

Supporting Information

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SI Text

S1. Calculation of L_{REF} The values we used for L_{REF} are summarized in Table S1 and were calculated by dividing estimated fuel-cycle methane emissions, multiplied by 1.1, by the sum of the gross production of natural gas (26 Tcf in 2009) and the methane lost at production wells (570 bcf in 2009). [This adjustment accounts for the composition of CH_4 in natural gas, conservatively assuming that each cubic foot of natural gas discharged to the atmosphere contains 90% CH_4 , on average, across the fuel cycle. This value varies by location and by industry segment. The average CH_4 content in U.S. natural gas has been reported to be 79%, 87%, and 93% for the production, processing, and transmission/distribution segments, respectively. See Shires TM, Harrison MR (1996) Methane Emissions from the Natural Gas Industry; Volume 6: Vented and Combustion Source Summary; Appendix A (Gas Research Institute and U.S. EPA). Available at www.epa.gov/gasstar/documents/emissions_report/6_vented.pdf. Combining the CH_4 composition data in Table A-1 of the EPA/GRI report with the relative distribution of EPA's CH_4 emissions data by industry segment, summarized in Table S2, would yield a weighted average of 84%.]

S2. Estimated Emissions from Natural Gas Fuel Cycles. For over a decade, official U.S. estimates of the amount of CH_4 emitted by natural gas systems have been based on dozens of equipment-specific emission factors published in 1996 (1). In 2011, the U.S. EPA adjusted several emission factors from the 1996 report after new data indicated the older values underestimated actual emissions. Specifically, EPA revised emission factors for gas well cleanups, condensate storage tanks, and centrifugal compressors. In addition, EPA added emissions for unconventional gas well completions and workover venting (2).

We estimate CH_4 emissions from natural gas systems largely following the life-cycle analysis performed by P. Jaramillo et al. (3). The main differences are: (i) we use EPA's 2011 emissions estimates (for 2009 calendar year) (4), except where noted; (ii) we include vented CO_2 emissions in addition to CH_4 ; and (iii) we include a portion of the CH_4 emissions from oil wells, based on the fraction of total energy content produced at U.S. oil wells that is due to natural gas. Like Jaramillo et al., our analysis excludes manufacture of materials for natural gas and electric infrastructure, and energy used in preproduction activities at gas and oil wells. One study quantified preproduction activities for shale gas being less than 10% of the emissions from well to city gate (5).

Table S2 summarizes the CH_4 and noncombustion CO_2 emissions used in our analysis. Combustion CO_2 emissions and the CH_4 emissions from oil wells that we assign to the natural gas fuel cycle were derived using parameters summarized in Table S3. Table S4 includes our estimates of total emissions of CH_4 and CO_2 from the production, processing, transmission/storage and distribution per unit of natural gas delivered.

For the power plant scenario, we convert the emissions in Table S4 into emissions per unit of electricity produced assuming a new Combined Cycle power plant with a heat rate of 6,798 Btu/kWh (6). The upstream emissions for power plant scenarios exclude the distribution segment, based on the assumption that most power plants receive their natural gas directly from the transmission system. The resulting values in Table 1 of the main paper were used for all calculations of the radiative forcing from natural gas power plants.

For the transportation scenario (see Table 1 of the main paper), we additionally include emissions from the distribution segment in the estimates of upstream CH_4 and CO_2 . The upstream CO_2 emissions also include an additional 3.4 kg CO_2 /mmBtu to account for electric compression to produce compressed natural gas (CNG), assuming 0.6 kWh per therm (7) and average U.S. power plant emissions of 1.24 lb/kWh (Table S3). In-use methane emissions from a CNG vehicle are assumed to be 20 times the gasoline value (8). CNG refueling emissions are not explicitly included, but have been reported to be insignificant (9). Vehicle emissions of CO_2 from CNG combustion were derived from values in EPA's Greenhouse Gas Inventory (10). As discussed in Section S6, our emission estimates for the fuel cycle of a CNG car appear to be conservative when compared to results using emissions from the widely cited GREET model.

As described in Section S5, we used different emission factors for the comparison of CNG and diesel for heavy-duty vehicles, adapted from a published analysis specific to that category of vehicle.

S3. Estimated Emissions from Coal Fuel Cycle. The scenario modeled for a coal power plant was based on a new supercritical pulverized coal plant with a heat rate of 8,687 Btu/kWh. The emissions were based on a life-cycle analysis performed by the National Energy Technology Laboratory (NETL) (11), except that NETL's emissions estimates were divided by 1.07 to reflect MWh produced (i.e., excluding electric transmission line losses assumed in their analysis) and that we used a range of assumptions about CH_4 emissions from coal mining. NETL's analysis assumed the plant was fueled with underground bituminous coal from a "gassy" mine (360 scf CH_4 per ton coal mined). The power plant scenario analyzed in the main paper (see Table 1) assumed the use of "low-gassy" coal (i.e. from a mine considered to produce less than 71 scf/ton) (12), with only 25% of the upstream CH_4 assumed in NETL's analysis for gassy coal. Such an assumption is also more reflective of CH_4 from surface-mined subbituminous coal, which has relatively low CH_4 emissions (40 scf CH_4 /ton coal or less) (13). To account for the large variability in CH_4 emissions from coal mines, we ran sensitivity tests using revised assumptions described in Section S6.

S4. Estimated Emissions for Gasoline Fuel Cycle. The emissions we used to simulate a gasoline fleet of automobiles are summarized in Table 1 of the main paper. Except for the vehicle CO_2 emissions, our estimates were derived from the baseline life-cycle analysis conducted by NETL in support of EPA's recent review of renewable fuels (14). More specifically, we derived our values from those reported in NETL's spreadsheet model (downloaded from <http://www.netl.doe.gov/energy-analyses/refshelf/PubDetails.aspx?Action=View&PubId=283> on April 27, 2011) with selection of the following user-defined inputs: (i) EPA RFS2 tailpipe values (i.e., the EPA November 2009 Profile under "Vehicle Operation" tab); and (ii) 2007 IPCC GWPs. To be consistent with our calculation of CH_4 emissions using Higher Heating Value (HHV), we converted NETL's values, reported as Lower Heating Values, by multiplying by 0.93. The vehicle CO_2 emission factor was independently calculated using EPA data for the gasoline carbon content per gallon (2.421 kg/gallon) (15) and U.S. Energy Information Administration data for the heat content of conventional gasoline (5.253 mmBtu per Barrel, HHV) (16).

In the transportation scenario, we assumed the efficiency of gasoline and CNG automobiles were equal.

55. Estimated Emissions for Heavy-Duty CNG and Diesel Fuel Cycles. As summarized in Table 1 of main paper, we adapted published emission factors in mg per ton-mile from a paper by Meyer et al. (17) to compare CNG heavy-duty vehicles to diesel. Because Meyer et al.'s emission factors were developed using a version of the GREET model (see Section S6), which did not yet reflect EPA's 2011 revisions for CH₄ emissions from natural gas systems, we doubled the CH₄ emissions reported by Meyer et al. for the upstream CH₄ from the CNG fuel cycle. Other than this adjustment, the rest of the emission factors in Table 1 were taken directly from Meyer et al.

In the calculations for Fig. 2B of the main paper, which use Eq. 7, we assumed the well-to-wheels leakage of natural gas for heavy-duty CNG vehicles was 3.0% of total gas produced, equal to the value for light-duty CNG vehicles (see Table S1). We made this assumption in the absence of data for pump-to-wheels losses for heavy-duty vehicles, comparable to that provided by Lipman and Delucchi for light-duty vehicles (8). The effective value in Meyer et al. for pump-to-wheels methane losses relative to total well-to-wheels CH₄ losses is 5 percent, which is one-fourth of the effective value we derived from Lipman and Delucchi. According to Eq. 7, which is linear with respect to the assumed leak rate, if our value of well-to-wheels leakage for heavy-duty CNG vehicles is too high, then the cross-over leak rates are also too high. Consequently, our calculation of the cross-over leak rate for heavy-duty CNG vehicles compared to diesel are a conservative estimate (even lower leakage than our calculated value of 1% may, in fact, be required to produce climate benefits on all time frames).

56. Sensitivity Tests. CNG vs. gasoline. To test the emissions assumptions in our light-duty automobile scenario, we calculated TWP using emissions factors from two versions of the widely cited GREET model (18). We ran the most recent version of GREET, GREET 1_2011, in its default mode. GREET 1_2011 includes, among other things, an update to the upstream CH₄ emissions in the CNG fuel cycle that reflect EPA's 2011 adjustment to the emissions of CH₄ from natural gas systems. We also ran GREET1.8d.1 in its default mode and with one notable modification. Because the GREET 1.8d.1 model does not reflect EPA's 2011 revisions to the CH₄ emissions from natural gas systems (which roughly amounted to a doubling of CH₄ emissions) we doubled the CH₄ emissions predicted by the GREET1.8d.1 model for its CNG Feedstock and Fuel components. Table S5 summarizes the three sets of emissions from the GREET model that we used. Table S6 summarizes the cross-over point of TWP calculations using emissions in Table 1 of the main paper and TWP calculations using the emissions from the GREET model summarized in Table S5. Our results described in the main paper are in closest agreement with the calculation based on emissions from GREET1.8d.1 with the upstream CH₄ adjustment to the CNG fuel cycle. With the modified GREET1.8d.1 assumptions, the onset of net climate benefits is predicted to be 93 years using the Fleet Conversion TWP (52 years for a pulse TWP). These results can be compared with our results presented in Fig. 1 of the main paper (80 and 46 years, for the Continuous TWP and Pulse TWP, respectively). We note that our results are more favorable to the CNG option than both GREET 1_2011 (the most current version of the model) and GREET1.8d.1, as adjusted, suggesting a possible underestimate of our CNG fuel-cycle emissions, an overestimate of the gasoline fuel-cycle emissions, or both. Our analysis using GREET emissions factors is consistent with the recent analysis by Burnham et al. using the GREET model, which concluded that on a 100-year time horizon, there was no statistically significant difference between the well-to-wheels GHG emissions on kilometer traveled basis of CNG passenger cars and transit buses compared to those using gasoline or diesel fuel (19). Because the results of Burnham et al. are based

on a GWP analysis, they should be compared to our Pulse TWP results presented in Figs. 1 and 2 of the main paper (showing about a 10% reduction and a 2% increase in the cumulative radiative forcing after 100 years for the CNG options relative to the gasoline and diesel options, respectively).

Natural gas vs. coal. To account for the uncertainty in the CH₄ emissions from natural gas and coal systems, and the potential for operators to control emissions, we conducted the following sensitivity analysis.

In addition to using the baseline assumptions, upstream CH₄ in the natural gas fuel cycle was doubled and halved to represent a large range of possible leakage in the natural gas system (EPA reports a range of -19% to +30% as the 95% confidence interval for its current inventory). The effect of CH₄ from coal mining was evaluated assuming emissions from a gassy bituminous coal mine (360 scf/ton) in addition to our baseline assumption of a low-gassy mine (75% lower CH₄ than the gassy mine case).

Fig. S2 shows the sensitivity to CH₄ leakage in each fuel cycle of the Fleet Conversion TWP of converting a fleet of efficient coal-fired power plants to combined cycle natural gas plants. Scenario A in Fig. S2 shows that if leakage in the natural gas supply is sufficiently increased then natural gas can become worse than coal for some period of time. We estimate that natural gas produces net climate benefits relative to low-gassy coal on all time frames as long as leakage in the natural gas system is less than 3.2% from well through delivery at a power plant (i.e., excluding the local distribution system). If the comparison is with less efficient coal plants using higher CH₄ coal types, even higher leakage in the natural gas system could still yield climate benefits. The potential for improving the carbon benefits of natural gas used for electricity can be seen by comparing scenarios C and E, or B and D in Fig. S2; halving upstream leakage in natural gas system reduces the TWP of natural gas power plants compared to coal by 20% and 12% after 20 and 100 years, respectively. Alternatively, capturing coal mine CH₄ emissions (or using coal from low-gassy underground or surface mines) can also improve coal's climate footprint compared to natural gas power plants, as can be seen by comparing scenarios B and C in Fig. S2.

57. Comparison to the Paper by Howarth et al. Our conclusion that natural gas produces net climate benefits relative to certain types of coal reaches the opposite conclusion of Howarth et al. (20) for three principal reasons. The main difference is that Howarth et al. assume much greater methane emissions than we do. As described above, we estimate that 2.1% of natural gas produced is lost annually between the well and the power plant (including the local distribution system, we estimate that the natural gas emitted is 2.4% of gross natural gas production). Howarth et al. used a range of 3.6–7.9% for shale gas and 1.7–6.0% for conventional gas (as a percentage of the CH₄ produced over the life-cycle of a well—also a different metric than we used). From our sensitivity analysis, we conclude that natural gas produces net climate benefits relative to low-gassy coal as long as leakage in the natural gas system is less than 3.2% from well through delivery at a power plant. Second, for the power plant scenario, we did not consider emissions from the local distribution system, under the conservative assumption that most natural gas power plants tie directly into transmission pipelines. Finally, Howarth et al. used higher methane GWP values, reflecting recent evidence that the indirect radiative forcing of methane is larger than the IPCC estimates that we used (21).

58. Data from City of Fort Worth Natural Gas Air Quality Study. The calculations of emissions and leak rates from well sites with no compressor engines were based on data reported in Appendix 3b of the Final Report of the City of Fort Worth Natural Gas Air Quality Study (22). Excel worksheets containing relevant data

from the Fort Worth study and our calculations are available in Dataset S1.

S9. SO₂ Emissions from Coal Plants in the United States and China.

Table S7 summarizes emissions and emission factors of SO₂ from coal-fired power plants in the United States and China. The main paper compares these values with the emission factors used by

Wigley to characterize the global fleet: 12 TgS/GtC in 2010, declining linearly to 2 TgS/GtC in 2060 (23). The values for the calculated emission rates for U.S. coal plants in Table S7 were our own calculations using the selected units to enable comparisons to the Wigley paper and to reported emission rates from China; otherwise the values are in the units reported in the source material.

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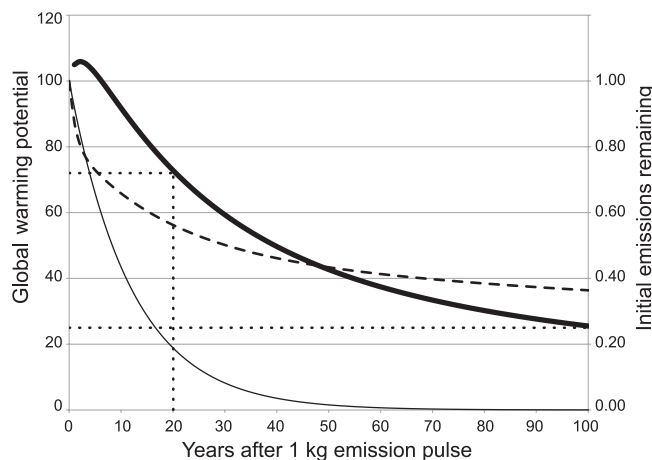


Fig. S1. (A) Decay of 1-kg pulses of CH₄ and CO₂ emissions emitted at time = 0 (thin solid line and dashed line, respectively, with scale shown on right axis) governed by the decay functions given by Eq. 5 and Eq. 6, respectively; (B) The bold solid line shows the variation with time horizon of the global warming potential (GWP) of CH₄, which is the analytic solution of Eq. 1 (scale shown on left axis). The commonly cited GWPs for CH₄ are indicated by the dotted lines (25 and 72 over 100-year and 20-year time horizons, respectively).

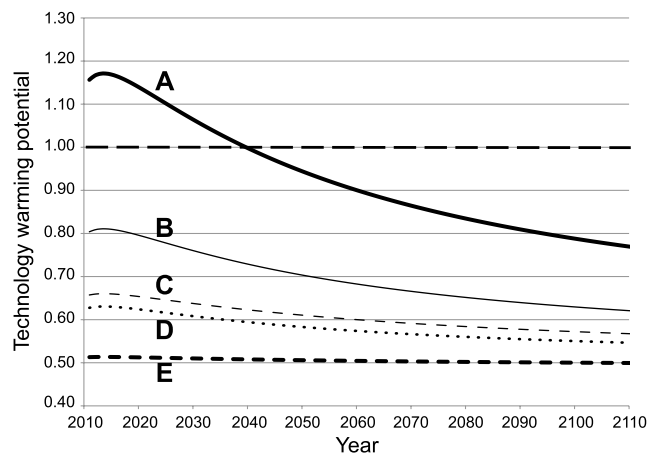


Fig. S2. Sensitivity to upstream CH₄ emissions of the Fleet Conversion TWP of a combined cycle natural gas (NG) plant relative to a supercritical pulverized coal plant, assuming the conversion occurs in 2011 and the following adjustments are made to the base CH₄ emission factors: (A) 2x Base NG CH₄ vs. low-gassy coal; (B) Base NG CH₄ vs. low-gassy coal; (C) Base NG CH₄ vs. gassy coal; (D) 0.5x Base NG CH₄ vs. low-gassy coal; (E) 0.5x Base NG CH₄ vs. gassy coal. Scenario B is the one shown in Fig. 1 of the main paper, which assumed natural gas emissions equal to EPA's 2011 leak rate for the U.S. natural gas supply to the City Gate (2.1%, see text) and low-gassy coal (approximately 90 scf per ton of coal). The natural gas leak rate was doubled (Scenario A) and halved (Scenarios D and E) from the base case. Gassy coal (representing underground coal mine CH₄ emissions of 360 scf per ton of coal) is simulated in Scenarios C and E. See discussion in Section S6.

Table S1. Reference leak rates, L_{REF} , used in this paper

Portion of fuel cycle covered	Applicable scenario	L_{REF}
Well-to-city gate (excludes local distribution systems)	Power plant	2.1%
Well-to-wheels *	Vehicles	3.0%

*In addition to the Well-to-city gate rate, includes 0.3% for local distribution systems and 0.6% for in-use CNG vehicle emissions.

Table S2. 2009 methane and noncombustion CO₂ emissions associated with U.S. natural gas supply (from EPA 2011 GHG EI)*

	CH ₄ Emissions (Tg CH ₄)	CO ₂ Emissions (MMTCO ₂)
Production	6.7	10.9
Processing	0.84	21.2
Transmission/storage	2.1	0.1
Distribution	1.4	0.0
Total	11.0	32.2

*35% of CH₄ emissions and 0% of CO₂ emissions from petroleum systems were allocated to the natural gas supply using the calculation described in Table S3.

Table S3. Data used in this analysis

Parameter	Value	Source
Emissions from natural gas (NG) and petroleum systems	See Table S2	EPA, Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2009, April 2011
NG delivered to consumers (*) (bcf, 2009)	20,966	EIA Natural Gas Annual 2009, Table 1
CO ₂ from power plant combustion of pipeline natural gas (lb CO ₂ /mmBtu, 2008)	117	EPA, Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2008, (April 2010) Table A-33
Heat content of natural gas (Btu/cubic foot, 2009)	1,026	EIA Annual Energy Review 2009, Table A4
CO ₂ from upstream natural gas combustion (lb CO ₂ /million scf)	112,860	Calculated from AP-42 emission factor for stationary gas turbines of 110 lb/MMBtu [AP-42, Table 3.1-2a (April 2000)] and heat content of 1,026 Btu/scf
Calculated mmBtu delivered (2009)	21,511,116,000	Calculated from NG delivered and heat content of natural gas
NG use—lease fuel (bcf, 2009)	913	EIA Natural Gas Annual 2009, Table 1
NG use—plant fuel (bcf, 2009)	362	EIA Natural Gas Annual 2009, Table 1
NG use—pipeline and distribution (bcf, 2009)	598	EIA Natural Gas Annual 2009, Table 1 (50% assumed to be for transmission pipeline)
Electric use pipeline compression (GWh, 2008)	2,937	Transportation Energy Data Book, Edition 29-2010 (50% assumed to be for transmission pipeline)
Total CO ₂ from U.S. power plants (MMTCO ₂ , 2009)	2,154	EPA, Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2009, April 2011, Table 3-1
Total electric power sector generation (GWh, 2009)	3,814,300	EIA, Annual Energy Review 2009, Table 8.2b
Average CO ₂ emissions factor U.S. power plants (lb/MWh, 2009)	1,242	Calculated from total CO ₂ divided by total electric generation
Gross heat content of U.S. crude oil production (mmBtu/barrel)	5.8	EIA, International Energy Annual 2006, Table C.3
U.S. crude oil production (billion barrels, 2009)	1.9	EIA, Annual Energy Review 2009, Table 5.2
Natural gas from oil wells (bcf, 2009)	5,813	EIA, Natural Gas Annual 2009, Table 3
Natural gas fraction of total energy content from oil wells (2009)	0.35	Calculated by dividing mmBtu of natural gas from oil wells into total mmBtu from oil and gas produced from oil wells

*Gross natural gas production in 2009 was 26.0 tcf. EIA Natural Gas Annual 2009, Table 3 (includes Gas Wells + Oil Wells + Coalbed Methane). Our TWP calculations use the amount of gas delivered to reflect the upstream fuel-cycle emissions per unit of gas combusted at the power plant or vehicle. Our calculations of the effective leak rates use the amount of gas produced to reflect losses over the entire fuel cycle.

Table S4. Natural gas fuel-cycle emissions (kg per mmBtu of natural gas delivered to consumers)

	Vented/ leaked CH ₄	Vented/ leaked CO ₂	Natural gas combustion CO ₂			Electric compression CO ₂	Power plant CO ₂
			Lease fuel	Plant fuel	Pipeline		
Production	0.31	0.5	2.2	0.0	0.0	0.0	0.0
Processing	0.039	1.0	0.0	0.9	0.0	0.0	0.0
Transmission/storage	0.098	0.0	0.0	0.0	0.7	0.04	0.0
Distribution	0.064	0.0	0.0	0.0	0.7	0.04	0.0
Power plant	0.0	0.0	0.0	0.0	0.0	0.0	53.1

Table S5. GREET model emission factors used in sensitivity tests (2010 fleet forecast)

	GREET1.8d.1 (unadjusted)		GREET1.8d.1 (adjusted)		GREET 1_2011	
	CNG	Gasoline	CNG	Gasoline	CNG	Gasoline
Vehicle CO ₂ (g CO ₂ /mi)	307	377	307	377	307	377
Vehicle CH ₄ (g CH ₄ /mi)	0.15	0.015	0.15	0.015	0.15	0.015
Upstream CO ₂ (well-to-pump) (g CO ₂ /mi)	58	78	58	78	59	80
Upstream CH ₄ (well-to-pump) (g CH ₄ /mi)	1.3	0.53	2.6*	0.53	3.3	0.65

*The GREET1.8d.1 default output was doubled to obtain this value (see text).

Table S6. Comparison of years to reach climate benefits for gasoline-to-CNG fuel switch using our TWP results compared to TWP calculated with emissions from the GREET model

Emissions source	Years until climate benefits reached	
	Fleet Conversion TWP	Pulse TWP
This analysis	80	46
GREET 1_2011	134	72
GREET 1.8d.1 (unadjusted)	7	5
GREET 1.8d.1 (adjusted)	93	52

Table S7. Emissions and emission factors for U.S. and Chinese coal plants discussed in this paper*

Year	U.S. coal plants				Chinese coal plants (24)		
	SO ₂ (million tons)	CO ₂ (million tons)	Heat input (10 ⁶ mmBtu)	Calculated emission rate (g SO ₂ /GJ)	Calculated emission rate (TgS/GtC)	SO ₂ (Gg)	Emission rate (g SO ₂ /GJ)
2000	10.7	2,122	20,661	445	9.3	9,959	849
2004	N/A	N/A	N/A	N/A	N/A	15,665	796
2008	N/A	N/A	N/A	N/A	N/A	12,486	433
2010	5.05	1,981	19,038	229	4.7	6,587	204
2014 (projected)	3.36	1,981 [†]	N/A	N/A	3.1	N/A	N/A

*U.S. values for 2000 and 2010 are from a query of the U.S. Environmental Protection Agency's Clean Air Markets, Data and Maps Web site <http://camddataandmaps.epa.gov/gdm/index.cfm?fuseaction=emissions.wizard>, performed on December 5, 2011. U.S. values for 2014 were obtained from the U.S. EPA's 2014 projection provided as part of the Technical Support Documents for the Final Cross-State Air Pollution Rule (CSAPR) and the Supplemental Notice of Proposed Rulemaking (SNPR): <http://www.epa.gov/airtransport/techinfo.html>; 2014 includes some emissions from noncoal units. We did not analyze data indicated by N/A for U.S. coal plants for 2004 and 2008 (intermediate years for our analysis but provided to capture estimates of Chinese emissions); no 2014 projection was available for Chinese emissions.

[†]We assumed this value would remain constant between 2010 and 2014.

Other Supporting Information Files.

Dataset S1 (XLSX)

Shows calculations of TWP and L* using Eq. 2 and Eq. 7 of the main paper, respectively (tabs "Calcs-Transp," "Calcs-Power," and "Calcs GREET LD"); GWP calculations using Eq. 1 (tab "GWP"); and calculations using data from the Fort Worth study (tabs "EDF analysis of FW data" and "Raw FW Data").