from NO$_x$ reductions in the A1FI H$_2$-FC scenario. Annual mean ground-level NO$_x$ concentrations decrease considerably along the coasts of North and South America, Western Europe, North Africa, the Middle East, New Zealand, and Japan (Fig. 8b), with percentage reductions as high as 80%. Although some remote areas have large relative changes, for example much of the remote Southern Hemisphere, the absolute changes are quite small there as the baseline NO$_x$ mixing ratio is rather low. The relative reductions are most pronounced in wintertime, when the boundary layer height is lower and ventilation is inhibited. In the B1 H$_2$-FC scenario, the reduction in NO$_x$ concentrations occurs in the same regions, but the percentages are about half of those in the A1FI scenario (Fig. 8c).

In the A1FI and B1 H$_2$-ICE scenarios the simulated surface NO$_x$ concentration change is not apparent in the NO$_x$ pollution areas (Fig. 8c and f), showing that the internal combustion engine type of H$_2$-based road transportation sector would not be as effective in ground-level NO$_x$ mitigation.

As in the global CAM-Chem simulations, the CMAQ model results show that higher NO$_x$ concentrations tend to appear in wintertime, so here we focus on the wintertime...
(February) changes. NO\textsubscript{x} peaks are concentrated in the northeastern United States and in locations in the west, e.g., Los Angeles and the San Francisco Bay Area of California (Fig. 9a and d). Wintertime concentrations are higher than summertime concentrations and concentrations are higher in the A1FI than B1 scenarios. There are large decreases in NO\textsubscript{x} concentrations with the adoption of a H\textsubscript{2} fuel cell transportation sector (Fig. 9b and e), up to \sim 13 ppbv (\sim 50 %) in winter over the northeast United States for the A1FI scenario. However, for the H\textsubscript{2}-ICE scenario there is almost no change in wintertime NO\textsubscript{x} concentrations evident (Fig. 9c and f).

4.5 CO

The tropospheric CO burdens decrease by 14 % and 18 % for the A1FI H\textsubscript{2}-FC and H\textsubscript{2}-ICE scenarios, respectively, and by 4 % and 7 % for the B1 H\textsubscript{2}-FC and H\textsubscript{2}-ICE scenarios, respectively (Tables 1 and 2).

In the A1FI baseline scenario, annual mean surface CO concentrations over North America and Eurasia exceed 200 ppbv (Fig. 10a). Higher values are present in the northeastern US, Central Europe and eastern China. There are also higher than background concentrations in the tropics due to biomass burning. In the B1 baseline scenario, annual mean surface CO concentrations over the NH continents are over 100 ppbv (Fig. 10b). Higher concentrations are seen in eastern China, northeastern US, and tropical regions.

In the A1FI H\textsubscript{2}-FC scenario, surface CO concentrations in remote areas of the NH decrease by at least 20 % (Fig. 10b). Relative CO concentration reductions in the H\textsubscript{2}-FC scenario are largest in hotspots of human activities, such as northeastern US, California, central and western Europe, eastern China, South Korea and Japan. Concentrations in New York City decrease by as much as 80 %. The CO mitigation is perennial but is most pronounced in winter. In the H\textsubscript{2}-ICE scenario the ground-level CO concentration reduction pattern is similar to that in the A1FI H\textsubscript{2}-FC scenario, but the magnitude of reduction is even larger (Fig. 10c) due to \sim 11 % higher OH concentrations in the A1FI H\textsubscript{2}-ICE scenario than the A1FI H\textsubscript{2}-FC scenario. This is a result of the increase in atmospheric oxidizing capacity in the H\textsubscript{2} internal combustion engine scenarios, which would lead to decreases in species that react with OH, e.g., CO and other VOCs.

In the B1 H\textsubscript{2}-FC and H\textsubscript{2}-ICE scenarios, the background CO concentrations in the NH decrease by 10–20 % relative to the baseline scenario (Fig. 10d–f). The largest relative reduction (up to 50 %) appears over northeastern United States. There is also a 30 % decrease in Western Europe. The relative and absolute CO decreases in B1 H\textsubscript{2} scenarios are smaller than those in the corresponding A1FI scenarios.

The regional US CMAQ simulations are consistent with the CAM-Chem global simulations for 2050. Baseline CO concentrations are generally highest over the US east coast in wintertime, with monthly average wintertime concentrations of greater than 400 ppbv and 150 ppbv over much of the eastern part of the United States in the A1FI and B1 scenarios, respectively (Fig. 11a and d). However, analysis shows that there is no violation of CO NAAQS, that is, 8 h average not to exceed 9 ppmv and 1 h average not to exceed 35 ppmv more than once per year, in any of the scenarios examined. In the H\textsubscript{2}-FC and H\textsubscript{2}-ICE scenarios there are large decreases in CO concentrations due to the decrease in CO emissions. The largest decreases occur over areas with the highest emissions in the baseline scenario and, in contrast...
Fig. 10. CAM-Chem simulated 2050 annual mean surface CO mixing ratios in the A1FI (a) and B1 (d) Baseline scenarios and percent differences for the H$_2$ fuel cell (H$_2$-FC) and H$_2$ internal combustion engine (H$_2$-ICE) scenarios (b, c, e, f). Note the different scales.

Fig. 11. February 2050 averages CO concentrations (ppbv) simulated by CMAQ for the A1FI (top) and B1 scenarios (bottom). Each row contains the BL scenarios (left: a and d), differences (ppbv) between the BL and H$_2$-FC scenarios (center: b and e), and differences (ppbv) between the BL and H$_2$-ICE scenarios (right: c and f). Data are from the lowest model level (~100 m). Note that the scales are different among panels.
to some other pollutants, there are only slight differences between the H2-FC and H2-ICE scenarios. Wintertime CO decreases are nearly 280 ppbv and 100 ppbv for the A1FI and B1 scenarios over polluted regions, corresponding to decreases over much of the US east coast of greater than 60% (Fig. 11b, c, e and f). Summertime decreases are nearly half as large as wintertime decreases (not shown) because the wintertime planetary boundary layer is shallower, there is less vigorous vertical mixing, and chemical loss is lower.

4.6 Aerosols

In the H2-FC scenarios, tropospheric aerosols, especially soot, sulfate aerosols and ammonium nitrate concentrations are significantly reduced as a result of emissions decreases. In the H2-ICE scenarios, tropospheric soot and sulfate aerosol concentrations decrease while ammonium nitrate aerosol concentrations increase due to the NOx concentration increase.

In CAM-Chem, PM$_{2.5}$ includes sulfate aerosol, nitrate aerosol, black carbon, organic carbon, secondary organic aerosol, and dust and sea salt aerosols with diameters no larger than 2.5µm. Since emission changes have little impact on dust and sea salt aerosols, we exclude these types of aerosols in the following discussion of PM$_{2.5}$.

Figure 12a and d show the CAM-Chem simulated annual mean surface PM$_{2.5}$ concentrations in the A1FI and B1 BL scenarios. In the A1FI BL scenario, eastern China is subject to high PM$_{2.5}$ concentrations of more than 100 µg m$^{-3}$. Surface PM$_{2.5}$ concentrations can be as high as 50 µg m$^{-3}$ in northern India and parts of Indonesia. In central Europe, near ground PM$_{2.5}$ concentrations reach 30 µg m$^{-3}$. In the eastern United States, surface PM$_{2.5}$ concentrations are 10–20 µg m$^{-3}$. Annual mean surface PM$_{2.5}$ concentrations in the B1 BL scenario (Fig. 12d) are generally lower than those in the A1FI BL scenario. In central Europe and eastern United States it is $\sim$10 µg m$^{-3}$ lower, whereas in eastern China it can be $\sim$50 µg m$^{-3}$ lower than in the A1FI BL scenario.

In the A1FI H2-FC scenario, annual mean surface PM$_{2.5}$ concentrations are significantly reduced in eastern China and northern India, by up to 10 µg m$^{-3}$ (Fig. 12b). In northeastern United States and Europe, PM$_{2.5}$ concentrations are reduced by 1–3 µg m$^{-3}$. In the A1FI H2-ICE scenario, surface PM$_{2.5}$
concentrations reductions in the northeastern United States, India and Europe are not as significant as in the A1FI H₂-FC scenario, whereas the reductions in eastern China is comparable to the A1FI H₂-FC scenario (Fig. 12c).

In the B1 H₂-FC scenario, there are only a few locations with annual mean surface PM₂.₅ concentrations reductions of more than 1 µg m⁻³ (Fig. 12e). The maximum reduction of ~5 µg m⁻³ appears in eastern China. In the B1 H₂-ICE scenario, surface PM₂.₅ concentrations reductions are even less significant than in the B1 H₂-FC scenario. Higher reductions are only seen in eastern China (Fig. 12f).

Again, the regional US CMAQ simulations are consistent with the global CAM-Chem simulations but with more localized peaks due to their higher resolution. In the A1FI and B1 baseline scenarios, the only location in the contiguous US exceeding the annual mean PM₂.₅ NAAQS (15 µg m⁻³) is around Buffalo, NY. In the H₂-FC and H₂-ICE scenarios in both the A1FI and B1 scenarios, annual mean PM₂.₅ concentration in Buffalo, NY, decreases but it is still above the NAAQS.

PM₂.₅ concentrations are typically higher in winter than in summer. For the baseline A1FI and B1 scenarios, highest PM₂.₅ concentrations over the contiguous United States appear generally along the Ohio River Valley and the East Coast (Fig. 13a and d). February average surface concentrations peak at roughly 9 µg m⁻³ for the A1FI scenario and around 8 µg m⁻³ for the B1 scenario. As with O₃, improvements in PM₂.₅ air quality depend on the technology adoption scenario. PM₂.₅ reductions are greatest with the H₂-FC scenarios’ NOₓ and VOC emissions reductions, while the reductions are much more modest with the VOC only emissions reductions of the H₂-ICE scenario. While the areas of largest absolute decrease correspond to the areas of highest initial concentrations, the areas of largest relative decrease are typically over areas with lower initial concentrations in the Midwest and west coast. For the H₂-FC scenario there are reductions of ~1.5 µg m⁻³ and ~1 µg m⁻³ for the A1FI and B1 scenarios in February over the regions of high concentration in the Midwest (Fig. 13b and e). For the H₂-ICE scenario, reductions are much more modest at around 1 and 0.5 µg m⁻³ for the A1FI and B1 scenarios, respectively, and more concentrated on the eastern seaboard (Fig. 13c and f). The smaller PM₂.₅ reduction in the H₂-ICE scenarios is primarily due to the impact of nitrate aerosol PM₂.₅ production from the NOₓ emissions, whereas in the H₂-FC scenario there are reductions in soot, sulfate, and nitrate aerosols as well as secondary organic aerosol formation from decreases in VOC emissions.

The changes in annual mean surface PM₁₀ concentrations (not shown) are the same as those for PM₂.₅, because the diameters of the aerosols reduced are less than 2.5 µm in the CAM-Chem simulation. The following sections discuss the aerosol components that are significantly reduced.

### 4.6.1 Soot

In the baseline scenarios, soot (black carbon) concentrations at the surface are rather low in remote regions (less than 1 µgC m⁻³) (Fig. 14a and d). It is nearly absent in the air of pristine regions (concentrations less than 0.1 µgC m⁻³). In the tropics, soot concentrations are higher due to biomass burning. Densely populated regions are plagued with soot pollution from intense fossil fuel combustion activities. These regions include eastern China, Europe, North America, Japan and India. The worst pollution occurs in eastern China, where annual mean soot concentration can be up to more than 10 µgC m⁻³ in the A1FI BL scenario.

The tropospheric soot burden would decrease by 17% and 7% in the A1FI and B1 scenarios, respectively (Tables 1 and 2), both in the H₂-FC and the H₂-ICE scenarios. Ground-level soot would be significantly reduced in most of the United States, Europe, and Japan (Fig. 14c). The A1FI scenario a H₂-based road transportation sector reduces surface soot concentrations by as much as 50% in the United States and Western Europe (Fig. 14b). Soot concentrations are also reduced by 30–40% in the North Atlantic Ocean region and other regions downwind of key polluted areas. There is also significant relative reduction (~20%) of soot in eastern China. The pattern of surface soot concentration reduction is almost identical in the A1FI H₂-ICE scenario to that in the A1FI H₂-FC scenario (Fig. 14c). In the B1 scenarios, soot reductions in the H₂-FC and H₂-ICE scenarios are 10–20% less than in the corresponding A1FI scenarios, but they are still significant reductions and bear a similar pattern (Fig. 14e and f).

Soot is a warming climate agent (IPCC, 2001), though there remain uncertainties in quantitative understandings. Reductions of tropospheric soot, as in with a H₂-based road transportation sector, would have a cooling effect on climate. This effect would be inhomogeneous since the reduction is not uniform.

### 4.6.2 Sulfate aerosols

Sulfate aerosols (SO₄) are associated with human activities (Fig. 15a and d). The highest annual mean surface concentrations (~10µgSO₄ m⁻³) are seen in eastern China, India, and Mexico in the A1FI BL scenario. In the B1 BL scenario, sulfate aerosol pollution in these densely populated areas is slightly less severe than in the A1FI BL scenario.

The tropospheric sulfate aerosol burdens decrease by 4% for both the A1FI and B1 H₂-FC scenarios (Tables 1 and 2). In the A1FI H₂-FC scenario, surface SO₄ mass concentrations decrease by approximately 5% around the world. Higher reductions of ~10% occur in southern California, Southeast Asia and New Zealand (Fig. 15b). The increase in Antarctica is not significant as the background sulfate aerosol concentration is extremely small there (less than 0.1 µg SO₄ m⁻³) (Fig. 15a and d).
Fig. 13. February 2050 average PM$_{2.5}$ concentrations ($\mu$g/m$^3$) simulated by CMAQ for the A1FI (top) and B1 scenarios (bottom). Each row contains the BL scenarios (left: a and d), differences ($\mu$g/m$^3$) between the BL and H$_2$-FC scenarios (center: b and e), and differences ($\mu$g/m$^3$) between the BL and H$_2$-ICE scenarios (right: c and f). Data are from the lowest model level (~100 m). Note that the scales are different among panels.

Fig. 14. CAM-Chem simulated 2050 annual mean surface soot concentrations in the A1FI (a) and B1 (d) Baseline scenarios and percent differences for the H$_2$ Fuel Cell (H$_2$-FC) and H$_2$ Internal Combustion Engine (H$_2$-ICE) scenarios (b, c, e, f). Note the different scales.
Figure 15. CAM-Chem simulated 2050 annual mean surface \( \text{SO}_4 \) concentrations in the A1FI (a) and B1 (d) Baseline scenarios and percent differences for the \( \text{H}_2 \) fuel cell (\( \text{H}_2 \)-FC) and \( \text{H}_2 \) internal combustion engine (\( \text{H}_2 \)-ICE) scenarios (b, c, e and f). Note the different scales.

The tropospheric sulfate aerosol burdens decrease by 3% for the two \( \text{H}_2 \)-ICE scenarios. In the A1FI \( \text{H}_2 \)-ICE scenario, there is around 10–15% decrease in \( \text{SO}_4 \) mass concentration over most Eurasia, North Africa, western United States, Mexico, and South America (Fig. 15c). The \( \text{H}_2 \)-ICE scenario is slightly less powerful than the \( \text{H}_2 \)-FC scenario in \( \text{SO}_4 \) mitigation (Fig. 15f) because the former has greater oxidizing power, which makes the oxidation of \( \text{SO}_2 \) to sulfate aerosols faster than in the latter scenario.

4.6.3 Ammonium nitrate

In 2050, as today, annual mean surface ammonium nitrate aerosols (\( \text{NH}_4\text{NO}_3 \)) concentrations are higher in regions with intense human activities, such as the northeastern United States, Europe and eastern China and India (Fig. 16a and d). The highest annual mean surface concentrations (\( \sim 10 \mu \text{g m}^{-3} \)) are seen in eastern China and India in the A1FI BL scenarios. Surface ammonium nitrate aerosols concentrations are slightly lower in the B1 BL scenarios than in the A1FI.

The tropospheric ammonium nitrate aerosol burdens decrease by 12% and by 9% in the A1FI and B1 \( \text{H}_2 \)-FC scenarios, respectively (Tables 1 and 2). In the A1FI \( \text{H}_2 \)-FC scenario, surface concentrations decrease significantly around the world. Concentrations decrease by as much as 80%, for example, in California (Fig. 16b). Reductions are nearly as significant in the B1 \( \text{H}_2 \)-FC scenario as in the A1FI \( \text{H}_2 \)-FC scenario in terms of percent change (Fig. 16e).

The tropospheric ammonium nitrate aerosol burdens increase by 3% and by 2% in the A1FI and B1 \( \text{H}_2 \)-ICE scenarios, respectively (Tables 1 and 2). Surface concentrations increase in remote regions (Fig. 16a and d) as a result of the increased \( \text{NO}_x \) concentrations there (see Sect. 4.4). In the A1FI \( \text{H}_2 \)-FC scenario, there are slight concentration reductions (\( \sim 10\% \)) in the Ohio River Valley and in eastern China (Fig. 16b). The relative increase in remote regions is not significant because the background concentration is extremely small there (less than 0.1 \( \mu \text{g m}^{-3} \)) (Fig. 16a and d).

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Fig. 16. CAM-Chem simulated 2050 annual mean surface \( \text{NH}_4\text{NO}_3 \) concentrations in the A1FI (a) and B1 (d) baseline scenarios and percent differences for the \( \text{H}_2 \) fuel cell (\( \text{H}_2 \)-FC) and \( \text{H}_2 \) internal combustion engine (\( \text{H}_2 \)-ICE) scenarios (b, c, e, f). Note the different scales.

5 Conclusions

We have evaluated the possible impacts of transitioning to a \( \text{H}_2 \) powered road transportation sector in 2050 using atmospheric chemistry-climate models and emissions scenarios based on the IPCC A1FI and B1 SRES growth scenarios and fuel cell and internal combustion engine technology options. Our results show that air quality would improve significantly with adoption of a \( \text{H}_2 \) fuel cell type road transportation sector (\( \text{H}_2 \)-FC scenarios). Primary gaseous (CO, \( \text{NO}_x \)) and particulate (soot) pollutants are reduced substantially once the fossil fuel-based road transportation sector transitions to \( \text{H}_2 \) fuel cells. Ozone, a major secondary pollutant that often exceeds air quality standards in some regions of the United States (and is a severe problem in developing countries), would be reduced to a significant extent as its precursors, CO, \( \text{NO}_x \) and NMVOCs emissions, are reduced. Additionally, concentrations of particulate matter, especially those with diameters less than 2.5 \( \mu \text{m} \) (PM\(_{2.5}\)), would be significantly reduced in densely populated regions.

Some aspects of air quality would also improve with application of a \( \text{H}_2 \) internal combustion engine type road transportation sector (\( \text{H}_2 \)-ICE scenarios). Ambient ozone concentrations would decrease slightly as its precursor emissions by fossil fuel combustion from the road transportation sector are eliminated. Ozone mitigation with the \( \text{H}_2 \)-ICE option is not nearly as effective as in the \( \text{H}_2 \)-FC scenarios but is somewhat more effective in the relatively few VOC-limited regions. Urban PM\(_{2.5}\) concentrations would not be significantly reduced except in eastern China.

For each specific type of \( \text{H}_2 \) technology applied in the road transportation sector, the reductions of key air pollution species are not as significant in the B1 scenario as in the A1FI scenario. However, since the emissions of key ozone and aerosol precursors are lower in the B1 scenarios, air quality in the B1 scenarios is generally better than that in the A1FI scenarios. For example, tropospheric ozone concentrations are substantially lower in all of the B1 scenarios than in all of the A1FI scenarios regardless of the \( \text{H}_2 \) technology adopted.

With a leakage rate of 2.5 \%, \( \text{H}_2 \) emissions in a \( \text{H}_2 \)-based road transportation sector are larger than the replaced \( \text{H}_2 \) emissions from fossil fuel burning, leading to increased \( \text{H}_2 \) concentrations in the atmosphere. The added \( \text{H}_2 \) would decrease tropospheric OH abundance, resulting in elongated
CH$_4$ lifetime and thus warming. However, when taking changes in emissions of CO, NO$_x$ and NMVOCs into account, the picture of the climate impact is more complicated. In the H$_2$-FC scenarios, the combined effect of emission changes is a 4% decrease in OH abundance, which contributes to warming by increasing CH$_4$ lifetime. On the other hand, concurrent decrease in tropospheric O$_3$ concentrations would provide a cooling mechanism. The overall effect is further complicated by the temporal and spatial inhomogeneity of the two offsetting mechanisms. In the H$_2$-ICE scenarios, both decrease in CH$_4$ lifetime due to OH increase and the decrease in tropospheric O$_3$ tend to cool the climate, while, again, inhomogeneities in space and time would complicate the problem.

It is worth noting that our emission scenarios assumed 100% market penetration of H$_2$ technologies in 2050. That is, all vehicles operating on the road are replaced with H$_2$ powered ones. We also assumed that the H$_2$ for road transportation was produced by renewable, nuclear, or fossil fuel facilities with advanced scrubbing technologies and with no emissions associated with its production. The positive and negative impacts presented here will decrease in magnitude to the extent that market penetration is not total and also whether there are emissions associated with H$_2$ production.

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