Life cycle air quality impacts of conventional and alternative light-duty transportation in the United States

Christopher W. Tessum[a], Jason D. Hill[b,1], and Julian D. Marshall[a,1]

[a]Department of Civil, Environmental, and Geo- Engineering, University of Minnesota, Minneapolis, MN 55455; and [b]Department of Bioproducts and Biosystems Engineering, University of Minnesota, St. Paul, MN 55108

Commonly considered strategies for reducing the environmental impact of light-duty transportation include using alternative fuels and improving vehicle fuel economy. We evaluate the air quality-related human health impacts of 10 such options, including the use of liquid biofuels, diesel, and compressed natural gas (CNG) in internal combustion engines; the use of electricity from a range of conventional and renewable sources to power electric vehicles (EVs); and the use of hybrid EV technology. Our approach combines spatially, temporally, and chemically detailed life cycle emission inventories; comprehensive, fine-scale state-of-the-science chemical transport modeling; and exposure, concentration–response, and economic health impact modeling for ozone (O_3) and fine particulate matter ($PM_{2.5}$). We find that powering vehicles with corn ethanol or with coal-based or "grid average" electricity increases monetized environmental health impacts by 80% or more relative to using conventional gasoline. Conversely, EVs powered by low-emitting electricity from natural gas, wind, water, or solar power reduce environmental health impacts by 50% or more. Consideration of potential climate change impacts alongside the human health outcomes described here further reinforces the environmental preferability of EVs powered by low-emitting electricity relative to gasoline vehicles.

LCA | pollution | bioelectricity | externality | spatial

Society is in the midst of a great effort to understand and mitigate anthropogenic greenhouse gas (GHG) emissions and their effects on the global climate (1–5). However, GHG damages are not the only environmental impact of human activities, and are often not even the largest. In transportation, for example, non-GHG air pollution damage externalities generally exceed those from climate change (6–8). Here, we explore the air quality impacts of several proposed transportation fuel interventions: liquid biofuels (9), electric vehicles (EVs) powered by conventional and alternative energy sources (3), biomass feedstocks to power EVs (10, 11), compressed natural gas (CNG) powered vehicles (5), and improved vehicle fuel economy.

The air quality impacts of biofuels, transportation electrification, CNG vehicles, and improved fuel economy have been studied (refs. 7, 8, and 12–21; results are summarized in Table S1); our work advances prior research by combining estimates of life cycle emissions [i.e., emissions from production ("upstream") and consumption ("tailpipe") of the fuel] with an advanced air quality impact assessment. In addition, we incorporate greater spatial, temporal, and chemical detail than have prior research efforts. We also report non-GHG air quality life cycle impacts of biomass-powered EVs, which to our knowledge have not yet been described.

We use a spatially and temporally explicit life cycle inventory model (22) to estimate total fuel supply chain air pollutant emissions for scenarios where 10% of US projected vehicle miles traveled in year 2020 are driven in 1 of 11 types of passenger cars: (i) conventional gasoline powered vehicles (abbreviation:

"gasoline"); (ii) grid-independent hybrid EVs ("gasoline hybrid"); (iii) diesel powered light-duty vehicles ("diesel"); (iv) internal-combustion CNG vehicles ("CNG"); (v) vehicles powered by ethanol from corn grain through natural-gas–powered dry milling ("corn ethanol"); (vi) vehicles powered by cellulosic ethanol from corn stover ("stover ethanol"); and battery EVs ("EV") powered by electricity from the following: (vii) the projected year 2020 US average electric generation mix ("EV grid average"); (viii) coal ("EV coal"); (ix) natural gas ("EV natural gas"); (x) the combustion of corn stover ("EV corn stover"); and (xi) wind turbines, dynamic water power, or solar power ("EV WWS"). Because year 2020 electric generation infrastructure is not predetermined, we explore a range of electricity technologies rather than attempting to predict future electrical generation and dispatch deterministically; our approach can inform transportation and electricity generation policies in tandem. Based on prior research, we assume that the difference among scenarios in emissions from manufacturing and disposal of vehicles and from upstream infrastructure is small relative to differences in vehicle operation emissions (8, 23, 24) with the exception of lithium ion EV battery production. To highlight battery-related impacts, we analyze them separately from fuel-related impacts.

Significance

Our assessment of the life cycle air quality impacts on human health of 10 alternatives to conventional gasoline vehicles finds that electric vehicles (EVs) powered by electricity from natural gas or wind, water, or solar power are best for improving air quality, whereas vehicles powered by corn ethanol and EVs powered by coal are the worst. This work advances the current debate over the environmental impacts of conventional versus alternative transportation options by combining detailed spatially and temporally explicit emissions inventories with state-of-the-science air quality impact analysis using advanced chemical transport modeling. Our results reinforce previous findings that air quality-related health damages from transportation are generally comparable to or larger than climate change-related damages.
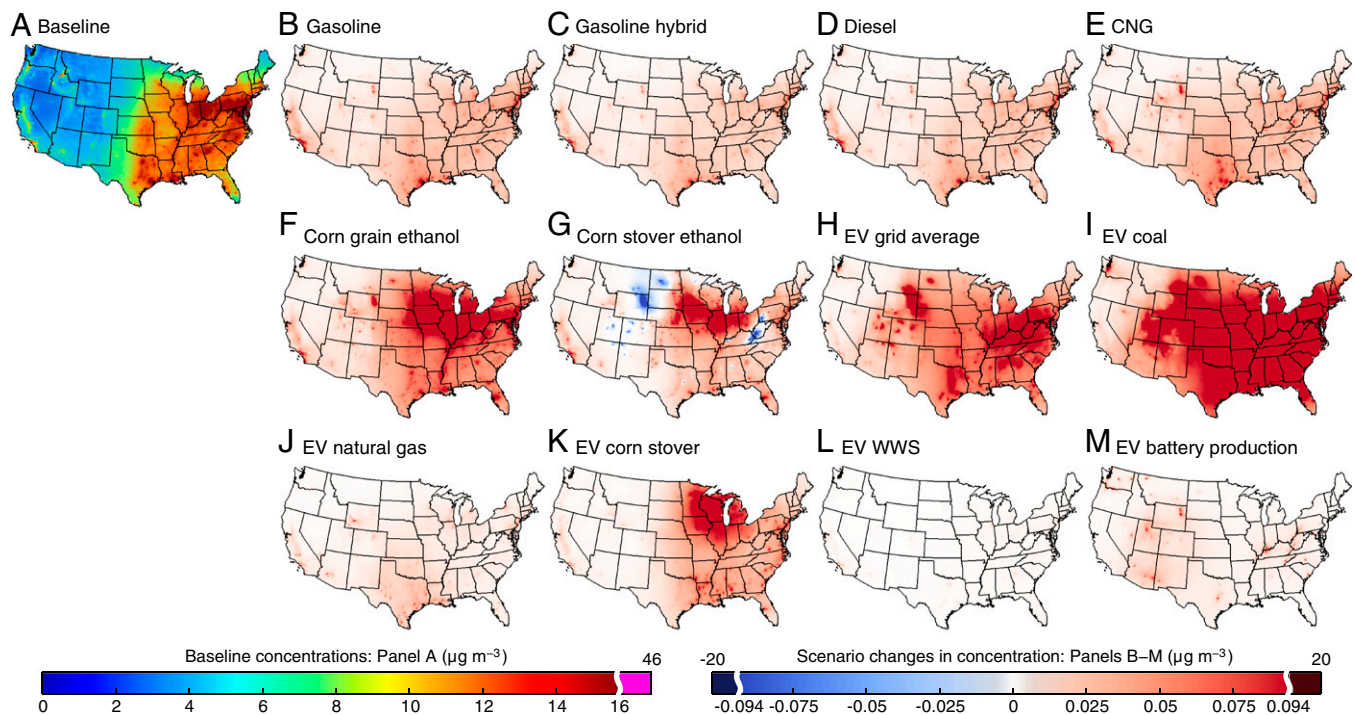
ENVIRONMENTAL SCIENCES

SUSTAINABILITY SCIENCE

Fig. 1. Annual average PM$_{2.5}$ concentrations. (*A*) Year 2005 baseline modeled concentrations. (*B–L*) Increase in concentration above the baseline attributable to replacement of 10% of year 2020 vehicle use with the given technology. (*M*) Increase in concentration attributable to EV battery manufacturing. Color scales contain a discontinuity at the 99th percentile of emissions. Abbreviations: CNG, compressed natural gas vehicle; EV, electric vehicle; WWS, wind, water, or solar. Analogous plots for O$_3$ are in Fig. S1.

We use spatially and temporally explicit simulations, including a state-of-the-science mechanistic meteorology and chemical transport model, to estimate for each scenario the changes in annual-average concentrations of the regulated pollutants fine particulate matter (PM$_{2.5}$) and ground-level ozone (O$_3$). We use spatially explicit population data (25) and results from major epidemiological studies (26, 27) to estimate increases in mortalities attributable to each scenario. We estimate monetized externalities from mortalities using a value of statistical life (VSL) metric. Results are given next; methods are described thereafter.

Results

Fig. 1 shows the spatial distributions of the background PM$_{2.5}$ concentrations (Fig. 1*A*) and of the changes in PM$_{2.5}$ concentrations for each scenario (Fig. 1 *B–L*). Spatial distributions of concentrations of O$_3$ (Fig. S1) and other species are in *SI Results*. As all scenarios represent increases in vehicle miles traveled, concentrations increase in almost all cases, as shown in Fig. 1. Changes in concentrations caused by alternative scenarios (Fig. 1 *C–L*) may be larger or smaller than the changes caused by the business-as-usual gasoline scenario (Fig. 1*B*). For the petroleum scenarios (gasoline, gasoline hybrid, and diesel; Fig. 1 *B–D*), vehicle tailpipe emissions are the largest source of impacts. Impacts from vehicle tailpipe emissions for the CNG (Fig. 1*E*) and ethanol (Fig. 1 *F* and *G*) scenarios are similar to those from the petroleum scenarios, but upstream processes, such as natural gas processing and compression (mainly in Texas) for the CNG scenario and agriculture (mainly in the upper Midwest) for the ethanol scenarios, cause additional substantial impacts. Industrial electricity use in the corn ethanol and CNG scenarios creates additional impacts in Wyoming and in the Appalachian Mountains owing to emissions from coal mining; emission credits from excess electricity generation in the stover ethanol scenario cause decreases in concentrations in the same geographic areas. Impacts are lower for the EV natural gas

scenario (Fig. 1*J*) than for the CNG scenario (Fig. 1*E*) for several reasons: emissions per vehicle-kilometer are lower for natural gas combustion in electricity generators than for CNG combustion in vehicles; natural gas must be compressed for use in CNG vehicles but not in electricity generation; and combustion emissions generally occur further from population centers for electricity generation than for vehicle tailpipe emissions. The EV corn stover scenario (Fig. 1*K*) causes farming-related impacts in the Midwest. Coal mining and combustion in the EV coal (Fig. 1*I*) and EV grid average (Fig. 1*H*) scenarios cause large impacts in Wyoming and the Appalachian Mountains; long-range transport of sulfur dioxide (SO$_2$) emitted during coal combustion causes impacts distributed over wide areas in those scenarios. Excluding battery production, impacts from wind, water, and solar (WWS) EVs (Fig. 1*L*) are notably lower than from the other scenarios because WWS electrical generators do not produce emissions while in use. Emissions from battery production for the EV scenarios are tracked separately. The main emission sources for EV battery production (Fig. 1*M*) are coal mining in Wyoming and the Appalachian Mountains and the extraction and refinement of raw material inputs, including copper in Arizona and Utah and aluminum in Washington State and the Appalachian Mountains.

Animated versions of Fig. 1 showing temporally explicit concentrations for PM$_{2.5}$ and O$_3$ are available in *SI Results*. Notable trends include spikes in PM$_{2.5}$ for the corn ethanol, corn stover ethanol, and EV corn stover scenarios caused by fertilizer application and nitrification emissions during the spring planting season. For these same scenarios, owing to complexities in the chemistry of O$_3$ formation and removal, O$_3$ concentrations in the Midwest "corn belt" tend to be increased relative to the baseline in the summer months, but decreased in the winter months.

Air pollution-related human health impacts for each scenario (PM$_{2.5}$- and O$_3$-related mortalities, and the corresponding monetized damages) are shown in Fig. 2. Total impacts range

from 230 mortalities per year ($0.14 per gallon gasoline-equivalent) for the WWS EV scenario to 3,200 mortalities per year ($1.94 per gallon gasoline-equivalent) for the coal EV scenario. Estimated mortality impacts from $PM_{2.5}$ are approximately an order of magnitude greater than those from O_3. Damages from the production of EV batteries are shown separately in Fig. 2. Scenarios with substantially decreased air quality-related health impacts compared with gasoline include gasoline hybrid vehicles (30% decrease) and EVs powered by natural gas or by WWS (50% and 70% decrease, respectively); scenarios with substantially higher damages than gasoline include corn ethanol (80% increase) and EVs powered by grid average or coal electricity (200% and 350% increase, respectively).

Changes in combined air pollution and climate damages attributable to each alternative scenario, relative to gasoline, are shown in Fig. 3. For most scenarios, air pollution impacts are comparable to or larger than climate change impacts. Although corn ethanol as modeled here emits marginally less GHGs than does gasoline, the combined climate and air quality impacts are greater than those from gasoline vehicles. (Our corn ethanol GHG results exclude impacts of indirect land-use change, and so likely are lower-bound GHG emission estimates.) EVs powered by grid-average electricity also have greater negative impacts than do vehicles powered by gasoline. Previous studies (3, 5) have argued that to meet stated goals for GHG emission reductions, it is necessary to both electrify vehicles and decarbonize electricity generation, for example through WWS electric generation. Our results suggest that such a strategy would have the strong cobenefit of substantially reducing air quality-related mortalities (by ~70%).

Given the considerable computational demands of our mechanistic meteorology and chemical transport modeling, comprehensive quantification of uncertainty within our analyses is impractical. Rather, we explore uncertainty via sensitivity analyses for several factors that are important in our analyses and that could impact the relative rankings of scenarios. Results are described below; additional detail, methods, and figures are in SI Results.

As shown in Fig. S2 and discussed in SI Results, EV battery production is unique among the life cycles investigated here in that up to about half of all emissions occur outside of our spatial modeling domain and are thus excluded from this analysis. We test the impact of this exclusion by doubling the impacts from EV battery production (Fig. S3). We find that this increases damage costs for EVs by $0.08 per gasoline gallon-equivalent, which is between a 4% increase for the coal EV scenario and a 57% increase for the WWS EV scenario, but does not change the rank order of air quality impacts among scenarios.

As emissions from coal mining are a major source of impacts in many of the scenarios we study, we examine the impact on our results of using lower coal mining emission factors (28, 29). As
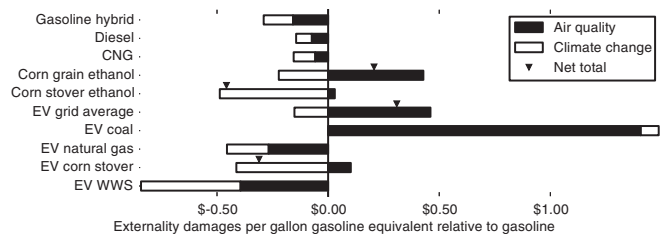
Fig. 3. Combined air quality plus climate change externalities attributable to each scenario, relative to the gasoline scenario. [The gasoline scenario impacts (air quality, $0.53/gallon; climate change, $0.46/gallon) would equal zero on this plot.] EV scenarios include battery production. Air quality impacts include $PM_{2.5}$ and O_3. For bars with both positive and negative values, the triangle above each bar shows the net total impact. GHG emissions from indirect land-use change are not included. See Fig. S4 for the impact of including indirect land-use change on net GHG emissions.

shown in Fig. S3, this shift does not change the rank order of air quality impacts among scenarios, although it decreases overall impacts from EVs powered by coal or grid average electricity by 37% and 33%, respectively.

To test the importance of incorporating full life cycle supply chain information when performing air quality impact assessment, we perform a sensitivity analysis that considers only emissions from the single phase of each life cycle most frequently associated with its environmental impacts (for internal combustion vehicles: on-road emissions; for EVs: emissions from electricity generation). This change causes corn ethanol to appear substantially less damaging because emissions during production and transport are overlooked (Fig. S3).

We also investigate the sensitivity of our results to a range of assumptions related to climate change (Fig. S4). We investigate the use of a lower carbon price, the inclusion of international emissions of climate forcers, the inclusion of indirect land-use change, the recently reported possibility of additional gas leakage during natural gas extraction (30), and sourcing crude oil from oil sands rather than from conventional sources (31). As illustrated in Fig. S4, conclusions of this study are generally robust to these perturbations.

Finally, we investigate the impact of model spatial resolution on estimated health impacts (Fig. S5). Higher resolution analyses tend to produce larger estimates for health impacts but do not alter the overall conclusions presented here.

Discussion

Results provided here combine spatially and temporally explicit life cycle assessment with state-of-the-science air quality modeling for a range of potential transportation technology interventions. The updated emissions estimates, better spatial resolution and coverage, better disaggregation of process locations, and more detailed air quality impact analysis incorporated here yield different overall conclusions from a similar recent study by Michalek et al. (8). For instance, when only air quality impacts are considered, Michalek et al. found that WWS EVs increase impacts by 160% compared with gasoline, whereas we find that WWS EVs reduce impacts by 70% compared with gasoline. Two major factors contributing to this difference are our use of a more recent version of the Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) model and Michalek et al.'s assumption that all emissions from EV battery manufacturing are colocated with automobile manufacturing facilities; our more detailed analysis places many of the most polluting processes in the battery production life cycle in remote areas. Additional discussion of this comparison is in SI Results.

We have considered here 10 alternative transportation fuel and technology options that have been put forward as potentially
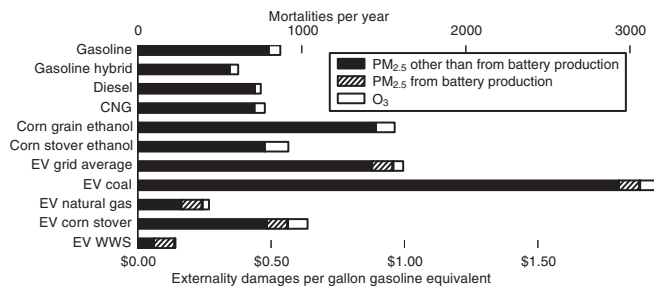

Fig. 2. Air quality health impacts in the United States for each scenario: attributable increases in annual mortality (upper scale) and the resulting monetized health impacts (lower scale).

environmentally preferable to conventional gasoline-powered vehicles based largely on reductions in fossil fuel use and GHG emissions. We find that, in some cases, such as for EVs powered by natural gas or WWS, considering air quality impacts alongside climate impacts increases the apparent environmental benefit of the alternative fuel relative to gasoline; in both cases (EVs from natural gas; EVs from WWS), the air pollution benefits relative to gasoline are larger than the climate-related benefits. Other fuels, such as corn ethanol [the climate impact of which is unclear (32)], are more damaging than conventional vehicles when air pollution impacts are considered alone or when air pollution and climate impacts are considered together. The difference between the least- and most-polluting electricity generation options for EVs increases almost sixfold when air pollution damages are considered alongside climate impacts, instead of when climate impacts are considered alone. Our findings thus reinforce the benefit of pairing EVs with clean electricity (3, 5).

Our work supports the inclusion of air pollution health impacts when assessing the environmental impact of transportation; in monetized terms, the health impacts can be as great, or greater than, effects on climate change. We also demonstrate the importance of spatial, temporal, and chemical detail and precision in life cycle air quality impact assessment. Although climate change and air quality are often considered the two main transportation-related environmental externalities (6), inclusion of other life cycle impact categories not considered here, such as environmental justice, water quality and availability (e.g., 33, 34), biodiversity, or vehicle safety could add additional insight.

Results given here should not be taken as a final statement that environmental improvements are best achieved by replacing existing light-duty vehicles with less-polluting light-duty vehicles, nor that EVs are the best technology for every transportation need. Instead, these results can be seen as an indication of how light-duty transportation fuels could shift to reduce or increase pollution, and as an encouragement into the research of less polluting, more sustainable transportation options for the future.

Materials and Methods

Scenario Selection. We use a functional unit of 388 billion miles per year, which is 10% of year 2020 projected US vehicle miles traveled (35). For the ethanol scenarios, this value corresponds to blending an average of 10% ethanol with gasoline on an energy equivalent basis. For EV scenarios, it corresponds to aggressive but plausible adoption of EVs (36).

We do not consider constraints on the availability or cost of resources (e.g., the viability of increased ethanol production; availability of WWS electricity). However, we have found that the air quality impacts we consider scale approximately linearly with changes in the size of the functional unit, so as a first approximation air quality impacts from different amounts of miles traveled or fuel produced could be interpolated from our results.

Life Cycle Inventory. We create a chemically, spatially, and temporally detailed life cycle inventory for each scenario using the GREET-chemical, spatial, and temporal (GREET-cst) model (22), which is based on the GREET model, version 1.8d1, from Argonne National Laboratory (31). It is worth noting that subsequent versions of GREET have been released since version 1.8d1. We investigate the sensitivity of our results to some of the changes in subsequent GREET versions, as well as to other important factors; investigating the effect of GREET version on the estimated air quality impacts is an area for future research. GREET-cst outputs emissions that are spatially allocated to a 12-km grid covering the continental United States and surrounding waters, chemically speciated according to the CB05 chemical mechanism (37), and temporally disaggregated in 1-h time increments. The GREET model can be obtained at greet.es.anl.gov/.

For assumptions specific to GREET-cst, including spatial modeling domain, spatial locations of emissions, chemical speciation, and temporal profiles, we configure the model as described by Tessum et al. (22). Briefly, we use the continental United States and its surrounding waters as a spatial model domain and exclude from the analysis all emissions occurring outside of the domain. We assume all emissions from individual processes, including vehicle

end use, occur at existing production or activity locations. For material transport emissions, we use network analysis and linear optimization to simulate transport behavior along roadways, rails, and navigable water routes between source and destination locations. We apply temporal and chemical speciation profiles from the 2005 National Emissions Inventory (NEI) (38) to the emissions from each process. We use the results of Bashash et al. (39) for EV battery charging temporal profiles, which assume batteries would be charged to optimize for long battery life and low electricity costs. We use these charging profiles only to determine the timing for emissions release from power plants; for the mix of electrical generation sources, we use the hypothetical scenarios discussed in the Introduction.

For most processes that use electricity, such as electricity generation for use in EVs in the EV coal and EV natural gas scenarios, we assume that electricity generation comes from the same North American Electrical Reliability Corporation (NERC) electrical grid region as the end use process as in Tessum et al. (22) and that the mix of generation fuels and technologies in each NERC region is the same as the national average. For electricity generation in the EV corn stover scenario, however, because of the logistical difficulty in transporting corn stover, we assume that electricity generation for EVs occurs in the same electrical grid region as the corn stover production instead of vehicle end use. For the EV grid average scenario, we assume that electricity generation for EV use is distributed according to total generation amounts in the year 2007 (40) so as to preserve the difference in fuel mixes for electric generation across NERC regions. Further details are in *SI Results*.

In GREET, we model year 2020 passenger cars using default settings with the following exceptions:

- The gasoline used is 100% conventional (0% reformulated gasoline) so as to disentangle the effect of ethanol blends.
- Because almost all oil extraction from oil sands occurs outside of our geographic modeling domain, we assume all oil is extracted conventionally (0% oil sands oil). The sensitivity of our results to this assumption is explored in *SI Results*.
- For gasoline hybrid vehicles, GREET assumes that the tailpipe emission factors for volatile organic compounds, CO, PM_{10}, $PM_{2.5}$, and N_2O are the same as for conventional gasoline vehicles on a per-mile basis. We therefore adjust gasoline hybrid tailpipe emissions of these species to be 71% (the ratio of hybrid to conventional fuel efficiency) of the conventional vehicle emissions factors.
- We do not include land-use change emissions for the ethanol or bioelectricity scenarios.
- We assume all corn ethanol plants use dry milling and 100% natural gas for process heat.
- We assume that ethanol is produced as 100% ethanol with no denaturant, but for ethanol scenarios we use tailpipe emissions factors for a 10% blend of ethanol with gasoline ("E10").
- We update the year 2020 projection for electricity generation mix to a more recent Energy Information Administration projection (41).
- We add a corn stover bioelectricity pathway to the GREET 1.8d1 spreadsheet based on parameters from the other bioelectricity pathways and the corn stover ethanol pathway.

We do not assume that marginal emissions from electrical generation or other sources are subject to cap-and-trade or other regulation (e.g., as in refs. 12 and 42). We use the GREET default assumption that electricity from biomass (corn stover) is generated using a dedicated biomass boiler. Cofiring biomass with coal may yield different emissions.

We assume that impacts related to the manufacture and disposal of equipment used to supply fuel (e.g., electrical generating units, oil wells, manufacturing plants), as well as those from the manufacture and disposal of vehicles, do not differ among the scenarios investigated here (8, 23, 24), and we exclude them from this analysis. Although these emissions do not differ much among scenarios, their impact on the overall air quality-related damages from transportation may be important (8, 23) and is an area for future research. We do include, however, production of EV batteries, which we model as a separate scenario to show explicitly the contribution of EV battery manufacturing to total impacts. We model the emissions from the production of enough lithium ion batteries to power our functional unit of 10% of the projected year 2020 US vehicle miles traveled by expanding GREET-cst to include the GREET2 vehicle cycle model, version 2012 (23). We do this step using the methods of Tessum et al. (22) and including additional spatial data on the locations of aluminum manufacturing (43), copper manufacturing (44), plastics manufacturing (38), steel manufacturing (38), and battery assembly (38). Table S2 shows, for each process, the fractions of emissions from battery production processes that are excluded from the analysis because they occur outside our spatial modeling domain. Information on

international fractions for other processes is reported by Tessum et al. (22). Fig. S2 shows amounts of domestic (included) and international (excluded) emissions from battery production. Full GREET-cst results for battery production are included in Dataset S1. We use the GREET2 default EV battery life assumption of 160,000 miles.

Emissions of GHGs and radiatively active aerosols are calculated using the same assumptions as above (i.e., the exclusion of indirect, market-mediated effects and the exclusion of emissions occurring outside the United States), and reported as CO_2 equivalent emissions using global warming potential conversions reported by Bond et al. (45). We use GREET default values for all emissions factors and process relationships not otherwise mentioned here.

Chemical Transport Modeling. To estimate changes in $PM_{2.5}$ and O_3 concentrations attributable to each scenario, we run the Weather Research and Forecasting with Chemistry (WRF-Chem) Eulerian meteorology and chemical transport model, version 3.4 (46), using a 12-km resolution grid with 444 rows, 336 columns, and 28 vertical layers. The modeling domain (5,328 km × 4,032 km) covers the continental United States, southern Canada, and northern Mexico [see the study by Tessum et al. (47)].

Setup of WRF-Chem as used here is detailed elsewhere (47). We use the Regional Atmospheric Chemistry Mechanism (RACM) (48) for gas phase reactions, and the Modal Aerosol Dynamics for Europe (MADE) (49) module for aerosol chemistry and physics. We use the Volatility Basis Set (VBS) (50) to simulate formation and evaporation of secondary organic aerosols.

We compare the change in emissions attributable to each scenario to a baseline or reference case. For the baseline emissions we use the 2005 NEI (38), which includes area, point, and mobile emissions from year 2005 for the United States, year 2006 for Canada, and year 1999 for Mexico. The performance of WRF-Chem as used here in reproducing observed $PM_{2.5}$ and O_3 concentrations is reported by Tessum et al. (47); the annual mean fractional bias and error, respectively, are −1% and 21% for $PM_{2.5}$, and 11% and 14% for daily peak O_3.

We use year 2005 baseline emissions and meteorology inputs. The NEI has been projected to future years, but we are only able to compare our model predictions to observed pollutant levels for years for which there is an available emissions inventory, pollution monitoring data, and meteorological observations. At the time the simulations described here were run, 2005 was the most recent year for which all of these inputs existed.

To predict changes in pollutant concentrations attributable to each scenario, we combine the emissions from each scenario with the baseline NEI, rerun WRF-Chem with the resulting emissions, and then subtract the baseline NEI-derived concentrations from the concentrations calculated using the combined emissions. Because some scenarios involve credits for displaced coproducts that we model as negative emissions, and because negative emissions cannot be represented in WRF-Chem, we deal with any grid cell with net negative emissions (where the baseline NEI value plus the scenario value is negative) by subtracting emissions from nearby cells, thereby zeroing out the negative cells while achieving the targeted local total mass of emissions.

Health Impact Assessment. We use as health endpoints respiratory mortalities attributable to ambient O_3 concentrations and mortalities from all causes attributable to ambient $PM_{2.5}$ concentrations. We use concentration–response values reported by Jerrett et al. (27) for O_3 and Krewski et al. (26) for $PM_{2.5}$. Jerrett et al. reported that, after accounting for covariation with $PM_{2.5}$ concentrations, a 10-ppb increase in April to September average daily peak-hour O_3 concentration causes a 4% increase in respiratory mortalities. Krewski et al. reported that a 10 $\mu g \cdot m^{-3}$ increase in annual average $PM_{2.5}$ concentration causes a 7.8% increase in all-cause mortalities; the Krewski et al. model does not account for covariation with O_3 concentrations. Both results are based on the Cox proportional hazards model, which assumes that the risk ratio (RR) varies exponentially with change in pollutant concentration (ΔC) as in Eq. **1**, where the β coefficients for O_3 (0.00392 ppb^{-1}) and $PM_{2.5}$ [0.00751 ($\mu g\ m^{-3}$)$^{-1}$] are calculated from the results of Jerrett et al. and Krewski et al., respectively. WRF-Chem is configured to output instantaneous concentrations at the beginning of each hour, rather than hourly average concentrations; when calculating ΔC for each scenario, we use the instantaneous concentrations output by WRF-Chem as a surrogate for hourly average concentrations. We use Eq. **2** to calculate total annual mortalities (D) attributable to each scenario, where n is the total number of ground-level grid cells, P is the population in each cell based on year 2000 US Census block group-level data (25), and M is the county-specific population average baseline all-cause (for $PM_{2.5}$) or respiratory (for O_3) mortality rate (51):

$$RR = \exp(\beta \Delta C), \quad [1]$$

$$D = \sum_{i=1}^{n} (\exp(\beta \Delta C_i) - 1) P_i M_i. \quad [2]$$

Economic Valuation. We estimate economic damages from the deaths attributable to air pollution impacts of each scenario using a VSL of $10.1 million (2012$) using US Environmental Protection Agency methodology for income year 2020 (52). We assume no lag time between pollutant emissions and the resulting health effects. We estimate the economic damages from emissions GHGs and radiatively active aerosols using a social cost of carbon of $180 MgC^{-1} ($49 MgCO$_2$$^{-1}$) (53) (mean value, 1% discount rate, adjusted to 2012$) in the main analyses and $23 MgC^{-1} ($6.19 MgCO$_2$$^{-1}$) (2012$) in a sensitivity analysis.

1. Intergovernmental Panel on Climate Change (2007) *Climate Change 2007: Synthesis Report* (Intergovernmental Panel on Climate Change, Geneva). Available at www.ipcc.ch/publications_and_data/publications_ipcc_fourth_assessment_report_synthesis_report.htm. Accessed October 10, 2012.
2. Pacala S, Socolow R (2004) Stabilization wedges: Solving the climate problem for the next 50 years with current technologies. *Science* 305(5686):968–972.
3. Williams JH, et al. (2012) The technology path to deep greenhouse gas emissions cuts by 2050: The pivotal role of electricity. *Science* 335(6064):53–59.
4. United Nations (1998) *Kyoto Protocol to the United Nations Framework Convention on Climate Change.* Available at unfccc.int/kyoto_protocol/items/2830.php/. Accessed October 10, 2012.
5. National Research Council (2013) *Transitions to Alternative Vehicles and Fuels.* Available at www.nap.edu/catalog.php?record_id=18264. Accessed April 10, 2013.
6. Delucchi MA (2000) Environmental externalities of motor-vehicle use in the US. *J Transp Econ Policy* 34(2):135–168.
7. Hill J, et al. (2009) Climate change and health costs of air emissions from biofuels and gasoline. *Proc Natl Acad Sci USA* 106(6):2077–2082.
8. Michalek JJ, et al. (2011) Valuation of plug-in vehicle life-cycle air emissions and oil displacement benefits. *Proc Natl Acad Sci USA* 108(40):16554–16558.
9. US Environmental Protection Agency (2010) Regulation of fuels and fuel additives: Changes to renewable fuel standard program; Final Rule. 75. *Federal Register* 58 (2010), pp 14670–14904.
10. Campbell JE, Lobell DB, Field CB (2009) Greater transportation energy and GHG offsets from bioelectricity than ethanol. *Science* 324(5930):1055–1057.
11. Ohlrogge J, et al. (2009) Energy. Driving on biomass. *Science* 324(5930):1019–1020.
12. Electric Power Research Institute (2007) *Environmental Assessment of Plug-In Hybrid Electric Vehicles, Volume 2: United States Air Quality Analysis Based on AEO-2006 Assumptions for 2030.* Available at www.epri.com/abstracts/Pages/ProductAbstract.aspx?ProductId=000000000001015326. Accessed November 24, 2014.
13. Jacobson MZ (2007) Effects of ethanol (E85) versus gasoline vehicles on cancer and mortality in the United States. *Environ Sci Technol* 41(11):4150–4157.
14. Boureima F-S, et al. (2009) Comparative LCA of electric, hybrid, LPG and gasoline cars in Belgian context. *World Elec Vehicle J* 3:1–8.
15. National Research Council (2009) *Hidden Costs of Energy: Unpriced Consequences of Energy Production and Use.* Available at www.nap.edu/catalog.php?record_id=12794. Accessed November 24, 2014.
16. Thompson T, Webber M, Allen DT (2009) Air quality impacts of using overnight electricity generation to charge plug-in hybrid electric vehicles for daytime use. *Environ Res Lett* 4(1):014002.
17. Cook R, et al. (2010) Air quality impacts of increased use of ethanol under the United States' Energy Independence and Security Act. *Atmos Environ* 45(40):7714–7724.
18. Brinkman GL, Denholm P, Hannigan MP, Milford JB (2010) Effects of plug-in hybrid electric vehicles on ozone concentrations in Colorado. *Environ Sci Technol* 44(16):6256–6262.
19. US Environmental Protection Agency (2010) *Renewable Fuel Standard Program (RFS2) Regulatory Impact Analysis*, EPA-420-R-10-006. Available at www.epa.gov/otaq/renewablefuels/420r10006.pdf. Accessed December 3, 2012.
20. Alhajeri NS, McDonald-Buller EC, Allen DT (2011) Comparisons of air quality impacts of fleet electrification and increased use of biofuels. *Environ Res Lett* 6(2):024011.
21. Thompson TM, King CW, Allen DT, Webber ME (2011) Air quality impacts of plug-in hybrid electric vehicles in Texas: Evaluating three battery charging scenarios. *Environ Res Lett* 6(2):024004.
22. Tessum CW, Marshall JD, Hill JD (2012) A spatially and temporally explicit life cycle inventory of air pollutants from gasoline and ethanol in the United States. *Environ Sci Technol* 46(20):11408–11417.

ENVIRONMENTAL SCIENCES

SUSTAINABILITY SCIENCE

23. Wang M, Burnham A, Wu Y (2012) *The Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation Model v2_2012*. Available at greet.es.anl.gov/. Accessed November 24, 2012.

24. MacLean HL, Lave LB (2003) Life cycle assessment of automobile/fuel options. *Environ Sci Technol* 37(23):5445–5452.

25. US Census Bureau (2000) Census 2000 Datasets. Available at www2.census.gov/census_2000/datasets/. Accessed June 12, 2013.

26. Krewski D, et al. (2009) Extended follow-up and spatial analysis of the American Cancer Society study linking particulate air pollution and mortality. *Res Rep Health Eff Inst* 2009(140):5–114; discussion 115–136.

27. Jerrett M, et al. (2009) Long-term ozone exposure and mortality. *N Engl J Med* 360(11):1085–1095.

28. US Environmental Protection Agency (2013) *Fugitive Dust from Mining and Quarrying (2325000000)*. Available at ftp://ftp.epa.gov/EmisInventory. Accessed August 12, 2013.

29. Xstrata Coal (2012) *Ravensworth Underground Mine—Coal Mine Particulate Matter Control Best Management Practice Determination*. Available at www.xstratacoalravensworth.com.au/EN/RavensworthUndergroundMine/Publications/Mt plans and programs/RUM%20Coal%20Mine%20PM%20BMP%20Determination.pdf. Accessed August 26, 2013.

30. Brandt AR, et al. (2014) Energy and environment. Methane leaks from North American natural gas systems. *Science* 343(6172):733–735.

31. Wang M (2010) *The Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation Model v1.8d1*. Available at greet.es.anl.gov/. Accessed March 13, 2011.

32. National Research Council (2011) *Renewable Fuel Standard: Potential Economic and Environmental Effects of U.S. Biofuel Policy*. Available at www.nap.edu/catalog.php?record_id=13105. Accessed November 24, 2014.

33. Costello C, Griffin WM, Landis AE, Matthews HS (2009) Impact of biofuel crop production on the formation of hypoxia in the Gulf of Mexico. *Environ Sci Technol* 43(20):7985–7991.

34. King CW, Webber ME (2008) Water intensity of transportation. *Environ Sci Technol* 42(21):7866–7872.

35. US Department of Transportation (2008) *Transportation Vision for 2030*. Available at www.rita.dot.gov/sites/default/files/rita_archives/rita_publications/transportation_vision_2030/index.html. Accessed December 13, 2012.

36. Balducci PJ (2008) *Plug-in Hybrid Electric Vehicle Market Penetration Scenarios* (US Department of Energy, Washington, DC), PNNL-17441. Available at www.pnl.gov/main/publications/external/technical_reports/pnnl-17441.pdf. Accessed November 24, 2014.

37. Yarwood G, Rao S, Yocke M, Whitten GZ (2005) *Updates to the Carbon Bond Mechanism: CB05* (US Environmental Protection Agency, Washington, DC), RT-04-00675. Available at www.camx.com/publ/pdfs/CB05_Final_Report_120805.pdf. Accessed November 24, 2014.

38. US Environmental Protection Agency (2009) *2005 National Emissions Inventory (NEI)*. Available at www.epa.gov/ttn/chief/emch/index.html. Accessed March 7, 2012.

39. Bashash S, Moura SJ, Fathy HK (2011) On the aggregate grid load imposed by battery health-conscious charging of plug-in hybrid electric vehicles. *J Power Sources* 196(20):8747–8754.

40. US Environmental Protection Agency (2010) 2007 eGRID Database. Available at www.epa.gov/cleanenergy/energy-resources/egrid/index.html. Accessed July 3, 2012.

41. US Energy Information Administration (2011) *Annual Energy Outlook 2011* (Department of Energy, Washington, DC), DOE/EIA-0383.

42. Murphy CW, Parker NC (2014) Impact of air pollution control costs on the cost and spatial arrangement of cellulosic biofuel production in the U.S. *Environ Sci Technol* 48(4):2157–2164.

43. US Geological Survey (1999) *Mineral Industry Surveys: Primary Aluminum Plants*. Available at minerals.usgs.gov/minerals/pubs/commodity/aluminum/alplants.html. Accessed January 8, 2013.

44. Edelstein BDL (2010) *U.S. Geological Survey 2010 Minerals Yearbook: Copper* (US Geological Survey, Reston, VA), Available at minerals.usgs.gov/minerals/pubs/commodity/copper/myb1-2010-coppe.pdf. Accessed January 28, 2013.

45. Bond T, Venkataraman C, Masera O (2004) Global atmospheric impacts of residential fuels. *Energy Sustain Dev* 8(2):20–32.

46. Grell GA, et al. (2005) Fully coupled "online" chemistry within the WRF model. *Atmos Environ* 39(37):6957–6975.

47. Tessum CW, Hill JD, Marshall JD (2014) Twelve-month, 12 km resolution North American WRF-Chem v3.4 air quality simulation: Performance evaluation. *Geosci Model Dev Discuss* 7:8433–8476.

48. Stockwell W, Kirchner F, Kuhn M, Seefeld S (1997) A new mechanism for regional atmospheric chemistry modeling. *J Geophys Res* 102(D22):25847–25879.

49. Ackermann IJ, et al. (1998) Modal aerosol dynamics model for Europe: Development and first applications. *Atmos Environ* 32(17):2981–2999.

50. Ahmadov R, et al. (2012) A volatility basis set model for summertime secondary organic aerosols over the eastern United States in 2006. *J Geophys Res* 117(D6):D06301.

51. US Centers for Disease Control and Prevention (2000) Compressed Mortality File (2000). Available at www.cdc.gov/nchs/data_access/cmf.htm. Accessed November 6, 2011.

52. US Environmental Protection Agency (2013) *Technical Support Document: Estimating the Benefit per Ton of Reducing $PM_{2.5}$ Precursors from 17 Sectors*. Available at www.epa.gov/airquality/benmap/models/Source_Apportionment_BPT_TSD_1_31_13.pdf. Accessed November 24, 2014.

53. Tol RSJ (2009) The economic effects of climate change. *J Econ Perspect* 23(2):29–51.

Supporting Information

Tessum et al. 10.1073/pnas.1406853111

SI Results

Sensitivity Analyses. We perform several sensitivity analyses to evaluate the impact of key assumptions. The results of the sensitivity analyses are shown in Figs. S3–S5 and are described below and in the main text. All sensitivity analyses in Fig. S3 are performed for $PM_{2.5}$-related health impacts for the month of September only; as shown in Fig. S6, health impacts based on September $PM_{2.5}$ concentrations for the scenarios are similar to health impacts based on annual average concentrations. The grid resolution sensitivity analysis shown in Fig. S5 was performed for the month of July to capture summer peak O_3 conditions.

As shown in Fig. S2, not all emissions from the fuel life cycles occur within our spatial modeling domain, and therefore some emissions are excluded from our analysis. Refer to Tessum et al. (1) for more information. The electric vehicle (EV) battery production scenario has the most substantial fraction of emissions assumed to occur outside of the United States, with around 30–40% of emissions of most pollutants from battery production being excluded from the analysis (Fig. S2). We explore the sensitivity of our results to this assumption by doubling health impacts from battery production (Fig. S3) and find that the rank order of impacts among the different scenarios remains unchanged. For scenarios other than battery production, in most cases more than 90% of emissions occur inside the spatial modeling domain (Fig. S2). A fraction (30–45%) of SO_x and NO_x emissions from the petroleum scenarios (gasoline, gasoline hybrid, and diesel) are also excluded from the analysis, but because the excluded emissions are mainly from the extraction of crude oil (1), which largely occurs over the open ocean or far from population centers, their exclusion is not likely to impact our overall conclusions. These international upstream emissions are also excluded from fossil fuel use in the corn grain and stover ethanol scenarios.

Additionally, as emissions from coal mining and cleaning cause a substantial fraction of the total health impacts for some scenarios, and recent estimates of emissions factors for coal mining and cleaning for surface mining (2) and for underground mining (3) exist, we explore updating the GREET model with the new emission factors and rerunning all analyses for the month of September. We find that the change in coal mining and cleaning emissions factors does not affect the rank order of scenario impacts (Fig. S3) but does substantially reduces the air pollution impacts of some scenarios.

We additionally compare the impacts from the full life cycle of the fuels to impacts from either the vehicle tailpipe only for internal combustion vehicles or the electrical generation units only for EVs. As shown in Fig. S3, this sensitivity analysis affects the rank order of scenario impacts, further demonstrating the importance of including the emissions entire life cycle when performing environmental impact assessment for transportation fuels.

We also perform a set of sensitivity analyses investigating the effects climate-related assumptions on our results as shown in Fig. S4.

i) Fig. 3 in the main text excludes emissions occurring outside the United States for pollutants affecting both air quality and climate change. We investigate the effects of including these international emissions (exclusive of indirect land-use change) on climate change impacts. We find that including international climate-related emissions does not change overall damage costs by more than $0.02 per gallon gasoline equivalent for any scenario.

ii) We investigate the impact of including indirect land-use change emissions as calculated by Plevin et al. (4) (using a value of 80 g $CO_2e \cdot MJ^{-1}$, which is near the middle of the range of estimates in that paper) for the corn ethanol scenario. This substantially increases the overall externality damages from that scenario and reinforces the overall conclusion that corn ethanol is not an attractive alternative fuel in terms of air pollution or climate change impacts.

iii) We investigate the sensitivity of our results to carbon pricing using market-based carbon price of $6.19 Mg^{-1} CO_2 (from www.pointcarbon.com/productsandservices/carbon/ as of March 9, 2013, adjusted to 2012$) as opposed to the $49 Mg^{-1} CO_2 price used in Fig. 3. We find that this changes the sign of the net externality damages of the EV corn stover scenario from negative to positive but does not impact the overall conclusions presented in this paper.

iv) Recent analysis by Brandt et al. (5) suggests that CH_4 emissions may be systematically underestimated in emissions inventories, especially for natural gas extraction. We update the GREET model with the middle value for CH_4 leakage during natural gas extraction from Brandt et al. (5), resulting in 1.2% leakage on average instead of the GREET 1.8d1 default of 0.35%. We find that this does not affect the overall conclusions presented here. Although increased CH_4 emissions may also impact O_3 concentrations, we assume the effect on our overall conclusions to be negligible because O_3 health impacts are small relative to $PM_{2.5}$ health impacts.

v) Because almost all oil extraction from oil sands occurs outside of our geographic modeling domain, our baseline analysis assumes all oil is extracted conventionally (0% oil sands oil). This sensitivity analysis assumes that the GREET 1.8d1 year 2020 default value of 21% of crude oil comes from oil sands. We find that this does not affect the overall conclusions presented here. The use of oil sands oil instead of conventionally extracted crude may also affect air pollution concentrations, but the difference in health impacts is likely small because both the Canadian oil sands and most conventional extraction locations are typically located far from population centers.

The final sensitivity analysis investigates the impact of model spatial resolution on calculated health impacts (Fig. S5). For $PM_{2.5}$, total estimated impacts increase ∼10–15% when going from 36- to 12-km resolution, and another 5% when going from 12- to 4-km resolution. (O_3 impacts are not highly dependent on grid resolution owing to the comparatively smaller spatial gradients in O_3 concentrations.) Our contiguous-United States, 12-km resolution analysis is an improvement over previous studies, which used 36-km or county-level resolution or considered only part of the United States; Table S1); still, our approach is potentially susceptible to underestimation of near-source exposures. This dependence of impacts on model spatial resolution is likely caused by numerical dispersion and is likely most pronounced in scenarios where the most emissions occur in urban areas (i.e., the gasoline, diesel, and gasoline hybrid scenarios). It currently is not computationally practical to perform the full methodology reported here at 4-km or finer resolution; current models capable of higher resolution analyses (e.g., Gaussian plume models) do so at the expense of the chemical and physical representation of processes that our findings suggest are important (e.g., formation of secondary $PM_{2.5}$).

Comparison with Michalek et al. (6). An analysis by Michalek et al. (6) finds that, in terms of air quality-related health impacts, EVs do not compare favorably to conventional gasoline vehicles: when only emissions from battery production and from brake and tire wear during vehicle use are considered (which is equivalent to the WWS EV scenario presented here), they find that EVs cause air quality-related damages 150% greater than do conventional gasoline vehicles. [Michalek et al., table S25, adjusted to make equivalent for comparison by excluding the following: (*i*) vehicle and battery production for gasoline vehicles; (*ii*) vehicle and electricity production for EVs; and (*iii*) GHG, CO, and oil premium impacts for both vehicles.] Our analysis, however, finds that WWS EVs reduce impacts by 70% compared with conventional gasoline vehicles. In both studies, the main source of WWS EV impacts is battery production. We are aware of two major reasons for the difference between our and Michalek et al.'s results: (*i*) differences in the estimates of amounts of emissions, and (*ii*) differences in the modeled locations of battery manufacturing processes.

i) Michalek et al. use a customized version of GREET 2.7 to calculate emissions from battery production, whereas we use the default settings in the more recently released GREET2_2012. (Note: GREET 2 is not an updated version of GREET 1. GREET 1.x models fuel pathways, and GREET 2.x models vehicle production pathways.) Comparing our emissions results (Dataset S1) to those of Michalek et al. (table S3 in their study), our emissions estimates are substantially lower than theirs—87% lower for SO_2 emissions (80% lower if international emissions are included). The battery size used in both studies is similar (66.1 kWh in Michalek et al.; 63 kWh in our study). The differences in emissions instead appear to be caused by differences between GREET versions—among other differences, GREET2_2012 uses $LiMn_2O_4$ batteries in place of the $LiCoO_2$ batteries used in GREET 2.7 (7)—and our use of year 2020 grid-average electric generation mix for electricity used in battery production, which is cleaner than the year 2010 mix used by Michalek et al.

ii) Michalek et al. assume processes upstream from EV battery manufacturing are colocated with automobile manufacturing facilities, but our more detailed analysis shows that, for example, copper ore smelting, which causes the majority of battery production SO_2 emissions, mainly occurs in the sparsely populated southwestern United States (8). Because the production of copper and other raw materials for batteries occurs far from people, even if impacts from battery production as calculated here are doubled to adjust for emissions that occur outside of our spatial modeling domain, impacts from WWS EVs would still be 57% lower than conventional gasoline vehicle impacts. We test this hypothesis by using the ratio of population-weighted average ground-level concentrations to area-weighted average ground-level concentrations as an imperfect surrogate for the proximity of emissions sources to people. The ratio for our results for EV battery production is 1.9, lower than any of the other scenarios. The ratios for the other scenarios

range between 3 and 9. If emissions from battery production were located so as to give a population-weighted average to domain average ratio of 9 instead of 1.9, impacts from WWS EVs would be ~30% greater than impacts from conventional gasoline vehicles, which is closer to the result reported by Michalek et al. GREET assumes zero transportation emissions between mining operations and smelting facilities. This implies that smelting occurs at the mining site; to maintain consistency with GREET, we have maintained that assumption in our analyses. Given the potentially large importance of those emissions in estimating the impacts of battery EVs, further investigation of this topic is warranted.

Sensitivity of Results to EV Battery Life. In these analyses, we use the GREET default assumption that EV battery life is 160,000 miles: the same as the life of the rest of the vehicle. To explore a hypothetical scenario where battery life is only 100,000 miles, we multiply our results for air quality impacts from battery production by a factor of 1.6. This gives results similar to the sensitivity analysis in Fig. S3 where we double battery impacts: the air quality impacts of the EV scenario increase (WWS EVs, 34% increase; natural gas EVs, 18%; corn stover EVs, 7%; grid average EVs, 5%; coal EVs, 2%), but the rank order of scenarios does not change.

Model Availability. The GREET model is available at greet.es.anl. gov/. GREET-cst is freely available upon request. WRF-Chem is available at www2.mmm.ucar.edu/wrf/users/. The SMOKE model is available at www.cmascenter.org/smoke/. Our program used to convert between SMOKE and GREET-cst output and WRF-Chem input formats is available at bitbucket.org/ctessum/emcnv/.

Additional Data. Additional supporting data files are available:

- Dataset S1: A Microsoft Excel file containing emissions amounts disaggregated by life cycle stage for each scenario. For more information on emissions, refer to Tessum et al. (1).
- DatasetS2.pdf: Maps of annual average ground-level concentrations of $PM_{2.5}$, O_3, PM_{10}, NO_x, HCHO, NH_3, particulate SO_4, particulate NH_4, particulate NO_3, organic aerosol, elemental carbon aerosol, particle number, and CO; maps of annual average daily peak O_3 concentrations; and maps of $PM_{2.5}$ and O_3 concentrations animated by month of year, day of week, and hour of day for the baseline simulation and each scenario. A PDF viewer that allows embedded Java-Script, such as Adobe Acrobat, is required to view the animations. Available in an external repository at dx.doi.org/10.13020/D6159V.
- VideoS1.mp4: A video showing temporal variation in $PM_{2.5}$ concentrations attributable to each scenario. Available in an external repository at dx.doi.org/10.13020/D6159V.
- VideoS2.mp4: A video showing temporal variation in O_3 concentrations attributable to each scenario. Available in an external repository at dx.doi.org/10.13020/D6159V.

1. Tessum CW, Marshall JD, Hill JD (2012) A spatially and temporally explicit life cycle inventory of air pollutants from gasoline and ethanol in the United States. *Environ Sci Technol* 46(20):11408–11417.
2. US Environmental Protection Agency (2013) *Fugitive Dust from Mining and Quarrying (2325000000)*. Available at ftp://ftp.epa.gov/EmisInventory. Accessed August 12, 2013.
3. Xstrata Coal (2012) *Ravensworth Underground Mine—Coal Mine Particulate Matter Control Best Management Practice Determination*. Available at www.xstratacoalravensworth.com. au/EN/RavensworthUndergroundMine/Publications/Mt plans and programs/RUM %20Coal%20Mine%20PM%20BMP%20Determination.pdf. Accessed September 26, 2013.
4. Plevin RJ, O'Hare M, Jones AD, Torn MS, Gibbs HK (2010) Greenhouse gas emissions from biofuels' indirect land use change are uncertain but may be much greater than previously estimated. *Environ Sci Technol* 44(21):8015–8021.
5. Brandt AR, et al. (2014) Energy and environment. Methane leaks from North American natural gas systems. *Science* 343(6172):733–735.
6. Michalek JJ, et al. (2011) Valuation of plug-in vehicle life-cycle air emissions and oil displacement benefits. *Proc Natl Acad Sci USA* 108(40):16554–16558.
7. Argonne National Laboratory (2012) *Summary of Expansions and Revisions in GREET2_2012 Version*. Available at greet.es.anl.gov/files/greet2-2012-memo. Accessed November 24, 2014.
8. Edelstein BDL (2010) *US Geological Survey 2010 Minerals Yearbook: Copper* (US Geological Survey, Reston, VA). Available at minerals.usgs.gov/minerals/pubs/commodity/copper/myb1-2010-coppe.pdf. Accessed January 28, 2013.

Fig. S1. April to September average daily peak O_3 concentrations. (A) Year 2005 baseline modeled concentrations. (B–L) Increase in concentration above the baseline attributable to replacement of 10% of year 2020 vehicle use with the given technology. (M) Increase in concentration attributable to EV battery manufacturing. Color scales contain a discontinuity at the 99th percentile of emissions. Abbreviations: CNG, compressed natural gas vehicle; EV, electric vehicle; WWS, wind, water, or solar.

	CH_4	CO	CO_2	N_2O	NH_3	NO_x	PM_{10}	$PM_{2.5}$	SO_x	VOC
Gasoline	96%	100%	97%	99%	100%	68%	86%	81%	55%	99%
Gasoline hybrid	96%	100%	97%	99%	100%	69%	87%	82%	55%	99%
Diesel	96%	98%	98%	99%	100%	71%	87%	83%	54%	97%
CNG	99%	100%	97%	99%	100%	89%	88%	92%	89%	99%
Corn grain ethanol	97%	100%	94%	96%	99%	93%	96%	94%	71%	97%
Corn stover ethanol	123%	100%	115%	95%	98%	97%	86%	91%	190%	97%
EV grid average	100%	98%	100%	100%	100%	98%	100%	100%	100%	99%
EV coal	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%
EV natural gas	99%	96%	99%	99%	80%	93%	98%	98%	98%	98%
EV corn stover	86%	99%	76%	97%	97%	98%	97%	97%	90%	87%
EV WWS	X	X	X	X	X	X	100%	100%	X	X
EV battery production	53%	59%	64%	70%	80%	52%	75%	68%	63%	43%

Fig. S2. Fractions of emissions from each scenario that occur within the spatial modeling domain. Boxes marked with "X" indicate that total emissions are zero. Emissions outside of spatial modeling domain are not included in the above analyses.

Fig. S3. Air pollution damages based on air quality modeling for the month of September for the baseline scenarios and sensitivity analyses where battery production impacts are doubled ("Doubled battery impacts"), where coal mining and cleaning emissions factors were updated to a recently published value ("Coal mining EF"), and where only emissions from vehicle tailpipes or electrical generation units are considered ("Tailpipe or EGU only"). The numbers at the base of each bar are rank orders where number 1 has the lowest impacts and number 11 has the highest impacts of all of the scenarios. Climate change and air pollution impacts of battery production are added to the EV scenarios assuming effects are additive. Abbreviations: CNG, compressed natural gas vehicle; WWS, wind, water, or solar.
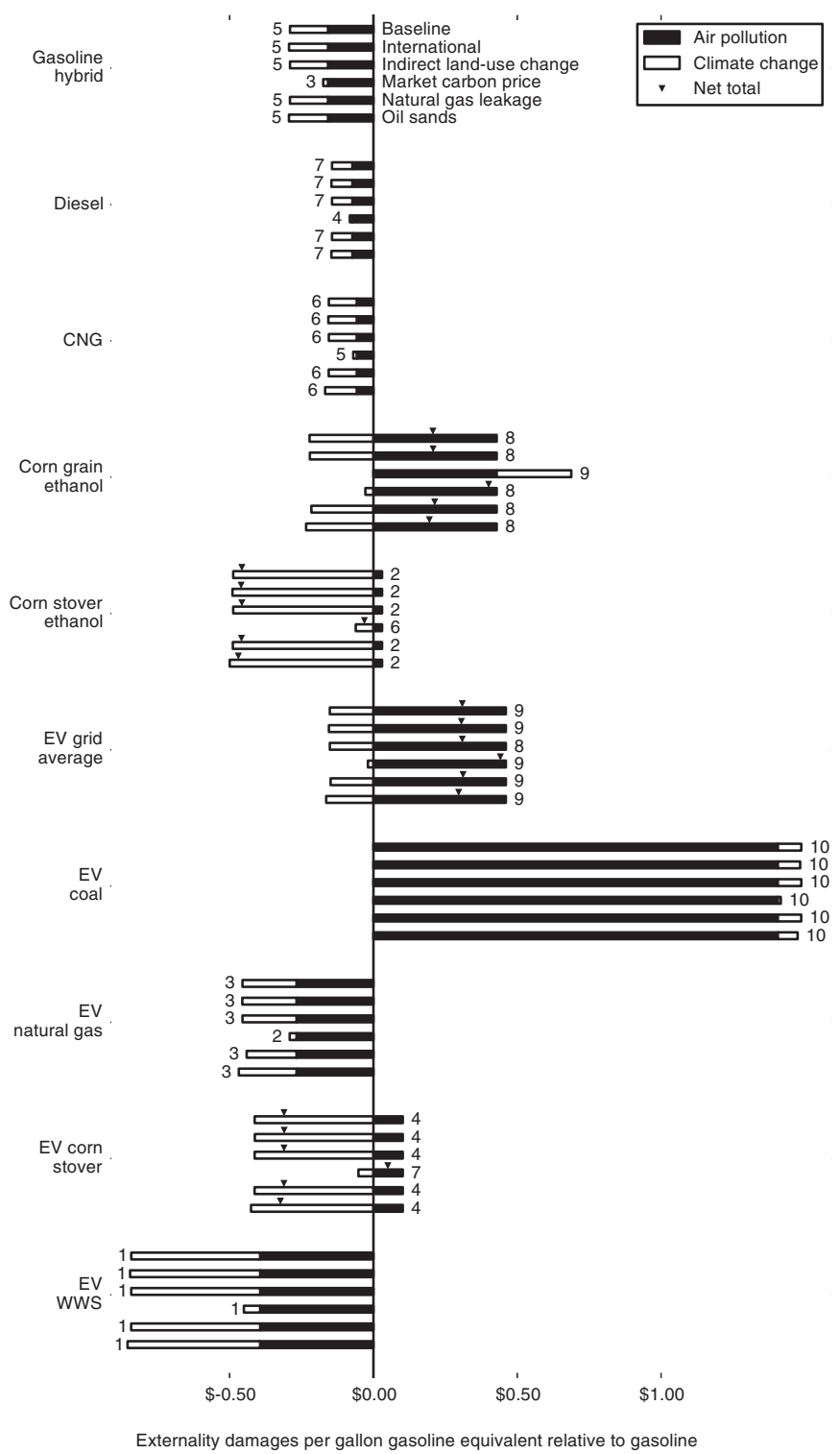
Fig. S4. Annual air pollution and climate change externalities attributable to each scenario relative to the gasoline scenario ("Baseline") and sensitivity analyses assessing the impacts of (*i*) "Indirect land-use change": including indirect land-use change emissions in the corn ethanol scenario; (*ii*) "International": including climate impacts of emissions outside of the United States; (*iii*) "Market carbon price": using a market-based carbon prices rather than the mitigation-based price used in the main analysis; (*iv*) "Natural gas leakage": assuming increased leakage of methane during natural gas extraction; and (*v*) "Oil sands": assuming 21% of crude oil comes from oil sands as opposed to the baseline assumption of 0%. The numbers at the end of each bar are rank orders where number 1 has the lowest impacts and number 10 has the highest impacts of all of the scenarios. Impacts from the gasoline scenario equal zero on this plot. Climate change and air pollution impacts of battery production are added to the EV scenarios assuming effects are additive. Abbreviations: CNG, compressed natural gas vehicle; WWS, wind, water, or solar.

Fig. S5. Impact of grid resolution on apparent health impacts for two scenarios.

Fig. S6. Average percent differences in number of deaths among all scenarios when only considering air quality modeling results from one month compared with all 12 months.

Table S1. Results of previous studies of air quality impacts from alternative transportation fuels and technologies

Article	Result	Notes	Peer-reviewed journal?
Alhajeri et al. (1)	Seventeen percent plug-in hybrid EV (PHEV) adoption leads to greater decreases in O_3 than 100% biofuel (E85) adoption.	Use detailed photochemical model, but only consider vehicle tailpipe and EGU emissions, and only estimate impacts in Austin, TX.	Yes
Boureima et al. (2)	Battery EVs greatly decrease air quality impacts compared with gasoline or hybrid vehicles.	Full life cycle analysis including battery production but does not include any spatial information and uses generalized emissions impact functions. Electric generation mix is Belgium average.	Yes
Brinkman et al. (3)	PHEVs decrease O_3 concentrations compared with gasoline vehicles.	Use detailed photochemical model, but only consider vehicle tailpipe and EGU emissions, and only estimate impacts in Denver, CO.	Yes
Cook et al. (4)	Increased ethanol use in the United States will increase O_3 concentrations in most areas, but decrease concentrations in some highly populated areas with poor air quality.	Full life cycle analysis with spatially explicit emissions, but the degree of spatial disaggregation is not clear. Impacts on $PM_{2.5}$ concentrations are not reported. Air quality model uses two separate 12-km resolution domains, each covering half of the United States.	Yes
EPA (5)	Standard mandating biofuel (both corn grain-based and cellulosic) production and consumption will cause 35–85 cases of adult $PM_{2.5}$ mortality and 36–160 cases of adult O_3 mortality compared with business as usual.	Use detailed air quality model for contiguous United States with life cycle inventory, and spatial data are included in the life cycle inventory. Impacts of corn grain and cellulosic ethanol are not reported separately.	No
EPRI (6)	PHEV adoption decreases O_3 and $PM_{2.5}$ levels compared with business-as-usual in almost all urban areas.	Use detailed photochemical model for contiguous United States, but only consider tailpipe, EGU, and petroleum supply chain emissions. Assume no marginal SO_x or NO_x emissions from EGUs. Air quality model uses 36-km spatial resolution.	No
Hill et al. (7)	$PM_{2.5}$ impacts from corn ethanol are ~60% greater than from gasoline, impacts from cellulosic ethanol are slightly better than from gasoline, and $PM_{2.5}$ impacts are larger than GHG impacts.	Full life cycle analysis at county-level spatial resolution for contiguous United States, with reduced-form air quality model.	Yes
Jacobson (8)	Ethanol vehicles cause increased O_3-related mortalities compared with business-as-usual.	Use detailed photochemical model for contiguous United States, but only consider tailpipe emissions. Air quality model uses 0.5 by 0.75° (~50 km × 75 km) spatial resolution.	Yes
NRC (9)	For year 2030, corn ethanol causes similar air quality impacts to gasoline; cellulosic ethanol, diesel vehicles, and compressed natural gas vehicles cause decreased impacts; EVs cause increased impacts.	Use reduced-form air quality model with full life cycle emissions inventory. Emissions inventory and air quality model have county-level spatial resolution.	No
Michalek et al. (10)	Using "base case" assumptions, EVs do not improve $PM_{2.5}$ and O_3 air quality impacts compared with gasoline, owing largely to emissions from battery production.	Use reduced-form air quality model with full life cycle emissions inventory. Emissions inventory and air quality model have county-level spatial resolution.	Yes
Thompson et al. (11)	PHEVs decrease O_3 concentrations compared with gasoline vehicles.	Use detailed photochemical model, but only consider vehicle tailpipe and EGU emissions, and only estimate impacts in Pennsylvania, New Jersey, and Maryland.	Yes
Thompson et al. (12)	PHEVs decrease O_3 concentrations compared with gasoline vehicles.	Use detailed photochemical model, but only consider vehicle tailpipe and EGU emissions, and only estimate impacts in Texas.	Yes

1. Alhajeri NS, McDonald-Buller EC, Allen DT (2011) Comparisons of air quality impacts of fleet electrification and increased use of biofuels. *Environ Res Lett* 6(2):024011.
2. Boureima F-S, et al. (2009) Comparative LCA of electric, hybrid, LPG and gasoline cars in Belgian context. *World Elec Vehicle J* 3:1–8.
3. Brinkman GL, Denholm P, Hannigan MP, Milford JB (2010) Effects of plug-in hybrid electric vehicles on ozone concentrations in Colorado. *Environ Sci Technol* 44(16):6256–6262.
4. Cook R, et al. (2010) Air quality impacts of increased use of ethanol under the United States' Energy Independence and Security Act. *Atmos Environ* 45(40):7714–7724.
5. US Environmental Protection Agency (2010) Regulation of fuels and fuel additives: Changes to renewable fuel standard program; Final Rule. 75. *Federal Register* 58 (2010), pp 14670–14904.
6. Electric Power Research Institute (2007) *Environmental Assessment of Plug-In Hybrid Electric Vehicles, Volume 2: United States Air Quality Analysis Based on AEO-2006 Assumptions for 2030*. Available at www.epri.com/abstracts/Pages/ProductAbstract.aspx?ProductId=000000000001015326. Accessed November 24, 2014.
7. Hill J, et al. (2009) Climate change and health costs of air emissions from biofuels and gasoline. *Proc Natl Acad Sci USA* 106(6):2077–2082.
8. Jacobson MZ (2007) Effects of ethanol (E85) versus gasoline vehicles on cancer and mortality in the United States. *Environ Sci Technol* 41(11):4150–4157.
9. National Research Council (2009) *Hidden Costs of Energy: Unpriced Consequences of Energy Production and Use*. Available at www.nap.edu/catalog.php?record_id=12794. Accessed November 24, 2014.
10. Michalek JJ, et al. (2011) Valuation of plug-in vehicle life-cycle air emissions and oil displacement benefits. *Proc Natl Acad Sci USA* 108(40):16554–16558.
11. Thompson T, Webber M, Allen DT (2009) Air quality impacts of using overnight electricity generation to charge plug-in hybrid electric vehicles for daytime use. *Environ Res Lett* 4(1):014002.
12. Thompson TM, King CW, Allen DT, Webber ME (2011) Air quality impacts of plug-in hybrid electric vehicles in Texas: Evaluating three battery charging scenarios. *Environ Res Lett* 6(2):024004.

Table S2. Fractions of processes related to battery production that occur outside of the spatial modeling domain

Process	Percent excluded, %	Data source, ref.
Steel production	9	1
$LiMn_2O_4$	100	2
Graphite production	100	1
Copper production	35	1
Aluminum production	13	1
$LiPF_6$	100	2

The spatial modeling domain includes the continental United States and surrounding waters. Processes not included in this table are assumed occur 100% inside the spatial modeling domain. Refer to Dataset S1 for total emissions for each of these processes after the excluded fractions have been removed.

1. US Geological Survey (2012) *Mineral Commodity Summaries 2012*. Available at minerals.usgs.gov/minerals/pubs/mcs/. Accessed January 28, 2012.
2. Wang M, Burnham A, Wu Y (2012) *The Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation Model v2_2012*. Available at greet.es.anl.gov/. Accessed November 24, 2014.

Other Supporting Information Files

Dataset S1 (XLS)