

Climate change and health costs of air emissions from biofuels and gasoline

Jason Hill^{a,b,1}, Stephen Polasky^{a,b}, Erik Nelson^c, David Tilman^{b,1}, Hong Huo^d, Lindsay Ludwig^e, James Neumann^e, Haochi Zheng^a, and Diego Bonta^a

^aDepartment of Applied Economics, ^bDepartment of Ecology, Evolution, and Behavior, University of Minnesota, St. Paul, MN 55108; ^cDepartment of Biology and Natural Capital Project, Woods Institute for the Environment, Stanford University, Stanford, CA 94305; ^dArgonne National Laboratory, Argonne, IL 60439; and ^eIndustrial Economics, Cambridge, MA 02140

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Environmental impacts of energy use can impose large costs on society. We quantify and monetize the life-cycle climate-change and health effects of greenhouse gas (GHG) and fine particulate matter (PM_{2.5}) emissions from gasoline, corn ethanol, and cellulosic ethanol. For each billion ethanol-equivalent gallons of fuel produced and combusted in the US, the combined climate-change and health costs are \$469 million for gasoline, \$472–952 million for corn ethanol depending on biorefinery heat source (natural gas, corn stover, or coal) and technology, but only \$123–208 million for cellulosic ethanol depending on feedstock (prairie biomass, *Miscanthus*, corn stover, or switchgrass). Moreover, a geographically explicit life-cycle analysis that tracks PM_{2.5} emissions and exposure relative to U.S. population shows regional shifts in health costs dependent on fuel production systems. Because cellulosic ethanol can offer health benefits from PM_{2.5} reduction that are of comparable importance to its climate-change benefits from GHG reduction, a shift from gasoline to cellulosic ethanol has greater advantages than previously recognized. These advantages are critically dependent on the source of land used to produce biomass for biofuels, on the magnitude of any indirect land use that may result, and on other as yet unmeasured environmental impacts of biofuels.

fine particulate matter | ethanol | biomass | greenhouse gas | life-cycle analysis

Since the beginning of the automobile era a century ago, oil has had a near monopoly as an energy source for transportation. In 2007, petroleum products accounted for >95% of U.S. transportation energy (1). An intensive search for alternatives is now underway, driven by a variety of concerns including volatile oil prices, increased global demand, reliance on imports from politically unstable regions, and recognition of the harmful effects of GHG and local air pollution. Among these many alternatives are liquid biofuels (2).

Biofuels may offer benefits relative to fossil fuels. Biofuels are a renewable energy source that can be produced domestically from a wide variety of plant materials and wastes. Because plants absorb CO₂ during growth and may increase stores of soil organic carbon (3), biofuels may reduce GHG emissions relative to petroleum-derived fuels. In addition, vehicle tailpipe emissions of many air pollutants harmful to human health may be lower with biofuels (4). Whether biofuels are desirable alternatives to fossil fuels, however, also depends largely on how each is produced (5). Energy inputs, fertilization rates, biomass yields, conversion efficiencies, pollution control technologies, and direct and indirect land-use change all affect the environmental impacts of both biofuels and conventional fuels (6–8).

We compare monetized costs of life-cycle emissions from gasoline and ethanol, including both current and proposed methods of ethanol production in the United States (Fig. 1). We focus on 2 categories of emissions: (i) GHG contributing to global climate change (9) and (ii) fine particulate matter (PM_{2.5}) with a diameter <2.5 μm linked to premature mortality and

other human health impacts (10, 11). Both pollutants are emitted at all stages of gasoline and ethanol production and combustion, including feedstock production (crude oil extraction or biomass cultivation), feedstock transportation, feedstock conversion (refining oil into gasoline or converting biomass into ethanol), fuel distribution, and fuel combustion by end users. GHG and PM_{2.5} constitute 2 of the largest categories of damages associated with energy production and combustion (9, 12, 13), and the transportation sector is a major emitter of both (14, 15).

We consider 3 methods of producing ethanol from corn (using natural gas, coal, or corn stover for process heat at biorefineries) and 4 methods of producing cellulosic ethanol (from corn stover, switchgrass, diverse prairie biomass, or *Miscanthus*). Biorefineries producing corn ethanol purchase electricity from the grid, whereas those producing cellulosic ethanol generate excess electricity for sale to the grid. We base our corn-ethanol analysis on current industry data, with natural gas providing process heat being the industry standard. We also present an advanced corn-ethanol scenario that incorporates potential major improvements in corn production and conversion technologies. Our analyses of cellulosic ethanol, which is not yet in full-scale commercial production, reflect near-term predictions for biomass crop-cultivation practices and biorefinery efficiencies.

We estimate increased emissions from a 3.78 billion liter (1 billion gallon) expansion in U.S. production and combustion of ethanol or an energy-equivalent volume of gasoline (2.49 billion liters or 0.66 billion gallons), approximately equal to the 2006–2007 increase in U.S. gasoline consumption (1). We express all volumes on a gasoline energy-equivalent basis. To make balanced comparisons across alternatives we assume that all production activity occurs in the U.S., and we hold production of all other goods and services in the economy constant. We assume that the additional corn or biomass needed for biofuel production is grown on land currently in the U.S. Conservation Reserve Program (CRP) as perennial grasslands, or, in the case of corn stover, is collected from existing cropland. The expansion of cropland is assumed to occur near currently planned expansion of ethanol facilities. In reality, much of the expansion of U.S. corn production would likely come from shifting acreage of soybeans or other crops (16). If crops in the U.S. are displaced for biofuel production, the resultant global indirect land-use change could result in a substantial carbon debt if native

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¹To whom correspondence may be addressed at: Department of Ecology, Evolution, and Behavior, University of Minnesota, 1987 Upper Buford Circle, St. Paul, MN 55108. E-mail: hill0408@umn.edu or tilman@umn.edu.

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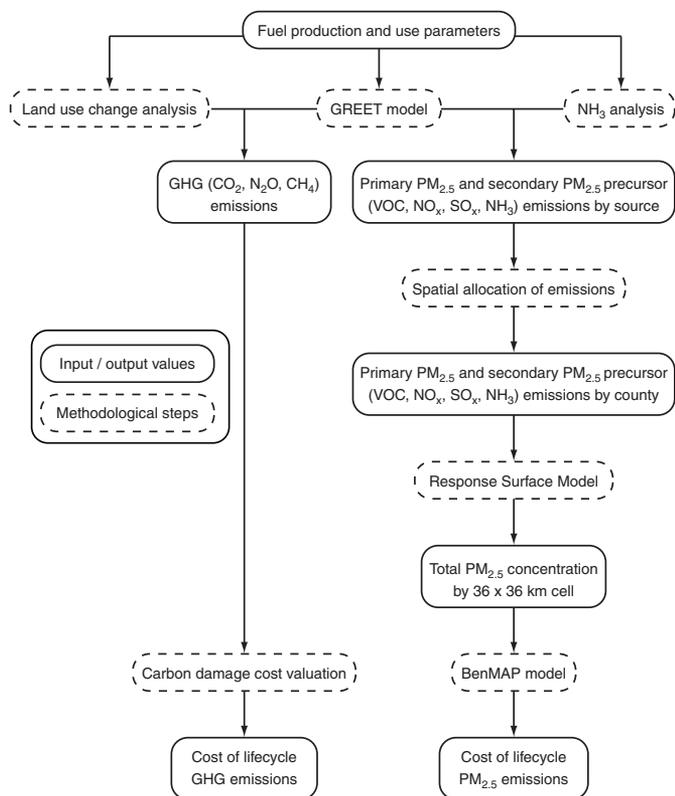


Fig. 1. Flowchart describing methodology for estimating life-cycle GHG and $PM_{2.5}$ costs. Details of various stages are provided in the *Methods* and *SI Methods*.

ecosystems elsewhere in the world are converted to agricultural production (8).

We quantify life-cycle emissions by using the Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) model (17), which tracks long-lived GHG and most $PM_{2.5}$ related emissions. We perform separate analyses to estimate life-cycle NH_3 emissions related to $PM_{2.5}$ formation (supporting information (SI) *Methods*), which are not tracked in GREET, to estimate GHG emissions from land-use change, which is a user-defined parameter in GREET (SI *Methods*). GHG emissions (CO_2 , N_2O , and CH_4) occur from fuel production, fuel combustion, land-use change, and related upstream processes. Air emissions of $PM_{2.5}$ occur directly from fuel combustion (primary $PM_{2.5}$) and indirectly from atmospheric reactions involving SO_x , NO_x , NH_3 , and VOCs emitted from stationary, mobile, and area sources (secondary $PM_{2.5}$).

We generate a monetary cost for GHG emissions based on independent estimates of carbon mitigation costs, carbon market prices, and the social cost of carbon. Carbon mitigation (capture and storage) costs for an integrated gasification combined-cycle electricity generating plant are \$92–\$147 $Mg^{-1} C$ (18); we use the midpoint of this range, \$120 $Mg^{-1} C$. This estimate is in the range of recent prices on the European carbon market [€23–28 $Mg^{-1} CO_2$, or U.S. \$133–\$162 $Mg^{-1} C$ at an exchange rate of \$1.58 €^{-1} in July 2008, and €18–23 $Mg^{-1} CO_2$, or U.S. \$88–\$112 $Mg^{-1} C$ at an exchange rate of \$1.33 €^{-1} in October 2008] (molecular weight ratio of $CO_2:C = 44:12$). It is also within the range of estimates for the “social cost of carbon,” which represents the expected future damages from enhanced climate change (e.g., damages from sea level rise, increased storm intensity, and crop losses from more intensive drought). The mean estimate of the social cost of carbon from peer-reviewed studies was \$43 $Mg^{-1} C$ (19), although there was large variation in the estimates from

below \$0 to $> \$300 Mg^{-1} C$. The large variation in estimates occurs largely because of disagreement on how to weigh future costs and benefits relative to the present (“discounting”) and how to weigh costs and benefits accruing to poor and rich countries (“equity weights”). In addition, there are large uncertainties about climate forcing impacts and future human adaptability.

Unlike long-lived GHG emissions, which globally mix in the atmosphere, the formation and health effects of $PM_{2.5}$ are regional and cannot be accurately determined without considering the spatial patterns of emissions relative to population density (20). We estimate $PM_{2.5}$ related emissions for each U.S. county (SI *Methods*) and use the Response Surface Model (RSM) developed by the U.S. EPA to determine changes in $PM_{2.5}$ levels in each of 6,358 grid cells ($36 km \times 36 km$) covering the continental U.S. (SI *Methods*). Our inclusion of fine-scale spatial patterning of emissions and their movement allows quantification of health impacts of exposure to $PM_{2.5}$ across rural, suburban, and urban areas that differ in emissions and population densities. For example, ethanol production, which occurs largely in rural areas, may have lower impacts on human health per unit of $PM_{2.5}$ emissions than gasoline refining, which occurs largely near urban areas. We use the U.S. EPA’s Environmental Benefits Mapping and Analysis Program (BenMAP) to estimate the human health impacts of increased $PM_{2.5}$ levels and the resultant costs associated with greater exposure to $PM_{2.5}$ (SI *Methods*).

Results

Our results show that the proposed methods of producing cellulosic ethanol we consider have lower GHG emissions than corn ethanol or gasoline (Fig. 24, Table S1), consistent with published findings (21, 22). For cellulosic ethanol produced from perennial plants grown on former CRP lands, higher biomass yields (*Miscanthus* vs. switchgrass) and lower nitrogen fertilizer inputs (diverse prairie biomass vs. switchgrass) result in lower GHG emissions. Whether corn ethanol has lower life-cycle GHG emissions than gasoline depends on biorefinery heat source, assumptions about technology, and land-use change. Before the effects of land-use change on GHG emissions are included, our analyses show that corn ethanol produced by using natural gas has lower GHG emissions than gasoline, but this benefit is lost once carbon emissions from converting CRP grassland to corn cultivation are included. If potential advances in production efficiency at the farm and biorefinery are assumed, GHG emissions from corn ethanol produced by using natural gas are lower than those of gasoline, even with land-use change included. Corn ethanol produced by using corn stover at the biorefinery has lower GHG emissions than gasoline, whereas coal use at the biorefinery results in higher GHG emissions regardless of land-use change assumptions. If C is valued at \$120 Mg^{-1} , the societal climate-change cost from production and consumption of gasoline is \$0.10 L^{-1} (\$0.37 gal^{-1}), between \$0.08–\$0.14 L^{-1} (\$0.31 and \$0.52 gal^{-1}) for corn ethanol, but only between \$0.01–\$0.02 L^{-1} (\$0.03 and \$0.09 gal^{-1}) for cellulosic ethanol (Table S2).

Although recent attention has focused on GHG emissions, the consequences of criteria air pollution from fuel production and combustion are equally important. Our spatially explicit analyses show that cellulosic ethanol has the lowest $PM_{2.5}$ costs of any fuel type analyzed (Fig. 2B and Table S3). Corn ethanol, regardless of whether a biorefinery generates process heat from natural gas, coal, or corn stover, has higher health costs from $PM_{2.5}$ than gasoline. The $PM_{2.5}$ health costs from gasoline are \$0.09 L^{-1} (\$0.34 gal^{-1}) and from ethanol range from \$0.04 L^{-1} (\$0.16 gal^{-1}) for cellulosic ethanol from prairie biomass to \$0.24 L^{-1} (\$0.93 gal^{-1}) for corn ethanol with coal for process heat (Table S4). Besides differences in overall magnitudes, the regional impacts of $PM_{2.5}$ emissions also differ (Fig. 3). Additional corn

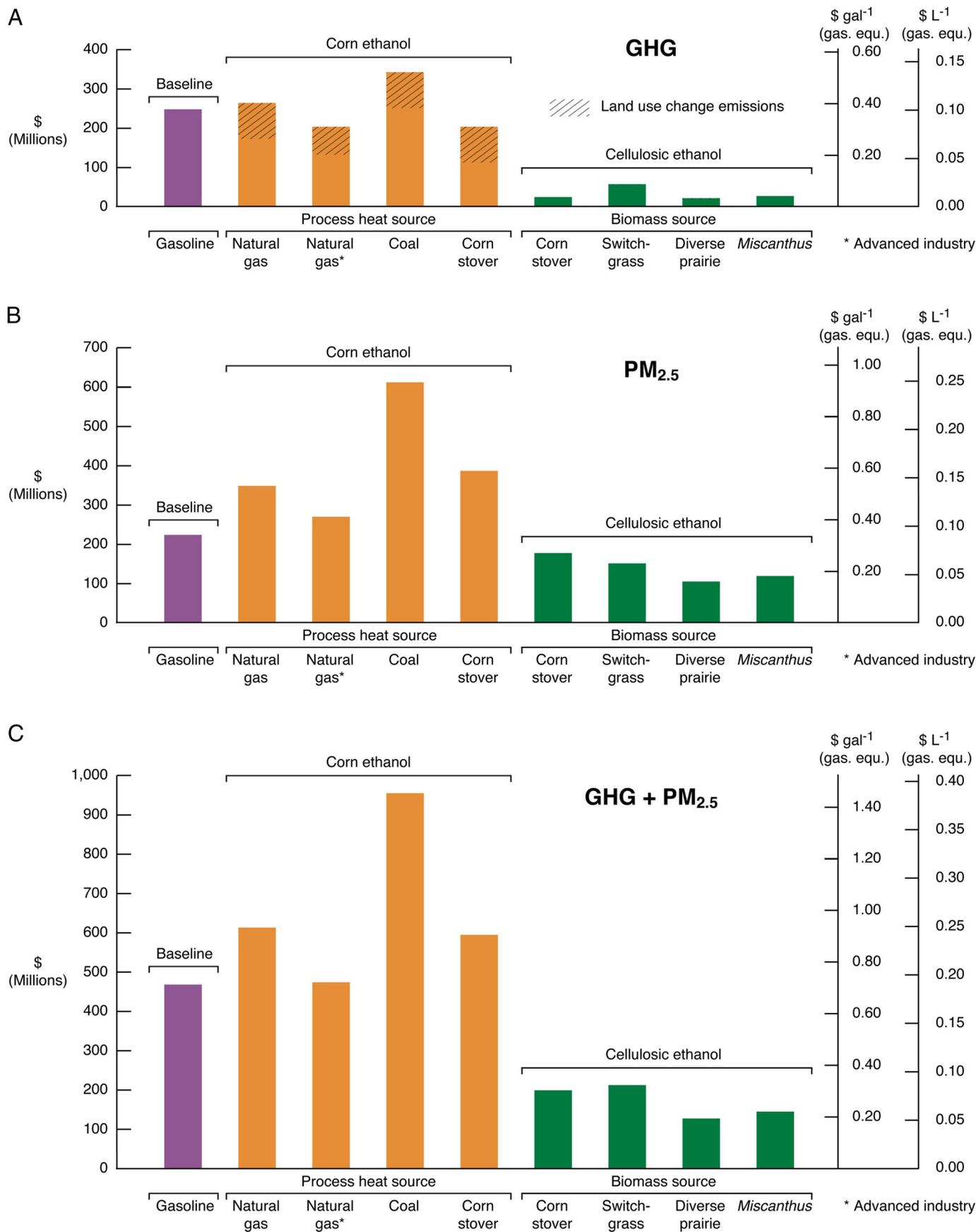


Fig. 2. Costs of GHG (A) and PM_{2.5} (B) emissions. Per liter and per gallon estimates are shown alongside total costs arising from production of an additional billion gallons of ethanol or an energy-equivalent volume of gasoline. (C) Combined costs of GHG and PM_{2.5}.

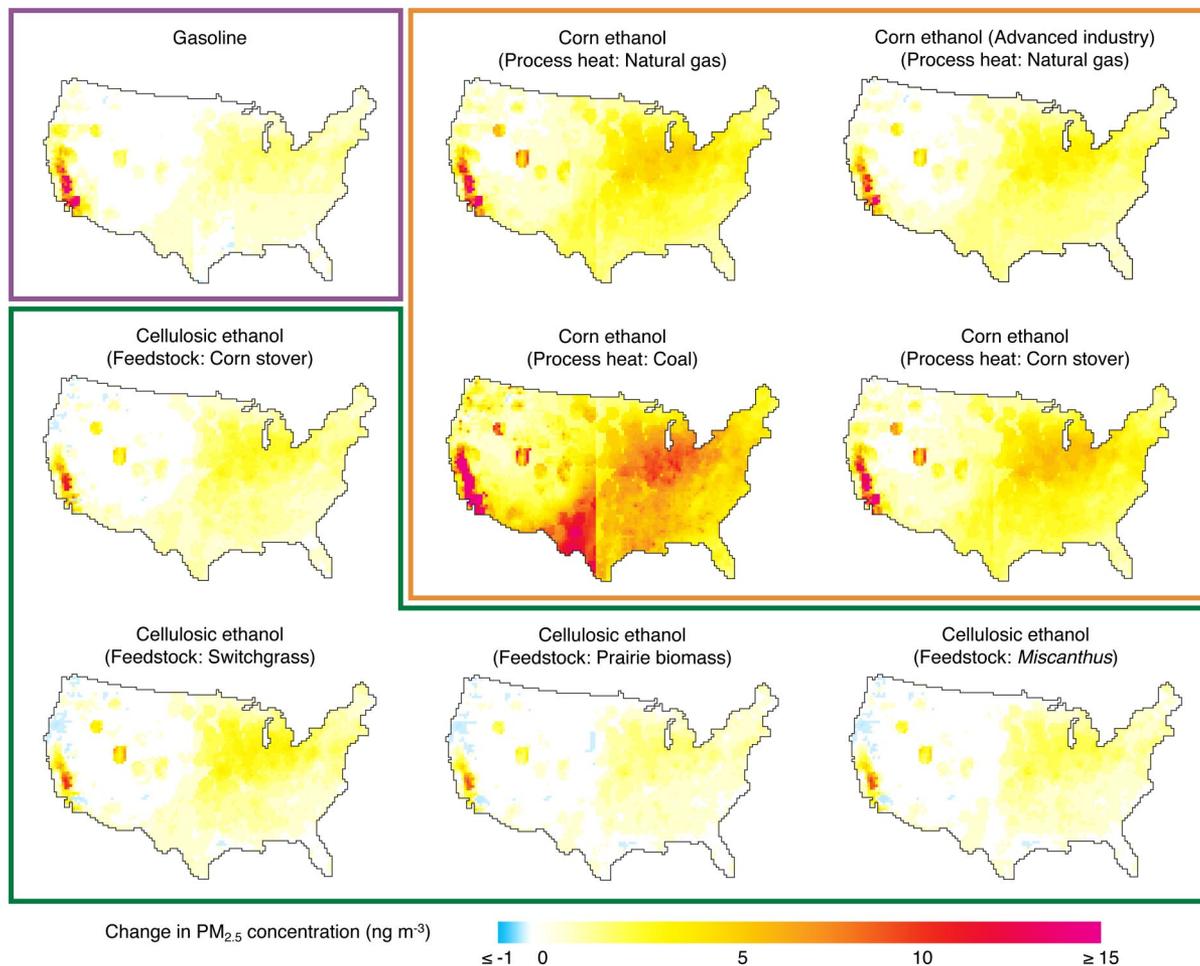


Fig. 3. Change in average annual atmospheric $PM_{2.5}$ concentration from producing and combusting an additional billion gallons of ethanol or an energy-equivalent volume of gasoline.

production increases $PM_{2.5}$ health impacts in the Midwest Corn Belt. Growing perennial biomass crops for cellulosic ethanol in this same region results in lower $PM_{2.5}$ levels than corn ethanol because less fossil fuel and fertilizer are required. Compared with gasoline and corn ethanol, our analysis predicts that cellulosic ethanol also offers improved air quality on the West Coast largely because (i) SO_x emissions from fertilizer production for cellulosic ethanol are lower than those for corn ethanol and SO_x emissions from refining oil into gasoline and (ii) cellulosic biorefineries on the West Coast generate excess electricity, which displaces some coal-based electricity and its $PM_{2.5}$ -related emissions.

Discussion

Our analyses show that air emissions from the production and combustion of liquid transportation fuels impose substantial costs on society. At $\$120 \text{ Mg}^{-1} \text{ C}$ and mean published $PM_{2.5}$ health costs, the total climate-change and health costs of life-cycle GHG and $PM_{2.5}$ emissions for an increase of 1 billion gallons of corn ethanol are $\$614$ million with natural gas for biorefinery process heat and current technology, $\$589$ million with stover, $\$952$ million with coal, and $\$472$ with natural gas and advanced technology on both the farm and biorefinery (Fig. 2C). Total climate-change and health costs for the same volume of cellulosic ethanol from a variety of biomass feedstocks are only $\$123$ – $\$208$ million, and for an energy-equivalent amount of gasoline (0.66 billion gallons) are $\$469$ million. Corn ethanol

fares poorly relative to alternatives because it requires, per unit of fuel produced, more fossil fuel and fertilizer inputs that emit large amounts of GHG and $PM_{2.5}$. Corn-ethanol emissions would be improved if the hypothesized advances in technology were to occur (i.e., reduced fertilizer inputs, increased yields on farm, and improved conversion), making combined environmental costs from corn ethanol by using natural gas for process heat similar to gasoline. Compared with corn ethanol, cellulosic ethanol from corn stover or perennial crops requires lower inputs and has lower emissions at the biorefinery because lignin combustion provides process heat and power, thereby displacing fossil fuel inputs and electricity production.

Estimates of climate-change and health costs are subject to uncertainty in both biophysical and economic parameters (Table S3 and Table S4). Much of the variance is because of a lack of consensus on the economic values of both climate stabilization and human health. While this affects the dollar value of cost estimates, it does not change the relative ranking among fuel alternatives.

Additional factors also affect the societal evaluation of alternative fuels, including their impacts on energy independence and security, economic development, and food production. There are also other environmental impacts not considered here (e.g., hazardous air emissions, acidification, soil fertility, erosion, sedimentation, habitat quality and quantity, water usage, fertilizer and pesticide contamination, and petroleum spills) (23, 24). For climate change, additional factors not considered here include changes in albedo from land-use change and the effects

of “black carbon” created during fuel combustion (25). Also, in addition to contributing to secondary PM_{2.5} formation, air emissions of NO_x and VOCs cause formation of ground-level ozone, which too is linked to premature mortality and other health concerns (26). As with PM_{2.5}, health costs of ozone are spatially dependent. Estimating the cost to society of many of these additional environmental impacts of alternative fuels will require spatially and even temporally explicit life-cycle analysis.

The energy industry is undergoing rapid technological transformations that may change the cost equation. Environmental costs per unit of ethanol decline with higher biomass yield, lower fertilizer and fuel inputs into biomass production, and improvements in biomass to biofuel conversion efficiencies (27). When particularly favorable improvements in technology over the next decade are assumed, the costs of GHG and PM_{2.5} emissions from corn ethanol could be approximately equal to, but unlikely less than, those of conventional gasoline. Cellulosic ethanol holds the promise of yet greater environmental benefits, but economical ways of producing it must first be discovered. New biofuel feedstocks (e.g., algae, *Jatropha*, hybrid poplar, new varieties of switchgrass, and better multispecies plant mixtures) may also yield substantial improvements. Conversely, other ways of increasing biofuel production may increase air pollutant emissions, such as shifting from traditional corn–soybean crop rotations to continuous corn production, which has greater nitrogen fertilizer requirements and returns lower yields. Although gasoline production is a mature technology, a shift from crude oil to oil sands or coal-to-liquids technology would greatly increase emissions unless accompanied by simultaneous improvements in abatement technology. Consideration should also be given to improved emissions controls (28) and increases in fuel efficiency and fuel conservation (29) that would reduce the need for increased fuel supplies.

We estimated the costs associated with increased levels of GHG and PM_{2.5} resulting from a 3.78 billion liter (1 billion gallon) expansion in fuel production, which is a relatively small change. The Energy Security and Independence Act of 2007 calls for biofuel production to increase to 36 billion gallons by 2022 from 6.5 billion gallons in 2007. Such a large increase could change the estimated cost of GHG and PM_{2.5} emissions per gallon as the marginal damage to health is greater with worsening air quality, and changes in market prices of biofuel feedstocks could alter crop rotations and cultivation methods, encourage crop production to expand into less fertile lands, and cause native ecosystems to be converted to agriculture either directly or indirectly.

Our analyses show that the debate over whether substituting biofuels for fossil fuels benefits or harms the environment needs to be expanded beyond GHG emissions to include a broad array of environmental quality dimensions. PM_{2.5} emissions impose costs on society of similar magnitude to GHG emissions, and as such, the benefits of shifting from gasoline and the current generation of food- and feed-based biofuels to next-generation cellulosic biofuels are approximately twice as large as previously thought, as long as the carbon debt from land-use change is minimal. Other environmental advantages of properly produced cellulosic biofuels (e.g., lower emissions of ozone precursors and reduced pesticide and nitrate loading of surface and groundwater) may make the economic benefits to society of this transition greater still.

Increasing liquid fuel production is not the only approach to meeting society’s growing transportation energy needs. Technological and behavioral solutions include improved vehicle efficiency, public transportation, redesign of urban landscapes, and hybrid, plug-in electric, natural gas, and hydrogen vehicles. In total, the considerable societal costs of GHG and PM_{2.5} emissions, and of other effects not yet quantified, should be given full weight in policy choices among energy sources, efficiency, and conservation.

Methods

Fuel Production and Use. We assume the corn needed to produce an additional billion gallon of ethanol is grown on converted CRP land and that the land area converted in each county is proportional to each county’s share of U.S. corn production in 2005. We exclude CRP land classified as having high soil conservation or wildlife benefit (e.g., Filter Strips, Rare and Declining Habitat, and Wetland Restoration). CRP conversion is assumed to occur on less sensitive enrollments (e.g., Introduced Grasses, Native Grasses, and Established Grass). The average yield on converted CRP land is estimated at 9,095 kg ha⁻¹, which is 8.3% lower than the GREET default U.S. average value of 9,917 kg ha⁻¹. This difference reflects lower corn yields in counties with CRP land available for conversion and not reduced productivity of CRP land compared with cropland currently farmed in that county, although such reduced productivity is likely. GREET default values are used for herbicide, insecticide, agricultural lime, and fertilizer application rates.

As with corn, biomass production is assumed to occur on compatible CRP land. Because of transportation cost constraints, we assume land conversion occurs in counties nearest to planned biorefineries. We use data on plant locations and production capacities of planned facilities as reported by the biofuel industry in January 2008 (30). The average switchgrass yield is assumed to be 7.1 Mg ha⁻¹, as has been achieved on farm-scale field trials experiments in the eastern Great Plains (31). We use a nitrogen fertilizer application rate of 74 kg ha⁻¹ (31). We assume comparable biomass yields from restored diverse prairie on land of identical quality, but with nitrogen supplied by legumes instead of synthetic fertilizers. Such yields are consistent with published measurements of prairie productivity (7). *Miscanthus* yields are assumed to be 21.3 Mg ha⁻¹, triple that of switchgrass and diverse prairie. This yield is nearly identical to an average *Miscanthus* yield of 22 Mg ha⁻¹ taken from 97 observations of small-plot trials (32). As with switchgrass, an annual nitrogen fertilization rate of 74 kg ha⁻¹ is used, which is nearly identical to an average *Miscanthus* agronomic practice application rate of 75 kg ha⁻¹ (33).

Unlike corn, the 3 biomass crops we consider are not currently widely grown for harvest on a large scale under typical farm conditions. It is worth stressing that the yields and nitrogen fertilizer rates used here for the 3 biomass crops are estimates based on data from field trials and small-plot trials. Actual yields and fertilizer application rates may differ when produced on large scales with on-farm conditions. Also, yields may improve through time with increased experience in growing these crops or with improved varieties. At present we lack fine-scale mapping of productivity potential within the U.S. to differentiate biomass yield by county as we do with corn. Also, little is known about yields of these 3 crops on comparable soil and climate types. As structured in this analysis, the results of the 3 biomass crops can be interpreted as a sensitivity analysis to show the effect of decreasing fertilizer input (prairie) or increasing biomass yield (*Miscanthus*) versus a base case (switchgrass).

Although corn stover production does not require conversion of CRP land, additional nutrient inputs are needed to maintain soil fertility and grain yield on existing cropland. GREET default application rates of an additional 15.9 kg ha⁻¹ of N, 8.2 kg ha⁻¹ of P, and 41.8 kg ha⁻¹ of K are assumed. The default stover removal rate of 50% is used, although the fraction of stover that can be harvested while maintaining soil organic carbon and moisture levels may vary across soil types, land grading, and average rainfall amounts (34). This variation is not accounted for in this study. As with dedicated biomass crop production, stover is harvested in counties closest to planned biorefinery expansions.

Ethanol production from corn grain is assumed to occur with dry mill technology yielding 0.405 liter kg⁻¹. We assume a near-future conversion rate of 0.340 liter kg⁻¹ for lignocellulosic biomass to ethanol for lignocellulosic biomass to ethanol (35), which is 10% greater than current demonstrated technology (36) and consistent with other published results (37). Combustion of lignin during cellulosic ethanol production results in cogeneration of electricity, the excess of which is exported to the grid at a rate of 0.544 MJ L⁻¹. Like corn and biomass production, crude oil extraction is assumed to occur within the continental U.S. Increased crude production is allocated to states in proportion to 2005 crude oil production. GREET default values for refining crude oil into gasoline are used. For all fuels, GREET default methods are applied for estimating material and energy coproduct credits. We allocate emissions from transportation of feedstocks, intermediates, and finished products by using a simple rule that half of all transportation emissions occur in the county where transportation originates, and half occur in the county where transportation ends.

For the advanced corn-ethanol scenario, we consider 5 major potential improvements in agricultural and conversion technologies reflecting published projections for the industry in 2020. We assume corn grain yields increase by 20% and nitrogen fertilization rates decrease by 33% to reflect improved breeding and cultivation practices (38, 39). We also assume biorefineries use combined heat and power technologies to decrease thermal

energy use by 19% and purchased electricity use by 79% (40). Finally, we assume process improvements, such as converting corn kernel fiber to fermentable sugars, increase ethanol yields by 10% (40).

We assume that the additional volumes of fuels produced are combusted in counties in proportion to current transportation fuel usage as measured by vehicle miles traveled within a given county. Additional ethanol production is assumed to be combusted as a 10% blend with gasoline (E10). Tailpipe emissions of gasoline and ethanol are modeled by using GREET default values.

Fuel Life-Cycle Emissions. Quantities of GHG and PM_{2.5} related emissions released from various sources are determined for each stage of fuel production and use. For gasoline, the following sources are included:

- Process emissions from exploration and extraction of crude oil
- Electricity generation for use in exploration and extraction of crude oil
- Transportation of crude oil to refineries
- Refinery process emissions
- Electricity generation for use at refineries
- Upstream natural gas and coal emissions (e.g., extraction and mining)
- Distribution of finished product (gasoline)
- Sales and combustion of finished product (gasoline)

For ethanol, the following sources of emissions are included:

- Land-use change
- Process emissions from lime and fertilizer production
- Electricity generation for lime and fertilizer production
- Process emissions from pesticide (herbicide and insecticide) production
- Fossil fuel use on farms
- Electricity generation for farm use
- Soil emissions of N₂O, NO_x, and NH₃ from N fertilizer application
- Transportation of corn or biomass to biorefineries
- Biorefinery process emissions
- Combustion of natural gas, coal, or biomass at biorefineries
- Electricity generation for use at corn-ethanol biorefineries
- Upstream natural gas and coal emissions (e.g., extraction and mining)

- Distribution of finished product (ethanol)
- Sales and combustion of the finished product (ethanol).

Spatial Allocation of Emissions. We use publicly available descriptions of the locations of the emission sources listed above to allocate emissions of primary PM_{2.5} and secondary PM_{2.5} precursor pollutants >310 counties (*SI Methods*). For gasoline, emissions are allocated to counties based on the following spatial data:

- Areas where crude oil is extracted
- Locations of electrical plants providing power for crude oil extraction
- Areas over which crude oil is transported
- Locations of refineries
- Locations of electrical plants providing power to refineries
- Areas from which natural gas is extracted and coal is mined
- Areas over which the finished product (gasoline) is transported
- Areas in which the finished product (gasoline) is sold and combusted

For ethanol, emissions are allocated to counties based on the following spatial data:

- Areas where agricultural lime and N, P, and K fertilizer is produced
- Locations of electrical plants powering lime and N, P, and K production
- Pesticide production facility locations
- Areas farmed for corn and biomass, and their relative productivity
- Locations of electrical plants providing power to farms
- Areas over which corn and biomass are transported
- Locations of biorefineries
- Locations of electrical plants providing power to biorefineries
- Areas from which natural gas is extracted and coal is mined
- Areas over which the finished product (ethanol) is transported
- Areas in which the finished product (ethanol) is sold and combusted

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1. Energy Information Administration (2008) *Monthly Energy Review* (U.S. Department of Energy, Washington, DC), DOE/EIA-0035(2008/03).
2. MacLean H, Lave L (2003) Evaluating automotive fuel/propulsion system technologies. *Prog Energy Combust Sci* 29:1–69.
3. Tilman D, Hill J, Lehman C (2006) Carbon-negative biofuels from low-input high-diversity grassland biomass. *Science* 314:1598–1600.
4. McCormick R (2008) The impact of biodiesel on pollutant emissions and public health. *Inhalation Toxicol* 19:1033–1039.
5. Robertson G, et al. (2008) Sustainable biofuels redux. *Science* 322:49–50.
6. Hill J, Nelson E, Tilman D, Polasky S, Tiffany D (2006) Environmental, economic, and energetic costs and benefits of biodiesel and ethanol biofuels. *Proc Natl Acad Sci USA* 103:11206–11210.
7. Fargione J, Hill J, Tilman D, Polasky S, Hawthorne P (2008) Land clearing and the biofuel carbon debt. *Science* 319:1238–1240.
8. Searchinger T, et al. (2008) Use of U.S. croplands for biofuels increases greenhouse gases through emissions from land-use change. *Science* 319:1238–1240.
9. Intergovernmental Panel on Climate Change (2007) *Climate Change 2007: Synthesis Report* (Intergovernmental Panel on Climate Change, Geneva).
10. Pope C, Dockery D (2006) Health effects of fine particulate air pollution: Lines that connect. *J Air and Waste Manage Assoc* 56:709–742.
11. Baccarelli A, et al. (2008) Exposure to particulate air pollution and risk of deep vein thrombosis. *Arch Intern Med* 168:920–927.
12. U.S. EPA (1999) *The Benefits and Costs of the Clean Air Act 1990 to 2010* (U.S. Environmental Protection Agency, Washington, DC), EPA-410-R-99-001.
13. Peng R, et al. (2008) Coarse particulate matter air pollution and hospital admissions for cardiovascular and respiratory diseases among Medicare patients. *JAMA* 299:2172–2179.
14. Fuglestedt J, et al. (2008) Climate forcing from the transport sectors. *Proc Natl Acad Sci USA* 105:454–458.
15. Abu-Allaban M, Gillies J, Gertler A, Clayton R, Proffitt D (2007) Motor vehicle contributions to ambient PM₁₀ and PM_{2.5} at selected urban areas in the USA. *Environ Monit Assess* 132:155–163.
16. Food and Agricultural Policy Research Institute (2008) *U.S. and World Agricultural Outlook* (Staff Report 08-F5R 1, Ames, IA).
17. Wang M (2007) "GREET" (Argonne National Laboratory, Argonne, IL), ANL/ESD/05-03.
18. Dooley J, et al. (2006) *Carbon Dioxide Capture and Geologic Storage* (Global Energy Technology Strategy Program, College Park, MD).
19. Tol R (2005) The marginal damage costs of carbon dioxide emissions: An assessment of the uncertainties. *Energy Policy* 33:2064–2074.
20. Rabl A, Nathwani J, Pandey M, Hurley F (2007) Improving policy responses to the risk of air pollution. *J Toxicol Env Heal A* 70:316–331.
21. Farrell A, et al. (2006) Ethanol can contribute to energy and environmental goals. *Science* 311:506–508.
22. Plevin R, Mueller S (2008) The effect of CO₂ regulations on the cost of corn ethanol production. *Environ Res Lett* 3, doi:10.1088/1748-9326/3/2/024003.
23. Lavigne A, Powers S (2007) Evaluating fuel ethanol feedstocks from energy policy perspectives: A comparative energy assessment of corn and corn stover. *Energy Policy* 35:5918–5930.
24. Parry I, Walls M, Harrington W (2007) Automobile externalities and policies. *J Econ Lit* 65:373–399.
25. Ramanathan V, Carmichael G (2008) Global and regional climate changes due to black carbon. *Nat Geosci* 1:221–227.
26. National Research Council (2008) *Estimating Mortality Risk Reduction and Economic Benefits from Controlling Ozone Air Pollution* (National Academy of Sciences, Washington, DC).
27. Liska A, Cassman K (2008) Towards standardization of life-cycle metrics for biofuels: Greenhouse gas emissions mitigation and net energy yield. *J Biobased Mater Bioenergy* 2:187–203.
28. Twigg M (2005) Controlling automotive exhaust emissions: Successes and underlying science. *Phil Trans R Soc A* 363:1013–1033.
29. Leigh J, Geraghty E (2008) High gasoline prices and mortality from motor vehicle crashes and air pollution. *J Occupat Environ Med* 50:249–254.
30. Renewable Fuels Association (2008) *Ethanol Biorefinery Locations* (Renewable Fuels Association, Washington, DC).
31. Schmer M, Vogel K, Mitchell R, Perrin R (2008) Net energy of cellulosic ethanol from switchgrass. *Proc Natl Acad Sci USA* 105:464–469.
32. Heaton E, Voight T, Long S (2004) A quantitative review comparing the yields of two candidate C4 perennial biomass crops in relation to nitrogen, temperature, and water. *Biomass Bioenergy* 27:21–30.
33. Khanna M, Dhungana B, Clifton-Brown J (2008) Costs of producing miscanthus and switchgrass for bioenergy in Illinois. *Biomass Bioenergy* 32:482–493.
34. Wilhelm W, Johnson J, Karlen D, Lightle D (2007) Corn stover to sustain soil organic carbon further constrains biomass supply. *Agron J* 99:1665–1667.
35. Sheehan J, et al. (2004) Energy and environmental aspects of using corn stover for fuel ethanol. *J Ind Ecol* 7:117–146.
36. Lynd L, et al. (2008) How biotech can transform biofuels. *Nat Biotech* 26:169–172.
37. Varvel G, Vogel K, Mitchell R, Follett R, Kimble J (2008) Comparison of corn and switchgrass on marginal soils for bioenergy. *Biomass Bioenergy* 32:18–21.
38. Fageria N, Baligar V (2005) Enhancing nitrogen use efficiency in crop plants. *Adv Agron* 88:97–185.
39. Cassman K, Liska A (2007) Food and fuel for all: Realistic or foolish? *Biofpr* 1:18–23.
40. Mueller S (2007) *An analysis of the projected energy use of future dry mill corn ethanol plants (2010-2030)* (Energy Resources Center, University of Illinois, Chicago, IL).