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THE AIR-CARBON-WATER SYNERGIES AND TRADE-OFFS IN CHINA’S NATURAL GAS INDUSTRY

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Princeton, NJ : Princeton University

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THE AIR-CARBON-WATER SYNERGIES AND TRADE-OFFS IN CHINA'S NATURAL GAS INDUSTRY

Yue Qin

A DISSERTATION
PRESENTED TO THE FACULTY
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Abstract

China's coal-dominated energy structure is partly responsible for its domestic air pollution, local water stress, and the global climate change. Primarily to tackle the haze issue, China has been actively promoting a nationwide coal to natural gas end-use switch. My dissertation focuses on evaluating the air quality, carbon, and water impacts and their interactions in China's natural gas industry.

Chapter 2 assesses the lifecycle climate performance of China's shale gas in comparison to coal based on stage-level energy consumption and methane leakage rates. I find the mean lifecycle carbon footprint of shale gas is about 30–50% lower than that of coal under both 20 year and 100 year global warming potentials (GWP_{20} and GWP_{100}). However, primarily due to large uncertainties in methane leakage, the lifecycle carbon footprint of shale gas in China could be ~15–60% higher than that of coal across sectors under GWP_{20}.

Chapter 3 evaluates the air quality, human health, and the climate impacts of China's coal-based synthetic natural gas (SNG) development. Based on earlier 2020 SNG production targets, I conduct an integrated assessment to identify production technologies and end-use applications that will bring as large air quality and health benefits as possible while keeping carbon penalties as small as possible. I find that, due to inefficient and uncontrolled coal combustion in households, allocating currently available SNG to the residential sector proves to be the best SNG allocation option.

Chapter 4 compares the air quality, carbon, and water impacts of China's six major gas sources under three end-use substitution scenarios, which are focused on maximizing air pollutant emission reductions, CO_2 emission reductions, and water stress index (WSI)-weighted water consumption reductions, respectively. I find striking national air-carbon/water trade-offs due to

SNG, which also significantly increases water demands and carbon emissions in regions already suffering from severe water stress and having the largest per capita carbon footprint. Gas sources other than SNG may bring national air-carbon-water co-benefits. However, end-use deployment can cause enormous variations in air quality, carbon, and water impacts, with notable air-carbon synergies but air-water trade-offs.

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Chapter 1: Introduction

China has been facing growing environmental challenges to improve air quality, address climate change, and reduce water consumption. Severe air pollution in recent years has drawn increasing attention as heavy smog has continually shrouded a large part of northern and eastern China, giving rise to broad discussion and dissatisfaction both inside and outside the country [1, 2]. Simultaneously, China has been facing rising international pressure on climate change ever since it overtook the U.S. and became the world's largest carbon dioxide (CO_2) emitter in 2007. In the 2015 Conference of the Parties (COP21), China pledged to peak national CO_2 emissions by 2030 and to decrease carbon intensity by 60-65% from the 2005 levels [3]. In addition, with one-third of global average per capita renewable water resources and increasing water consumption, China has long been experiencing severe water shortages[4, 5]. The coal-dominated energy structure (roughly 65%) is believed to be one important reason for all three environmental issues. In comparison, natural gas (NG) is widely considered as the cleanest fossil fuel. Earlier studies found net lifecycle emissions reduction of Particulate Matter (PM), SO_2, and NO_x for natural gas substitution of other fossil fuels[6, 7]. In addition, it has relatively low carbon intensities and water consumption coefficients[8, 9]. Thus, the Chinese government has been vigorously promoting natural gas in its energy structure as one way to tackle air pollution, and in the hope of achieving co-benefits of carbon emission and water consumption reductions.

From 2000 to 2015, China's natural gas consumption increased from ~25 bcm to ~190 bcm, at an annual rate of 14%[10]. However, the percentage of natural gas in China's energy structure only increased slightly from 2% to 6% during the same period (Fig.1), which was still far below the global average of 24%[11]. According to China's 12th Five-Year-Plan (FYP), this percentage

will increase to 7.5% by 2015, and over 10% by 2020[12, 13]. Historically, China has been characterized as rich in coal but poor in gas and oil resources [14]. In order to increase gas supply, the Chinese government has been actively encouraging domestic conventional gas production, promoting domestic unconventional gas development (mainly shale gas, coalbed methane, and coal-based synthetic natural gas (SNG)), and increasing natural gas imports in the form of both pipeline gas (mainly from Central Asia and Russia) and liquefied natural gas (LNG). Thus, China's natural gas will come from a variety of sources. Meanwhile, natural gas can also be used in various economic sectors (i.e. industrial, power, and residential sectors) to replace coal end-uses.

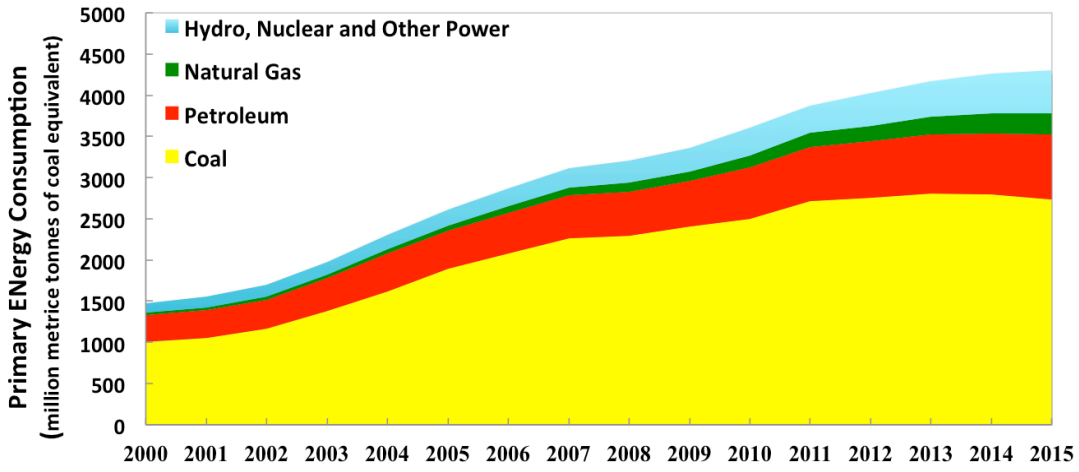


Fig.1 China's historical primary energy consumption (2000-2015).

Despite the fact that natural gas substitution for coal generally brings net air quality benefits, varying sources and deployment strategies of natural gas can result in substantial variations in the magnitude of the air quality, carbon, and water impacts, with potential synergies and trade-offs among various environmental impacts. However, little work has been done to quantitatively evaluate the air quality, carbon, and water benefits of China's natural gas. One reason is that

China's natural gas consumption is constantly small, thus not generating much attention. Also, only in recent years, the landscape of China's gas supply and demand has been quickly changing due to recently identified large unconventional gas resources and increasingly expanding gas imports. Therefore, this work aims to explore the role of natural gas in affecting regional air quality and associated human health, local water stress, and global climate change.

Broadly speaking, this work aims to answer three questions: 1) Based on lifecycle emission studies, will a coal-to-natural gas end-use switch in China facilitate a decrease in lifecycle greenhouse gas (GHG) emissions? 2) How to design end use gas deployment strategy to achieve larger air quality and associated human health benefits at carbon penalties, if any, as small as possible? 3) Whether air-carbon-water synergies or trade-offs exist due to either natural gas source choices or end-uses, and what is their relative importance? I use three analytical chapters to answer the three questions.

Chapter 1 introduces the background of the study. **Chapter 2** answers the question of whether switching from coal to shale gas can bring net carbon reductions in China from the lifecycle emissions perspective. **Chapter 3** evaluates the air quality and the associated human health, and climate impacts of China's coal-based synthetic natural gas development plans. **Chapter 4** characterizes the air-carbon-water synergies and trade-offs due to both gas sources and deployment strategies and compares their relative importance. **Chapter 5** concludes the findings in the three analytical chapters and proposes suggestions on policy making in China's natural gas industry and energy industry in general, with a particular focus on the challenges of using natural gas as a carbon mitigation option in China.

Among the three analytical chapters, **Chapter 2** uses the shale gas industry as a case study to answer the question "Can switching from coal to natural gas bring net climate benefits in

China?" I build a framework to estimate the lifecycle greenhouse gas emissions for China's shale gas industry based on stage-level energy consumption data and methane leakage rates. I then compare the GHG emissions with those from conventional natural gas and coal used in the power, residential, and industrial sectors. As robust decision-making is usually concerned with upper bound estimates, I also estimate the upper bound GHG emissions of China's shale gas industry based on different industrial practices (i.e., green completion, complete gas flaring). In addition, I estimate the breakeven methane leakage rates to help identify how much methane mitigation is sufficient to deliver net climate benefits when switching from coal to shale gas in China. Also, how would the breakeven methane leakage rates change in response to China's efforts in increasing methane recovery rates during coal mining and end-use efficiency improvements in the coal industry? This study finds that: 1) the mean lifecycle carbon footprint of China's shale gas is about 30–50% lower than that of coal in all sectors under both 20 year and 100 year global warming potentials (GWP_{20} and GWP_{100}). 2) Primarily due to large uncertainties in methane leakage, the upper bound estimate of the lifecycle carbon footprint of shale gas in China could be approximately 15–60% higher than that of coal across sectors under GWP_{20}. 3) To ensure net GHG emission reductions when switching from coal to shale gas, I also estimate the breakeven methane leakage rates to be approximately 6.0%, 7.7%, and 4.2% in the power, residential, and industrial sectors, respectively, under GWP_{20}. This project has important implications: 1) Because China's coal, produced primarily from underground mining, has high carbon intensities, shale gas in China may have a better chance to bring net carbon benefits than in the U.S. 2) However, China's continual efforts to upgrade coal-fired power plants and to increase CH_4 recovery rates for coal mining will significantly reduce the carbon intensities of coal. Thus, increasingly strict methane leakage regulations of the shale gas industry are needed to

deliver sustainable net climate benefits of shale gas substitution for coal. 3) From the shale gas production side, either green completions or full flaring of flowback gas results in a lower-than-coal upper bound of lifecycle carbon footprint for shale gas cooking stoves. Green completions, which have already played a significant role in reducing methane emissions from the shale industry in the U.S., can be a critical component in realizing net carbon benefits from shale gas development in China and bringing air quality and economic co-benefits.

Chapter 3 uses the coal-based synthetic natural gas (SNG) industry to evaluate the air quality, human health, and climate impacts of China's natural gas deployment strategies. To increase domestic natural gas supplies, China has planned multiple coal-base synthetic natural gas projects. When combusted, SNG emits negligible SO_2, NO_x, and fine particles. Thus, SNG substitution for coal does help to reduce air pollutants emissions and to improve local air quality. However, compared with direct coal combustion, SNG production and consumption leads to more CO_2 emissions due to energy losses during two stages of energy transformation. Based on earlier 2020 SNG production targets, I conduct an integrated assessment to identify production technologies and end-use applications that will bring as large air quality and health benefits as possible while keeping carbon penalties as small as possible. This study finds that 1) using all production of SNG in the residential sector results in an annual decrease of approximately 32,000 (20,000 to 41,000) outdoor-air-pollution-associated premature deaths. These numbers are roughly 10 and 60 times higher than what can be achieved supposing the same amount of SNG are used to substitute coal in the industrial and power sectors, respectively. 2) All SNG deployment strategies achieve air quality and human health benefits at the expense of climate penalties. Due to inefficiencies in current household coal use, utilization of SNG in the residential sector results in only 20-30% of the carbon penalty compared to using it in the

industrial or power sectors. 3) Compared with direct coal combustion, SNG can bring air quality and carbon co-benefits if carbon capture and storage (CCS) is conducted during SNG production. However, even with CCS, lifecycle CO_2 emissions of SNG are still 22-40% higher than that from direct combustion of conventional natural gas. This study has important policy implications: 1) China's SNG projects can help address its regional air pollution issue, but may worsen climate change. 2) In the U.S., increases in natural gas supplies have been primarily deployed to the power sector. However, in China, due to inefficient and uncontrolled coal combustion in households, allocating currently available SNG to the residential sector provides the largest air quality and health benefits and smallest climate penalties compared with allocation to the power or industrial sectors. 3) The air quality and the associated health benefits brought by SNG can be easily achieved by other natural gas sources (i.e. tight gas, shale gas, coalbed methane, and imported natural gas). These gas sources, assuming proper methane leakage control, shall also bring net climate benefits, making them better options than SNG.

Chapter 4 use an integrated assessment approach in conjunction with lifecycle analysis to quantify net changes in China's air quality, carbon, and water impacts due to gas substitution for coal for each of China's six major gas sources under three substitution scenarios. This study demonstrates striking air-carbon/water trade-offs due to coal-based synthetic natural gas, which also transfers water and carbon stress to regions already suffering from the severest water scarcity and the largest carbon intensity resulting from environmental injustice caused by the energy industry[8, 15]. For gas sources other than SNG, end-use is the key in determining the magnitude of the resulting air, carbon, and water impacts, as well as the air-carbon co-benefits but air-water trade-offs. This study indicates that in some cases, the regional and sectoral energy deployment can play a far more important role in affecting the environmental impacts than

energy source choices. Similar to many emerging economies, China is facing multiple tough environmental challenges. Thus, it is critical to integrate the air quality, climate, and water concerns when designing both energy development and deployment policies.

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Chapter 2. Can Switching from Coal to Shale Gas Bring Net Carbon Reductions to China?

1. Introduction

Facing serious domestic air pollution and growing international pressure to address climate change, the Chinese government has been vigorously promoting the substitution of natural gas for coal to address both issues. Inspired by the U.S.'s shale gas boom, China has been encouraging domestic shale gas development. According to the U.S. Energy Information Administration (EIA), China has the world's largest technically recoverable shale gas resources at around 31.6 trillion cubic meters (tcm)[1]. However, due to challenges including greater formation depth, inadequate water resources, limited experience with horizontal drilling and hydraulic fracturing, underdeveloped pipeline infrastructure, and government-controlled gas prices, China's current shale gas development is sluggish[2-4]. Until recently, commercial shale gas production only existed in Sichuan and Chongqing regions, with a total output of 1.3 billion cubic meters (bcm) in 2014, meeting just 1% of China's total domestic natural gas production that year[5]. However, China's shale gas has large future potential, driven by factors such as large shale resources, favorable policies, improving infrastructure, natural gas market reform, and advancing technologies[6-9].

In the U.S., substantial efforts have been made to compare the lifecycle carbon footprint of conventional gas, shale gas, and coal[10-15]. Concerns exist that methane leakage from shale gas production may result in a worse lifecycle climate performance for shale gas than coal[14, 16]. Thus, it is critical to determine whether shale gas in China can bring net greenhouse gas (GHG)

emission reductions. Constrained by its production size and data availability, there have been very few studies that quantified the environmental impacts of China's shale development. Chang et al. (2014, 2015) estimated the shale-to-well energy consumption and air pollutant emissions, as well as lifecycle GHG emissions of shale gas-fired power generation based on data from China's first shale well in Sichuan basin, using an input-output (IO) lifecycle inventory (LCI) modeling approach[17, 18]. However, as less than 20% of China's natural gas is used in the power sector[19], a detailed comparison of the lifecycle GHG emissions between coal and shale gas across China's major natural gas end-uses is needed. Overall, the climate impacts of China's shale gas development have received less attention than they deserve.

This work serves as one of the first attempts to build a framework to estimate the lifecycle GHG emissions for China's shale gas, based on the major differences between conventional and shale gas systems. We compare the lifecycle GHG emissions of shale gas with that of coal in China's major natural gas end-uses, using China's domestic data whenever available, and supplementing with U.S. data otherwise. In addition, we conduct a sensitivity analysis to identify key factors affecting our estimates of lifecycle carbon emissions of China's shale gas system. We also estimate the breakeven methane leakage rates to ensure net carbon benefits from a coal-to-shale gas switch in China's major end-uses to inform industrial practices and aid policy design.

2. Method

This section describes the method we use to estimate China's lifecycle GHG emissions from conventional natural gas, shale gas, and coal. Both upstream (energy extraction, processing, transport, and distribution) and end-use (power generation, residential cooking, and industrial boilers) processes are included within the lifecycle system boundary (Table S1). Embodied emissions from infrastructure construction are not included in this study. GHG emissions

included in our assessment are: 1) Fugitive CH_4 emissions from both unintentional gas leakage and intentional vents due to safety concerns or engineering design[20]; 2) CO_2 emissions from both energy used to run the entire system and from end-use combustion. Global warming potential (GWP) values from the most recent IPCC report are used to convert CH_4 to CO_2 equivalent (CO_{2eq}) [21]. IPCC estimates that over a 100-year time horizon, the same mass of CH_4 traps 28 times more heat than CO_2 (84 times for 20-years)[21]. As time scale choices have significant implications when evaluating the climate impacts of various fossil fuels, we present the results based on both time horizons.

As data scarcity prevents us to effectively characterize the underlying distributions of emission sources[12, 22], we estimate the lower bound, mean, and upper bound lifecycle GHG emissions of natural gas (both conventional and shale) and coal under a corresponding low, most likely, and high estimate of upstream fugitive emissions and energy combustion emissions (Table S13).

2.1 Upstream GHG Emissions for Natural Gas and Coal in China

2.1.1 Conventional Natural Gas in China

As China's domestic natural gas production is small and is under the control of three national oil companies (NOCs), there is limited publicly available measurement data regarding fugitive emission factors (EFs). Zhang et al. (2010, 2014)[23, 24] estimated China's CH_4 emissions based on Liu et al. (2008)[25], which provided CH_4 EFs from various components of the oil & gas system on the basis of local conditions but primarily rely on the 2006 IPCC Guidelines for Greenhouse Gas Inventories[26]. Meanwhile, U.S. researchers found that fugitive EFs of the U.S. conventional natural gas system are larger than the 2006 IPCC Guidelines based on field measurements in recent years[16, 27-31]. Conventional natural gas industries are long-standing businesses in both

China and the U.S. Thus, we apply the conventional gas methane leakage rates reported in the latest U.S. field studies to China as the "most likely" (mean) case, to take advantage of the U.S. recent measurement and estimation efforts[27-30]. Also, we summarize China's (primarily based on 2006 IPCC guidelines) and U.S.' studies that reported stage-level methane leakage rates for the conventional natural gas industry (Table S3)[10, 12-15, 26, 32], and set the lowest and highest estimates as the lower and upper bound methane leakage rates for China's conventional natural gas system. Table 1 shows the fugitive EFs used in this study for China's conventional natural gas system. Details are shown in the supporting information (SI). Table S4 shows the energy efficiency and energy mix information of upstream processes for China's conventional natural gas industry[33-35]. Recent EFs estimates for fossil fuel combustion are used to estimate CO_2 emissions from upstream energy combustion[36].

2.1.2 Shale Gas in China

Shale gas and conventional natural gas typically have similar chemical composition[37], and their primary difference is that the permeability of shale gas geological formations is approximately six orders of magnitude less than that of conventional gas[38, 39]. Therefore, shale gas extraction is more technically demanding, energy intensive, and costly. As shown in Table S1, the upstream natural gas system has four stages including 1) pre-production and production (referred hereafter as '(pre-) production') & gathering, 2) processing, 3) transmission & storage, and 4) distribution. In the (pre-) production stage, shale gas undergoes additional processes compared to conventional natural gas including horizontal drilling and hydraulic fracturing to enhance the flow of gas into the well. After production, shale gas goes through the same processes as conventional natural gas to reach end users[14, 40, 41]. Thus, the main differences in processes involving shale gas and conventional natural gas are in the (pre-) production stage.

Unlike conventional natural gas production, which is relatively mature in both countries, shale gas production in China is still at a nascent stage. Hence, different policy regulations and industrial practices between China and the U.S. may result in substantial differences in GHG emissions from China and the U.S.' shale gas production. Nevertheless, the primary emission sources for shale gas production are likely to be the same since both countries use the same drilling technologies. Thus, instead of directly applying the U.S. shale gas methane leakage rates to China, we build a framework to estimate GHG emissions from China's shale gas system based on the primary differences between the shale gas and conventional natural gas systems.

We compare additional GHG emissions from the shale gas system with the conventional natural gas system and add them to the shale gas (pre-) production stage. Additional GHG emissions from the (pre-) production stage, including fugitive emissions (Section 2.1.2.1) and CO_2 emissions due to energy combustion (Section 2.1.2.2), are described in the two sections below. GHG emissions in each of the subsequent stages are then calculated according to how much natural gas enters each stage, and the fugitive emissions and fossil fuel energy consumed at each stage (Fig. S1).

2.1.2.1 Upstream Fugitive GHG Emissions

Previous U.S. studies found that well completion and workover are the primary sources of additional fugitive emissions for shale gas due to its much longer flowback period compared to conventional gas (SI)[12, 14, 41]. Well completion is the process to make a drilled and stimulated well ready for production, which happens after hydraulic fracturing and before being connected to permanent processing equipment[41]. Large volumes of hydraulic fracturing fluid and produced water flow back to the surface via the wellbore typically for 1-2 weeks, causing substantial fugitive CH_4 emissions[42]. Workover is occasionally needed to re-stimulate production (e.g., re-

fracturing) and its fluid return process is similar to that of well completion in the pre-production stage[41]. Though well completion and workover are needed for both shale and conventional gas wells, the flowback period for conventional gas wells is usually less than one day or is not necessary[14], thus generating near zero fugitive emissions[14, 41].

To date, commercial shale gas production only occurs in the Sichuan basin, thus we only consider the Sichuan Shale in this study. The geological characteristics of China's Sichuan Shale are similar to the Haynesville Shale in the U.S. (Table S5), so we approximate China's shale gas production profiles using those from the Haynesville Shale[43]. We estimate Sichuan Shale's estimated ultimate recovery (EUR) by scaling a typical Haynesville Shale production curve, similar to the method used by ERINDRC[43]. Our estimates can be updated when China's shale development expands geographically.

Fig.1 shows the predicted gas production rate curve as well as the cumulative production curve for an average well in the Haynesville Shale. The curves are fitted to observed production data from 1,253 Haynesville Shale wells[44]. A detailed description of the model is in Edwards et al. (2015)[45]. Most of these surveyed wells reach their maximum production rates after approximately one month of production, with an average flowback period of about 10 days[14]. We assume this applies also to the Sichuan Shale. Average early shale gas production in Sichuan is currently reported as approximately 60,000 (20,000-100,000) m^3/day[18] and we assume a linear increase in shale gas production before reaching its peak production rate. Thus, we estimate on average total natural gas that comes out during well completion to be about 0.12 million m^3 for the Sichuan Shale, which we assume is either flared at a 98% combustion efficiency or vented directly to the atmosphere (Fig. S2)[13]. As no methane mitigation regulations exist for China's shale gas production, we assume the average flaring ratio in China is the same as that in the U.S.

(51%[18]) prior to the implementation of its methane regulations in 2012[46]. On average, we assume workover is conducted once in each shale well's lifetime in Sichuan. Detailed calculations of fugitive emissions during well completion and EUR for China's shale wells are shown in SI. Table S13 lists the values of key parameters for estimating the lower bound and upper bound well completion and workover fugitive emissions.

2.1.2.2 Upstream GHG Emissions from Energy Consumption

Besides additional fugitive emission sources discussed in 2.1.2.1, CO_2 emissions due to energy consumption from horizontal drilling and hydraulic fracturing, transportation of water and chemicals for hydraulic fracturing, and wastewater transport, treatment and disposal are the three major additional energy-related GHG emissions for shale gas production compared with conventional gas[47, 48]. Chang et al. (2014)[17] reported detailed shale-to-well energy consumption data for China's first shale well covering all three sources, which is used in this study to determine the additional energy consumption for China's shale gas production compared to that of conventional natural gas. Table S7 lists our estimated additional energy consumption and CO_2 emissions for shale production. Following Chang et al. (2014)[49], we assume a success rate for shale well exploration of 45-100%.

2.1.3 Coal in China

The lifecycle system boundary for coal includes all stages of coal mining, processing, transport, and combustion (Table S1). Table S8 shows fugitive EFs for coal mining in China.[23] National average CH_4 recovery rate is estimated to be 7%[24, 50], with a low and high estimate of 0% and 90%, respectively[15, 51]. Table S9 shows the energy efficiency and energy mix for upstream coal processes[33, 35]. In China, 95% of coal mining is from underground mining, of which 47% is from high-methane mines[24]. The average loss in coal transport is about 1.23%[52].

2.2 End Use GHG Emissions for Natural Gas and Coal in China

Natural gas and coal have competing uses in the power, residential and industrial sectors, which together account for approximately 85% of China's current natural gas consumption[19]. Thus we estimate GHG emissions for natural gas and coal combustion across the three major sectors. In China, coal-fired power plants have been continually updated, with subcritical (49%) and supercritical (20%) units being the current mainstream, and high pressure (10%), very high pressure (15%), and ultra-supercritical (6%) units making up the rest[53]. Thus, emissions from the average fleet of various pulverized coal-fired units (PC) are used to represent GHG emissions of China's average coal-fired power plants. More advanced coal-fired power plants, integrated gasification combined cycle (IGCC), are currently under the demonstration stage in China[54]. However, we also calculate the lifecycle GHG emissions for IGCC, given the possibility of its future higher penetration. We use natural gas combined cycle (NGCC), which accounts for ~98% of China's existing gas-fired power generation units[55], to estimate the lifecycle carbon emissions for gas-fired power plants. The average electricity loss in transmission and distribution is assumed to be 6.6%[53]. Additionally, we estimate the lifecycle carbon emissions of coal and gas burned in residential cooking stoves and industrial boilers. Tables S10 and S11 show combustion EFs and energy efficiencies for each sector in China. We evaluate the climate performance of various fossil fuels via comparing their lifecycle carbon intensities (grams of CO_{2eq} per energy produced).

3. Results

3.1 Comparing Lifecycle GHG Emission Intensity for Shale Gas, Conventional Natural Gas, and Coal

As shown in Fig. 2a, using GWP_{100}, the mean estimate of lifecycle GHG emission intensity of a shale gas-fired NGCC (625 g CO_2eq/kWh) is 6% higher than that of conventional gas (590 g CO_2eq/ kWh) under GWP_{100}. Shale and conventional gas-fired NGCC are about 45% and 50% lower than the average PC fleet (1130 g CO_2eq/kWh), respectively. Upgrading coal-fired generators to more efficient types decreases GHG emission intensity due to reduced coal consumption. For instance, we find switching from high-pressure PC (1400 g CO_2eq/ kWh) to ultra-supercritical PC (980 g CO_2eq/ kWh) and IGCC (920 g CO_2eq/ kWh) decreases lifecycle carbon intensity by about 30% and 35%, respectively. Nevertheless, even IGCC (57% higher than conventional gas, 47% higher than shale gas) and ultra-supercritical (65% higher than conventional gas, 55% higher than shale gas) coal-fired generators have higher mean lifecycle GHG emission intensity than a conventional gas- or a shale gas-fired NGCC, indicating the climate importance of switching away from coal power generators (if carbon capture and storage is not employed).

However, large variations exist in the estimate of the lifecycle carbon intensity for shale gas, especially between the mean and upper bound estimates. This can potentially change the relative climate performance of coal-generated electricity and shale gas-fired NGCC. Using GWP_{100}, the upper bound estimate of lifecycle carbon footprint of shale gas generators surpasses that of IGCC, ultra-supercritical and supercritical fleets, which together account for ~30% of China's current coal-fired power fleets. In addition, at the upper bound, we find the lifecycle carbon intensity for shale gas-fired NGCC is 45% higher than average PC fleets using GWP_{20}, ranging from 18% higher than high-pressure PC fleets to 70% higher than ultra-supercritical generators, and 80% higher than IGCC. Thus, without cautious shale development, switching from coal to shale gas could cause worse climate impacts.

We draw similar conclusions in the residential and industrial sectors (Fig. 2b and 2c). The mean estimate of lifecycle carbon intensity for shale gas is marginally larger (~6% with GWP_{100}, ~8% with GWP_{20}) than that of conventional gas in both sectors. Under both time horizons, the mean estimate of lifecycle carbon intensity for conventional gas and shale gas is about 50% and 30% lower than coal to generate 1 MJ energy for residential cooking and industrial boilers, respectively. However, at the upper bound, the lifecycle carbon footprint of shale gas partially overlaps that of coal in both sectors using GWP_{100}. Using GWP_{20}, the upper bound estimate of the shale gas lifecycle carbon intensity is about 15% and 60% higher than that of coal burned in residential cooking stoves and industrial boilers, respectively. This again highlights the importance of cautious shale gas development in China.

3.2 Uncertainties in Estimating Lifecycle GHG Emission Intensity for China's Shale Gas System

Large variations exist among our estimated lower bound, mean, and upper bound lifecycle carbon intensities for each fuel type (Fig. 2). Using GWP_{100}, we find that for average PC fleets, variations are largely resulted from uncertainties in end-use combustion EFs (-8% to +10%), followed by CH_4 recovery rate for coal mining (-9% to +1%), and upstream energy consumption (±1%) (Fig. S3). Similar results are found for coal used in the residential and industrial sectors. Similarly, uncertainties in upstream methane leakage from both well completion and workover (-6% to +48%), and from the conventional gas system (-9% to +35%) contribute the largest variations in our estimated lifecycle carbon intensity for China's shale gas system. In comparison, variations due to uncertainties in lifecycle energy use (-4% to 4%), exploration success rates (-3% to 6%), and end-use combustion EFs (-2% to 3%) are relatively small (Fig. S3).

To understand the importance of each input factor used to estimate additional fugitive emissions from shale gas production, we conduct a sensitivity analysis on well completion and workover (Fig. 3) (refer to SI for details). We find green completions reduce the carbon intensity of shale well completion and workover the most. Green completions, reported to reduce 90% of methane emissions during well completion[41], is conducted during well completion that separates natural gas from liquids and sand to reuse the gas[42]. Flaring ratio results in either a 74% decrease (fully flared) to a 77% increase (fully vented) in well completion and workover carbon intensity. Likewise, the flowback period contributes to an uncertainty range of -51% to +96%, indicating that gas producers can mitigate methane leakage by capturing natural gas as early as possible. As we assume a workover frequency of 0-2 times over shale wells' lifetime, it results in ±50% uncertainties. EUR greatly influences well completion and workover carbon intensity. Even under a conservative estimation (SI), EUR alone can contribute an uncertainty of -40% to +210% to carbon intensity of well completion and workover. Comparatively, gas composition and shale well lifetime have small impacts on carbon intensity.

Additionally, we find that green completions, complete gas flaring during flowback, a high EUR, a low flowback period, and no workover can each reduce the upper bound of shale gas lifecycle carbon intensity by approximately 30%, 30%, 30%, 24%, and 23%, respectively, under GWP_{20} (Table S14). Applying all five practices together bring down the upper bound of carbon intensity by about 40%, which is significantly lower than the upper bound for coal in the power and residential sectors. Indeed, green completions or complete flowback gas flaring alone can generally achieve a lower upper bound for shale gas than for coal in the residential sector.

3.3 Breakeven Methane Leakage Rates that Provide Net Carbon Benefits from a Coal-to-Shale Gas Switch

We further estimate the breakeven methane leakage rates that offer net reductions in lifecycle carbon intensity from a coal-to-shale gas switch in China's major end uses (Fig. 4a) (refer to SI for details). Similar work has been done in the U.S.[16, 56, 57], which help to identify how much methane mitigation is sufficient to deliver net carbon benefits when switching from coal to shale gas. We find that under GWP_{100}, the lifecycle carbon intensity of shale gas used in the power sector, and particularly in the residential sector, is much lower than that of coal even with a methane leakage rate as high as 10%. Over a 20-year time horizon, switching from coal to shale gas can generally bring net reductions in lifecycle carbon intensity in the power, residential, and industrial sectors if the methane leakage rate is below roughly 6.0%, 7.7%, and 4.2%, respectively. Thus, across sectors, shale gas used for residential cooking is most likely to bring net climate benefits. China has traditionally encouraged the substitution of natural gas for coal in the residential sector to maximize air quality benefits, because coal can be burned relatively cleanly in the power and industrial sectors with end-of-pipe control technologies but has extremely high emissions in the residential sector[58,59]. Our analysis supports the pro-residential-sector natural gas allocation policy as it also minimizes GHG emissions and is beneficial for climate.

We find that breakeven methane leakage rates vary with the type of coal-fired power plant fleet used in the comparison (Fig. 4b). Using GWP_{20}, methane leakage must be less than around 8% to ensure that shale gas-fired NGCC has lower lifecycle carbon intensity than China's coal-fired power plants (specifically, IGCC, ultra-super-, super-, sub- critical, very high pressure, and high pressure coal-fired power plants require leakage rates below approximately 4.2%, 4.7%, 5.2%, 5.8%, 6.8%, and 8.1%). With growing penetrations of advanced generators in China's

coal-fired fleets, increasingly strict methane leakage regulations for China's natural gas system are needed to ensure net climate benefits from a coal-to-shale gas switch in the power sector.

Additionally, we evaluate the impact of CH_4 recovery rates for coal mining on the relative climate performance of coal and shale gas. Fig. 4c illustrates the effects for the residential sector. We find that when CH_4 recovery rates increase from 7% (China's current national average) to 90% (EPA reported high value in the U.S.), the breakeven methane leakage rate for shale gas in the residential sector decreases from about 7.7% to 5.4%. Essentially, higher CH_4 recovery rates substantially reduce the lifecycle carbon intensity of coal. This leads to the need for stricter methane leakage regulations on the natural gas industry to ensure net climate benefits when switching from coal to gas. Notably, recovered coalbed methane is also an important source of natural gas. Similar results hold in the power and industrial sectors.

As GWP only considers a single emission pulse instead of a stream of emissions, we also estimate the breakeven methane leakage rates based on technology warming potential (TWP)[16] and obtain similar results: approximately 6.1%, 8.0%, and 4.3% in the power, residential, and industrial sectors, respectively (SI).

Our estimated breakeven methane leakage rates for China are generally larger than the U.S. estimates. For instance, Alvarez et al. (2011)[16] identified that lifecycle methane leakage rates should be below 3.2% to ensure better climate performance for shale gas-fired NGCC than supercritical PC using TWP. Higher breakeven methane leakage rates identified in our study are mainly due to higher lifecycle carbon intensities for coal in China than in the U.S. This primarily results from a much larger fraction of underground coal mining in China (95%) than in the U.S. (40%)[11]. Sub-surface mining has roughly 5 times higher CH_4 emission factors than surface mining[23, 24, 26], and China has a lower CH_4 recovery rate for coal mining[15, 24].

We emphasize that two mitigation efforts, green completions and full flaring of flowback gas, can individually keep the lifecycle methane leakage rates of China's shale gas roughly below 6.5%. Thus, shale gas in China has a large chance to bring air quality and carbon co-benefits with well-regulated industrial practices, particularly in the residential sector. Importantly, U.S. experiences have proved the enforceability of green completions, which have a short payback period and associated air quality co-benefits (reducing O_3 production via mitigating methane and volatile organic compounds emissions) [42]. This technology can be even more attractive to China than to the U.S., as China has higher natural gas prices and much severer air pollution than in the U.S.

4. Discussion and Policy Implications

In this study, we build a framework to evaluate the lifecycle carbon emissions of China's shale gas system and compare it with conventional gas and coal across China's major natural gas end-uses. Under both GWP_{100} and GWP_{20}, the mean estimate of China's lifecycle carbon intensity for shale gas is marginally (~6-8%) higher than conventional gas across all sectors. Shale gas carbon intensity is approximately 45%, 50% and 30% lower than coal for generating 1 kWh electricity in the power sector, producing 1 MJ energy for residential cooking, or 1 MJ energy from industrial boilers, respectively. However, data scarcity, particularly on upstream methane leakage, is a challenging issue, resulting in large variations in our estimates of lifecycle carbon intensity of China's shale gas system. Using GWP_{20}, the upper bound estimate of lifecycle carbon footprint for shale gas surpasses that of coal in all sectors we evaluate here (about 45% higher than average PC fleets, and approximately 15% and 60% higher than typical coal stoves and coal boilers, respectively).

To provide robust conclusions based on existing data, we estimate the breakeven methane leakage rates that ensure net carbon reductions by switching from coal to shale gas, which are roughly 6.0%, 7.7%, and 4.2% in the power, residential, and industrial sectors, respectively, under GWP_{20}. Because China's coal, produced primarily from underground mining, has high carbon intensity, shale gas in China may have a better chance to bring net carbon benefits than in the U.S. However, China's continual efforts to upgrade coal-fired power plants and to increase CH_4 recovery rates for coal mining will significantly reduce the carbon intensity from coal. Thus, increasingly strict methane leakage regulations of the shale gas industry are needed to deliver sustainable net climate benefits of shale gas substitution for coal.

From the shale gas production side, we identify several carbon mitigation opportunities. Requiring green completions, full flaring of flowback gas, achieving a high EUR, reducing flowback periods, and eliminating workover together can reduce the upper bound of shale gas lifecycle carbon intensity by ~40%, well below that for coal in the power and residential sectors. In particular, either green completions or full flaring of flowback gas results in a lower-than-coal upper bound of lifecycle carbon footprint for shale gas cooking stoves. Green completions, which have already played a significant role in reducing methane emissions from the shale industry in the U.S.[27], can be a critical component in realizing net carbon benefits from shale gas development in China and bringing air quality and economic co-benefits.

From the consumption side, we also identify an opportunity to maximize air quality and climate co-benefits by substituting gas (both conventional and shale gas) for coal in the residential sector. Continuing and strengthening the current policy that favors allocation of natural gas to the residential sector can maximize this opportunity at the national level. At a finer

resolution, factors such as local burning conditions and actual energy efficiencies may need to be evaluated to better inform case-specific policy design.

Notably, large uncertainties identified in this work call for field measurements of methane leakage along the whole lifecycle chain of China's natural gas industry. Particularly, based on U.S. experience, a better understanding of methane emissions from high-emitting facilities is essential to characterize the underlying distributions of emission sources. Such an understanding would reduce uncertainties in emission estimates and facilitate a determination of suitable mitigation strategies[22]. Development of a Chinese database of methane leakage would be valuable to address climate, economic and health concerns. Methane leakage often occurs along with the emission of hydrocarbons that are both directly and indirectly (via O_3 formation) harmful to human health. It is also a loss of resource and revenue. The framework built in this study can be used in future updates when more information on methane leakage from domestic measurements becomes available.

In addition, we emphasize that, even if the shale gas industry eliminates its methane leakage, natural gas still only provides an interim step in China's energy decarbonization process. Development of fully decarbonized energy sources is ultimately necessary to avoid dangerous levels of climate change.

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Table 1. Methane leakage rates for conventional natural gas system.

Processes	Mean (%)*	[Low-High] (%)
Gas Production & Gathering	0.86 [27, 28]	[0.05-2.17] [14, 26]
Gas Processing	0.11 [28]	[0.02-0.19] [14, 26]
Gas Transmission & Storage	0.32 [30]	[0.03-2.2] [14, 26]
Gas Distribution	0.09 [29]	[0.01-1.4] [14, 29]
Natural Gas System Total	1.37	0.11-5.96

* Methane leakage rates are calculated as the ratio of total methane emissions within that stage to the national total gross natural gas production reported in that year. Refer to Table S2 for details.

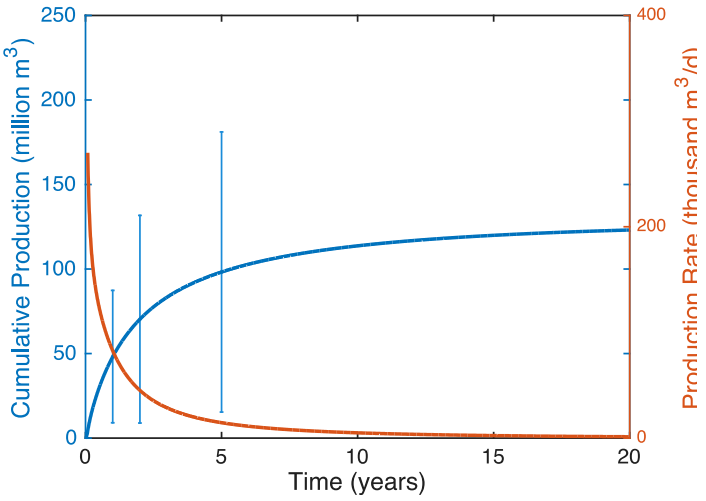
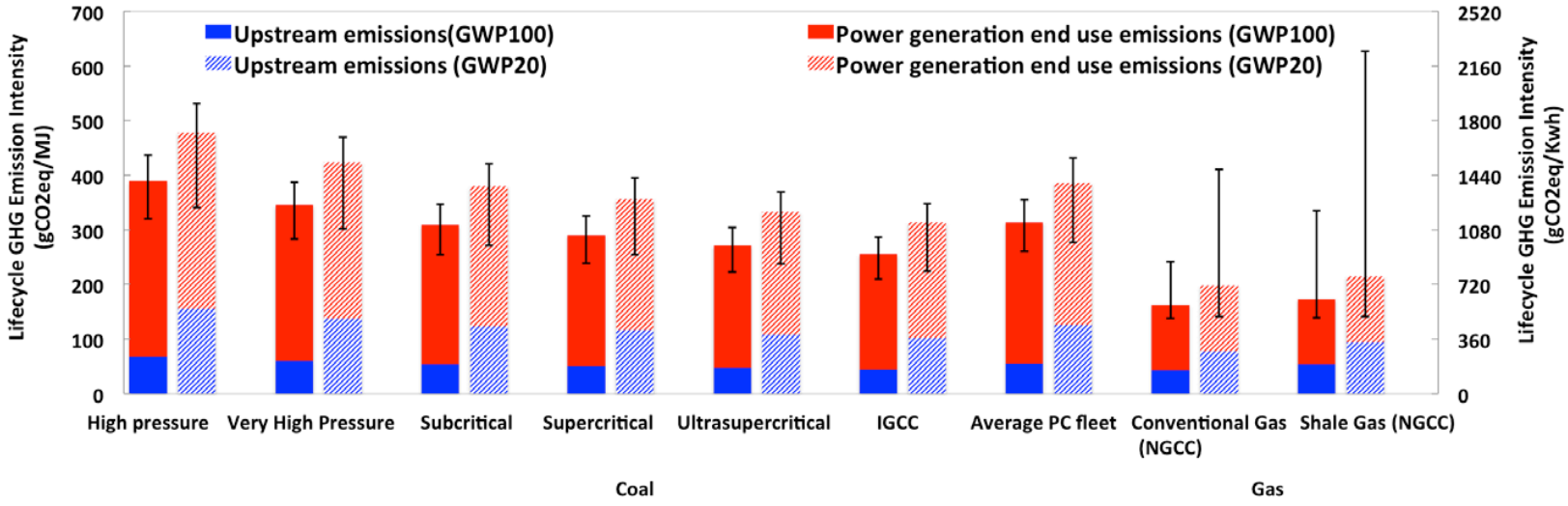
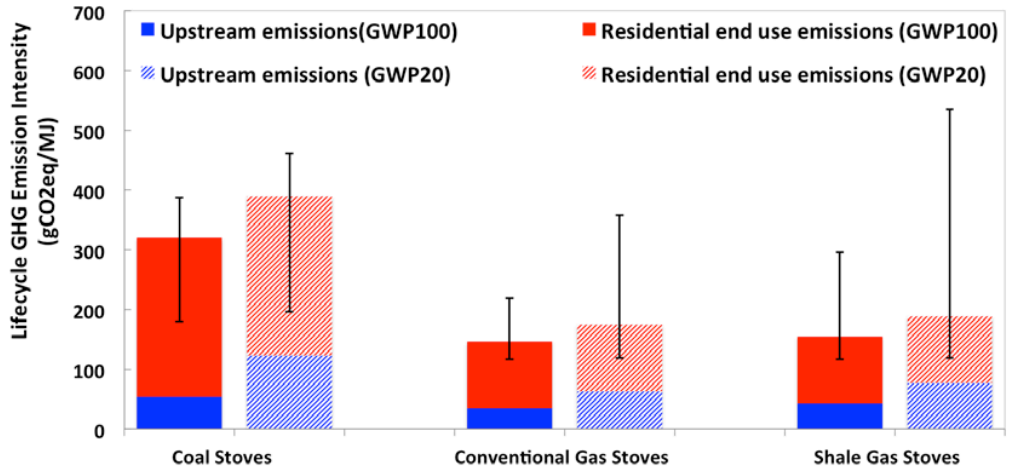


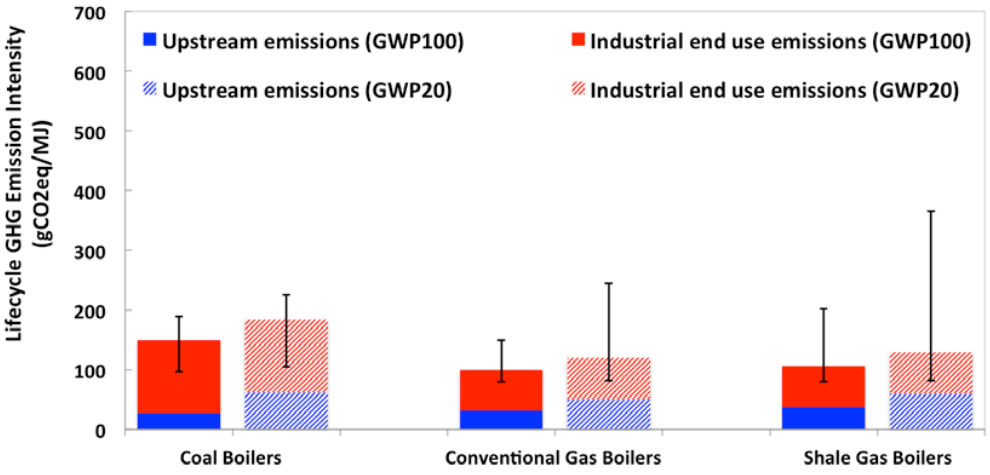
Figure 1. Projected shale gas cumulative production curve (left axis) and production rate (right axis) for an average Haynesville Shale well. Cumulative production plus and minus two standard deviations for individual wells at the 1st, 2nd, and 5th year of production are shown with blue bars.


Upstream emissions(GWP100)
Upstream emissions (GWP20)
Power generation end use emissions (GWP100)
Power generation end use emissions (GWP20)
Lifecycle GHG Emission Intensity (gCO2eq/MJ)
Lifecycle GHG Emission Intensity (gCO2eq/Kwh)
0
100
200
300
400
500
600
700
0
360
720
1080
1440
1800
2160
2520
High pressure
Very High Pressure
Subcritical
Supercritical
Ultrasupercritical
IGCC
Average PC fleet
Conventional Gas (NGCC)
Shale Gas (NGCC)
Coal
Gas

(a)



(b)



(c)

Figure 2. Lifecycle GHG emission intensity for coal, conventional natural gas and shale gas in the (a) power sector, (b) residential sector, and (c) industrial sector. Figures show results for GWP_{100} and GWP_{20}. Error bars show the lower and upper bound estimates of lifecycle GHG emission intensity as defined in Table S13. Note average PC fleet does not include integrated gasification combined cycle (IGCC).

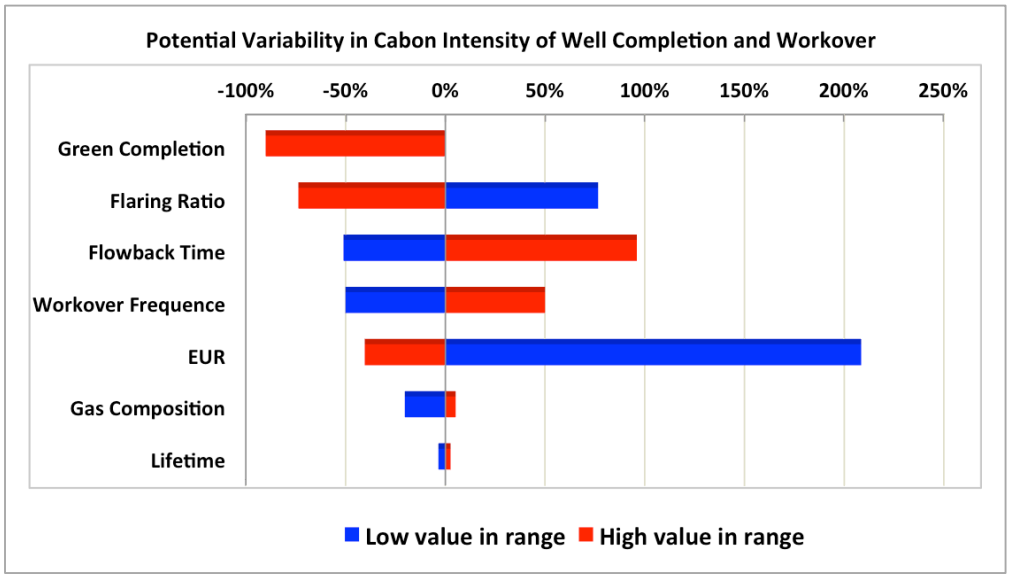
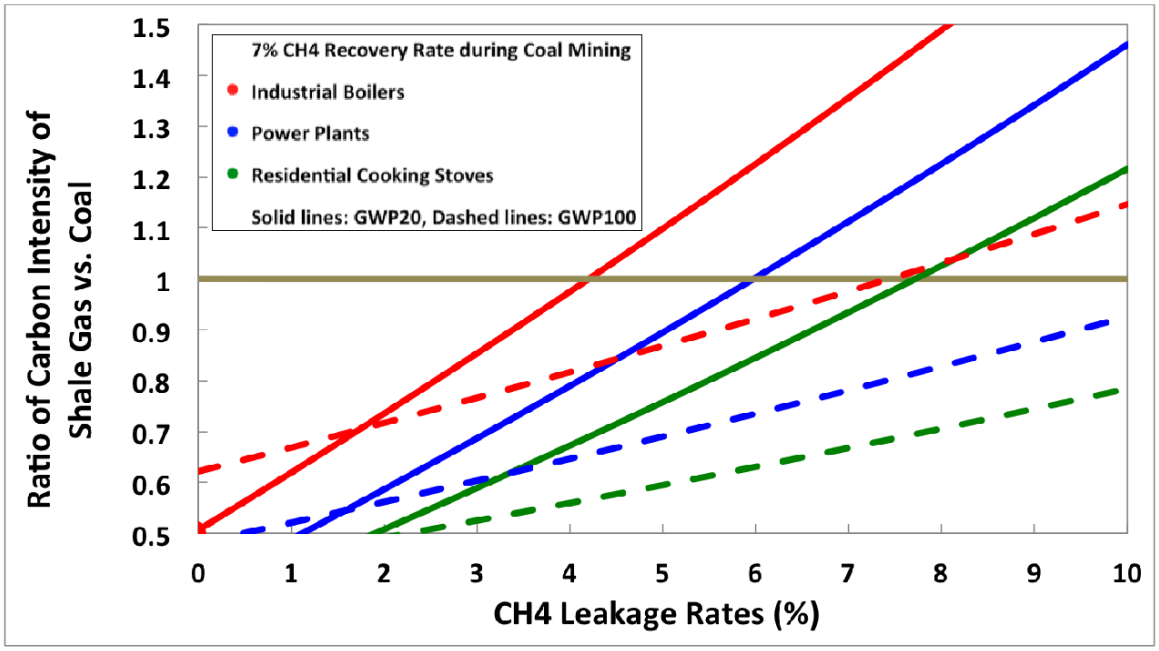
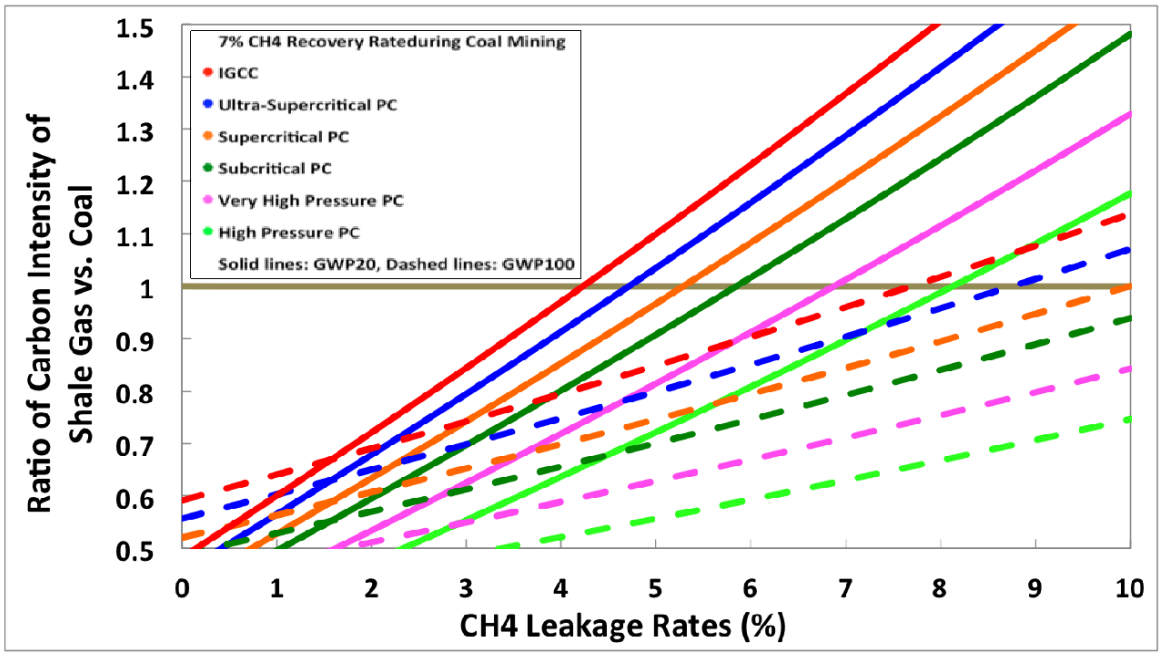


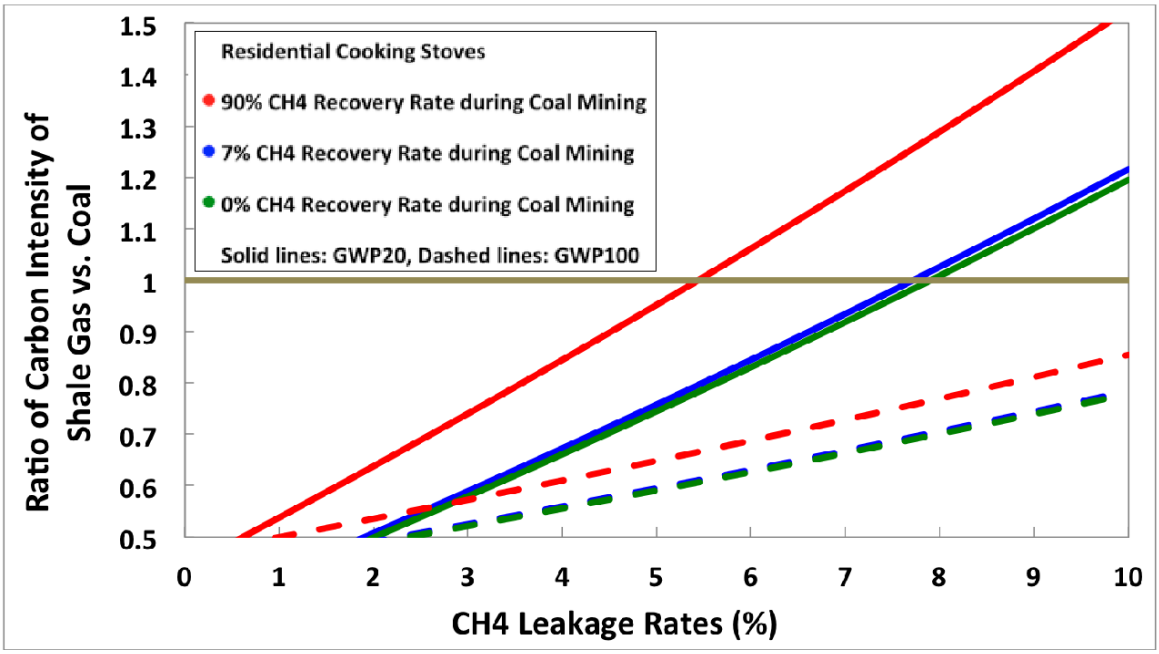
Figure 3. Sensitivity analysis for GHG emission intensity during well completion and workover for shale gas over a 100-year time horizon. 0% indicates the mean estimate. Positive (negative) % refers to percentage increases (decreases) in the estimated carbon intensity due to each factor in the y-axis, while keeping all other factors at the mean value.


7% CH4 Recovery Rate during Coal Mining
Industrial Boilers
Power Plants
Residential Cooking Stoves
Solid lines: GWP20, Dashed lines: GWP100
Ratio of Carbon Intensity of Shale Gas vs. Coal
CH4 Leakage Rates (%)
1.5
1.4
1.3
1.2
1.1
1
0.9
0.8
0.7
0.6
0.5
0
1
2
3
4
5
6
7
8
9
10

(a)


7% CH4 Recovery Rateduring Coal Mining
IGCC
Ultra-Supercritical PC
Supercritical PC
Subcritical PC
Very High Pressure PC
High Pressure PC
Solid lines: GWP20, Dashed lines: GWP100
Ratio of Carbon Intensity of Shale Gas vs. Coal
CH4 Leakage Rates (%)
1.5
1.4
1.3
1.2
1.1
1
0.9
0.8
0.7
0.6
0.5
0
1
2
3
4
5
6
7
8
9
10

(b)



(c)

Figure 4. Lifecycle carbon intensity breakeven ratio of shale gas compared with coal. (a) Comparison of three end-use sectors – industrial boilers, power plants, and residential cooking stoves. In the power sector, we use China's national average PC fleet composition to represent the lifecycle carbon intensity for coal, and NGCC for shale gas. A 7% CH_4 recovery rate for coal mining is used for all sectors. (b) Comparison of various coal-fired power plant fleet types in the power sector. A 7% CH_4 recovery rate is used for all fleet types. (c) Comparison of CH_4 recovery rates for coal mining when gas is used in residential cooking. In each figure solid lines indicate the results for GWP_{20}, and dashed lines for GWP_{100}. Color legends work for both the solid and dashed lines with the same color.

Chapter 3. Air Quality, Human Health and Climate Implications of China's Synthetic Natural Gas Development

1. Introduction

China's ongoing coal-based synthetic natural gas (SNG) development is largely motivated by efforts to reduce its extreme ambient air pollution and dependence on foreign natural gas. Over the past fifteen years, China's total natural gas consumption has increased from 25 billion cubic meters (bcm) in 2000 to 187 bcm in 2014, an annual growth rate of 15%[1]. However, domestic natural gas production failed to keep pace with the increases in demand. In 2007, China's natural gas consumption surpassed domestic production for the first time and by the end of 2014, dependence on foreign natural gas had increased to 30%[1]. After publicity surrounding high air pollution levels in 2013[2-4], the Chinese government aimed to further increase both the quantity and the proportion of energy obtained from natural gas[5]. Until 2014, natural gas accounted for approximately ~9% and ~5% of China's energy consumption in the residential (including both residential and commercial cooking and heating) and industrial sectors, respectively, as well as ~2% of national total electricity generation, suggesting large potential for further increases in natural gas use in these sectors[1].

Historically, China has been characterized as rich in coal but poor in gas and oil[6]. Recently, to increase natural gas supplies, the central government has emphasized domestic unconventional gas development, including SNG. As a result, 2013 was considered a "golden year" for the SNG industry. Through 2013, a total capacity of 37.1 bcm yr^{-1} of SNG production had been approved by the central government, with another 40 projects (~200 bcm yr^{-1}) proposed by the industry[7].

Total planned SNG capacity is ~1.25 times China's 2014 total natural gas consumption[1]. Also, the Chinese government set targets for annual SNG production of 15-18 bcm by 2015 and 32 bcm by 2017, with a potential production of approximately 57 bcm by 2020[8-11]. Notably, government plans for SNG production are continuously changing, likely due to a mix of concerns about the coal industry, local economy, air pollution in eastern China, energy security, local water stress, and global climate change[7, 12-15].

China's coal-based SNG strategy converts low-quality dirty coal in western parts of China into synthetic natural gas via coal gasification and methanation. As methanation catalysts are prone to sulfur poisoning, hydrogen sulfide is removed from the coal and converted to elemental sulfur prior to methanation. This essentially eliminates emissions of sulfur compounds including SO_2. Additionally, the SNG production process emits negligible NOx, which would occur if the coal were burned in steam and power-generating boilers[16]. After converting coal to SNG, it is transported to and used in eastern provinces. Compared with direct combustion of coal, gas combustion emits negligible fine particulates and SO_2, and little NOx[17-19]. Thus, replacing coal with SNG substantially reduces emissions of air pollutants at the location of the end-user[20]. However, the SNG cycle emits more CO_2 than direct use of coal. For electricity generation, up to 60% higher lifecycle CO_2 emissions occur with SNG than with ultra-supercritical coal-fired power plants per kWh electricity generated[12]. As a result, plans for increasing use of SNG are projected to dramatically increase CO_2 emissions.

An integrated analysis of the impacts of China's SNG plans on national air quality and associated health benefits as well as on global carbon emissions is needed to provide guidance to the Chinese government on SNG development. Moreover, as multiple SNG projects are already in place or under construction, it is important to determine production technologies and end-use

applications that will bring as large air quality and health benefits as possible while keeping carbon and energy penalties as small as possible.

This paper quantifies the air quality, human health and CO_2 emission impacts of China's SNG strategy using an integrated assessment approach. We use the ECLIPSE_V5a_CLE scenario for 2020 as our base case as it reflects the air pollution policies and regulations in place for China's 12th Five-year plan (FYP)[21-24]. Approximately 85% of natural gas in China is consumed in the power, industrial, and residential sectors[25]. Thus, we construct three SNG sectoral allocation scenarios (SNG_Power, SNG_Industrial, and SNG_Residential) by deploying all potentially available SNG (57 bcm) in 2020 into each key demand sector in turn. We substitute SNG for coal in each sector we analyze in proportion to the gas required to displace that quantity of coal in the subsector under the base case across provinces targeted to receive SNG (Table S1). Due to large uncertainties in actual SNG production in 2020 and practical constraints on SNG deployment, SNG allocation scenarios built in this study are effectively sensitivity analyses to identify the potential impacts of SNG use in each sector. They are not intended to be analyzed as to their actual executability in the real world. We estimate changes in air pollutant emissions resulting from SNG substitution for coal under each scenario (medium scenarios in Table S1). We then simulate the resulting changes in $PM_{2.5}$ concentrations, associated health impacts, and resulting changes in CO_2 emissions. Fig. 1 shows the potential provincial distribution of SNG production and consumption based on government plans and gas pipeline infrastructure[7, 8, 12, 26].

Through regional atmospheric chemistry model simulations, we evaluate the monthly mean $PM_{2.5}$ surface concentrations in January, April, July and October at a horizontal resolution of 27 $\times$ 27 km^2 for the base case and for each SNG scenario. The mean concentration for the year is

assumed to be the average of these four months. We also estimate the population-weighted (P-W) annual average $PM_{2.5}$ surface concentrations at the provincial level.

Burnett et al. (2014) developed disease-specific integrated exposure response (IER) functions for $PM_{2.5}$ that cover the global range of exposures[27], including ischemic heart disease (IHD), cerebrovascular disease (stroke), chronic obstructive pulmonary disease (COPD), and lung cancer (LC) for adults (≥25 years old), and acute lower respiratory infection (ALRI) for children (<5 years old). Based on the IER functions and P-W provincial $PM_{2.5}$ surface concentrations, we estimate the disease-specific population attributable fraction (PAF) from exposure to ambient $PM_{2.5}$ for each province and the corresponding air-pollution related premature mortality. Differences in national total premature deaths between each SNG scenario and the base case are used to estimate the avoided premature deaths under each SNG scenario. In parallel, we estimate the net changes in CO_2 emissions resulting from SNG substitution for coal under each scenario. Calculation details are described in the supporting information (*SI Materials and Methods*).

2. Results

2.1 Impacts of SNG Substitution for Coal on Air Pollutant and CO_2 Emissions

For all scenarios, substituting SNG for coal results in net reductions in air pollutant emissions: -40 to -0.7 g SO_2/m^3 SNG, -5 to -0.5 g NO_x/m^3 SNG, -21 to -0.7 g PM_{10}/m^3 SNG, -19 to -0.4 g $PM_{2.5}/m^3$ SNG, varying primarily on the end-use application (Fig. 2). This occurs because natural gas has higher energy efficiencies and lower air pollutant emission factors than coal per unit energy input.

Use of SNG in the residential sector results in the largest reductions in the emissions of all air pollutants considered here (Fig. 2). For instance, allocating all planned SNG to the residential sector reduces SO_2 emissions more than twice as much as allocating all SNG to the industrial

sector, and 15 times more than allocating it to the power sector. SNG allocation to the power sector reduces air pollutant emissions the least. This is primarily because the power sector has the most stringent emission controls on coal combustion among the three sectors, while the residential sector coal emissions are generally uncontrolled. This results in the lowest average abated air pollutant emission factors in the power sector (SI 1.2, Fig. S1). The industrial sector also has a large fraction of low-emitting coal boilers due to efficient control technologies, particularly for SO_2 and particulate matter. In addition, thermal efficiency improvement from a coal to gas switch is the largest in the residential sector (Table S2). Thus, proportionally substituting coal with the same amount of SNG in the residential sector leads to much larger air pollutant emission reductions than in the other two sectors. We also present the spatial distribution of monthly mean SO_2 emission reductions under each scenario as an illustration (Fig. S2), and identify substantial reductions across four seasons when all planned SNG is allocated to the residential sector, particularly in Beijing (28%), Tianjin (13%), and Hebei (18%) provinces.

Across scenarios, however, we observe energy (7-28 MJ/m^3 SNG) and CO_2 (0.5-2.5 kg CO_2/m^3 SNG) penalties. This is because the higher energy content of gas over coal per carbon atom is offset by the larger quantity of coal used to produce SNG. Although energy and CO_2 penalties cannot be avoided completely, we find that SNG allocation to the residential sector, in addition to providing the largest reductions in air pollutant emissions, results in the least energy losses and the smallest increases in CO_2 emissions. This is again because the largest thermal efficiency improvement of switching from coal to gas occurs in the residential sector (SI 1.2, Table S2)[28].

2.2 Impacts of SNG Substitution for Coal on $PM_{2.5}$ Surface Concentrations

We simulate China's 2020 baseline monthly mean $PM_{2.5}$ surface concentrations for January, April, July and October (Fig. S4a). Model evaluation is shown in the supporting information (*SI Model Evaluation*). $PM_{2.5}$ concentrations are significantly higher in January and October than in April or July. This is due to higher emissions resulting from residential heating in winter, and more stagnant meteorological conditions and less precipitation in January and October. In addition, relatively low simulated $PM_{2.5}$ concentrations occur in April partly because dust emissions are not included in our simulations. We find the Beijing-Tianjin-Hebei (BTH) region has extremely high area wide $PM_{2.5}$ concentrations, with maximum monthly mean $PM_{2.5}$ levels reaching 170 $\mu g/m^3$ at the grid level in January. Even in April and July, the maximum monthly mean $PM_{2.5}$ concentrations in BTH can be more than 80 $\mu g/m^3$.

Monthly mean $PM_{2.5}$ surface concentrations are reduced in all SNG scenarios, but by far the largest decrease occurs when all planned SNG is allocated to the residential sector (Fig. 3). This is because of reductions in both primary $PM_{2.5}$ emissions and less formation of secondary $PM_{2.5}$ due to reduced emissions of major precursors (i.e. SO_2 and NO_x). Grid-level reductions are generally more than 5 $\mu g/m^3$ across all seasons in the BTH region, and can reach 60 $\mu g/m^3$ (~30% reduction) in the dirtiest season (winter). In comparison, the $PM_{2.5}$ concentration reductions are less than 2 $\mu g/m^3$ in the BTH region year-round when all available SNG is allocated to the power sector, and less than 5 $\mu g/m^3$ when it is allocated to the industrial sector (Fig. S4 b-c).

China's base case annual average population-weighted $PM_{2.5}$ surface concentrations at the provincial level in 2020 are shown in Fig.4a. Annual average P-W $PM_{2.5}$ concentrations are projected to be approximately 70 $\mu g/m^3$ in Beijing, Tianjin and Hebei provinces. This is twice China's annual average $PM_{2.5}$ national standards (35 $\mu g/m^3$) (GB 3095-2012) and 7 times that of the WHO (10 $\mu g/m^3$) standards[29, 30].

Figure 4b-4d show changes in annual average P-W $PM_{2.5}$ surface concentrations across SNG sectoral scenarios. Using SNG in the residential sector to replace coal leads to the largest provincial-level P-W $PM_{2.5}$ concentration reductions. For instance, $PM_{2.5}$ concentrations are reduced by 19 $\mu g/m^3$ (26%), 12 $\mu g/m^3$ (17%) and 13 $\mu g/m^3$ (18%) in Beijing, Tianjin and Hebei provinces, respectively. In addition to the BTH region, several other SNG producing and consuming provinces, such as Henan, Shandong, Jilin, Shanxi and Inner Mongolia, also exhibit approximately 10% $PM_{2.5}$ concentration reductions because of substantial reductions in SO_2 and $PM_{2.5}$ emissions. In contrast, provincial-level $PM_{2.5}$ concentration reductions are virtually small when all planned SNG is allocated to the industrial or power sectors (generally less than 0.6 $\mu g/m^3$ (<1.5%) and 0.2 $\mu g/m^3$ (< 0.5%), respectively).

2.3 Impacts of SNG Substitution for Coal on Premature Mortality

National total avoided premature deaths under our SNG scenarios are shown in Fig. 5, with stroke and IHD contributing roughly 60% of total reductions in premature mortality. Across scenarios, use of SNG in the residential sector results in the largest decreases in total adult premature deaths of approximately 32,000 (20,000 to 40,000) and child deaths of 320 (200 to 400) annually, with the range resulting from the low and high estimates of relative risks. These reductions are roughly 10 and 60 times higher than reductions obtained by deploying SNG in the industrial and power sectors, respectively. Consistent with Liu et al. (2016) findings for the BTH region[31], our results highlight enormous benefits for China's air quality and associated human health by switching from coal to cleaner fuels in the residential sector across the country.

2.4 Comparison of SNG Scenarios' Air Quality and Climate Impacts with and without utilization of CCS

Using all SNG in the residential sector clearly provides the largest air quality and human health benefits, with the smallest energy and CO_2 penalties among the scenarios we evaluate (Fig.6). However, even under the SNG_Residential scenario, coal substitution with SNG results in an increase of 28 Mtonnes of CO_2 emissions, approximately 0.2% of national total projected CO_2 emissions (~12 billion tonnes) in 2020[24] (Table S5). For comparison, we replace coal with the same amount of conventional natural gas as SNG in the residential sector, and follow the same allocation strategies shown in Table S1 (medium scenarios). We find such replacement can reduce CO_2 emissions by 214 Mtonnes relative to household use of coal, or reduce CO_2 emissions by 242 Mtonnes relative to SNG while providing the same amount of air quality and health benefits.

As all SNG allocation strategies increase CO_2 emissions relative to coal, we explore whether carbon capture and storage (CCS), if employed with SNG production, can make SNG an attractive option for both air quality and climate. China has been one of the major players working on CCS demonstration projects in recent years[32]. In SNG production CO_2 is separated from syngas before methanation regardless of conducting CCS because this optimizes the economics of the process by cost savings from higher methanation efficiency and lower volume of input syngas[16, 33]. Thus, CO_2 is emitted as a by-product of SNG production and it does not require additional energy to separate the CO_2, therefore causing a relatively low energy penalty when conducting CCS[16]. We estimate the net energy penalty for conducting CCS at SNG plants to be ~9%-15%, with the range dependent on where the energy for conducting CCS comes from (*SI Section3*).

Nevertheless, even with CCS employed for SNG production, net CO_2 emissions are still 22-40% higher than occurs with the same amount of conventional natural gas, with the range

depending on end uses and the energy sources for CCS (Table S5). Substituting coal with SNG equipped with CCS requires 20-100% additional energy input compared with directly burning coal, varying primarily depending on end uses (Table S5). Thus, we find that SNG cannot simultaneously address the multiple objectives facing China: air quality improvement, carbon emissions mitigation, and energy intensity reduction.

3. Discussion

This study identifies deploying SNG to the residential sector as the allocation strategy providing the largest net environmental benefits among the substitution scenarios analyzed. We leave it to policy makers to decide how much they are willing to pay for these benefits. We did not include economic analysis of each SNG scenario. We realize that even though deploying SNG to the residential sector results in much larger health benefits and lower climate penalties than to the power and industrial sectors, it may also require higher investment on last-meter distribution pipelines, particularly for rural areas with low population density[28]. However, given only 40% of China's urban population used natural gas in 2015[1], there may still be large opportunities to distribute SNG to urban residents using solid fuels before making large investments in reaching rural residents. Additionally, in low population-density regions, SNG can be compressed or liquefied and then transported by trucks that can be more cost-effective than pipelines[34, 35]. Some provincial governments are already expanding natural gas uses to rural areas by subsidizing gas pipeline construction for rural households and transporting natural gas in the form of LNG and CNG to improve the clean fuel accessibility for rural residents[36].

The absolute environmental impacts estimated in this study are subject to uncertainties in actual SNG production, how well the ECLIPSE_V5a_CLE scenario reflects the actual energy and emission status in 2020, and the representation of $PM_{2.5}$ in our atmospheric chemistry model.

Our model captures the magnitude and trend of $PM_{2.5}$ concentrations fairly well, particularly in eastern regions where air quality and human health improvement predominantly occur (*SI Model Evaluation*). The actual energy use and emissions in 2020 will likely differ from the ECLIPSE_V5a_CLE scenario. Indications are that actual SNG production will be lower than the production target we use here due, in part, to the Chinese central government's frequent downward adjustment of SNG production plans. However, our finding that substantial health benefits and relatively small climate penalties occur when available SNG is allocated to the residential sector is likely to persist as long as inefficient and uncontrolled coal use continues in the residential sector.

Our atmospheric chemistry model places all emissions into the surface layer (0-32 m) and air pollutant concentrations in each grid box are instantly well mixed. The power and industrial sectors discharge much of their emissions via stacks above 32 m, while the residential sector primarily discharges closer to the surface than is resolved in the model. Thus, our study may have overestimated the $PM_{2.5}$ associated health benefits under the SNG_Power and SNG_Industry scenarios, and underestimated that under the SNG_Residential scenario. In addition, as household outdoor emissions are released closer to populations, they have higher intake fractions (dose or exposure effectiveness) than those from industry or power[37], which would widen the differences in benefits even further if taken into account. Finally, the significance of the residential sector would be even more striking if we included benefits to the household environment itself, i.e. health impacts from indoor and near field air pollution[38]. Nevertheless, SNG may have to be allocated to other sectors if too much future SNG production causes saturation of the residential sector or if costs of pipeline construction limit the spread of SNG to the more densely populated areas. Total 2020 SNG evaluated here (~30% of currently

industry-planned SNG projects) replaces ~60% of baseline coal consumption in the residential sector. Supposing all of the ~200 bcm yr^{-1} SNG projects are implemented, this will quickly use up the opportunities for coal replacement in the residential sector and lead to significantly lower marginal health benefits but larger marginal carbon penalties.

Our study does not consider the interactions between SNG and renewable energy, which can potentially increase the air quality and climate benefits obtained from the power sector and potentially allow electrification of the residential sector at a low-carbon intensity. However, the role that natural gas can play in facilitating wind and solar on-grid integration is likely to be limited in China in the near future given the small amount of natural gas in the power sector (even including all planned 2020 SNG). Also, the primary barriers of China's renewable integration lie in elsewhere (i.e., over-supply of coal-fired electricity with fixed annual operation hours and inadequate transmission capacity)[39].

4. Conclusions

China's SNG development has important implications for both regional air quality and global climate. Since 2013, China's severe air pollution has drawn enormous public attention and a commitment from the government to implement measures that improve air quality[4]. Switching from coal to gas is identified in the national action plan as a strategy to improve air quality[5], and efforts to increase natural gas supply including via development of SNG production are under way. However, wide concerns about China's SNG strategy exist as CO_2 emissions per unit of end use energy delivered from SNG projects greatly exceed that associated with most other energy sources and will have lasting and significant impacts on climate change[7, 15].

We find that sectoral allocation makes a huge difference in the environmental performance of a limited quantity of SNG. For instance, SNG substitution for coal in the residential sector can

reduce $PM_{2.5}$ concentrations in the BTH region by approximately 20%. These are among the most densely populated regions with the worst air quality in China. Additionally, it can avoid 32,000 (20,000 to 41,000) air pollution-related premature mortality nationwide in 2020. In contrast, allocating all SNG to the power or industrial sectors barely improves air quality and avoids only 560 (230-740) and 3,100 (1,300-4,300) premature deaths, respectively. Similarly, due to relative efficiencies in the use of coal and gas in the industrial and power sectors compared with the residential sector, net increases in CO_2 emissions when all planned SNG is used in the industrial or power sectors to replace coal are 2 and 4 times higher, respectively, than if it is used in the residential sector. We also compared the health impacts and net carbon emissions from two regional allocation scenarios, and find that allocating SNG to affluent provinces or proportionally to provinces based on their baseline gas needs for coal replacement leads to similar reductions in national total premature mortality and carbon emission increases (*see SI Section 4 for details*).

Critically, energy and CO_2 penalties exist across all scenarios. Thus, without CCS used in SNG production, the air quality and human health benefits of SNG substitution for coal are achieved at the expense of CO_2 emission increases. Even with CCS, however, the climate performance of SNG remains worse than conventional gas (SNG+CCS emits 22-40% more CO_2 than the same amount of conventional gas), and applying CCS for SNG production results in approximately 9-15% additional energy losses than SNG without CCS. In China's 2015 intended nationally determined contributions (INDC), China pledged to peak its CO_2 emissions by 2030 or earlier, and lower CO_2 emissions per unit of gross domestic product (GDP) by 60-65% from the 2005 levels by 2030[40]. Thus SNG development is inconsistent with China's efforts to reduce energy and carbon intensity, but does provide substantial air quality improvement with relatively

small climate co-benefits if done in conjunction with CCS. To achieve its goals, China may wish to limit the scale of SNG development and to conduct pilot projects on pairing CCS with SNG production to facilitate achievement of its international climate commitment while addressing its domestic air pollution issue.

Allocating SNG to the residential sector is likely to bring the largest air quality benefits with the smallest carbon penalties even without CCS. Importantly, the air quality benefits brought by SNG can easily be achieved by other sources of natural gas, which have a lower carbon footprint. Given the multiple challenges facing China today, other domestic gas sources, including tight gas, coalbed methane (CBM), and shale gas (with methane leakage well controlled)[41], as well as increased energy efficiency and an increasingly electrified energy economy driven by renewable energy, are likely to provide equal air quality benefits with lower negative climate impacts. Large challenges exist, however, in switching from coal to natural gas. Challenges include an underdeveloped pipeline infrastructure, high infrastructure costs particularly in low population regions, and low price-competitiveness of natural gas compared with cheap coal. However, the Chinese central government has demonstrated a political willingness to address these issues and has set ambitious near- and long- term natural gas usage targets, designed and approved substantial natural gas pipeline expansion projects (i.e., Xinjiang SNG pipeline project) while reforming China's natural gas market[8, 42]. Meanwhile, provincial and lower-level governments are also subsidizing local pipeline construction and natural gas consumption for rural residents[36]. Nevertheless, a reasonable price on carbon would facilitate a more accurate valuation of natural gas relative to both carbon-intensive coal and carbon-free renewables.

5. Materials and Methods

We use an integrated assessment approach to estimate the air pollution associated human health impacts in mainland China under each SNG scenario. The ECLIPSE_V5a_CLE scenario, developed by the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) model at the International Institute for Applied Systems Analysis (IIASA, http://gains.iiasa.ac.at/models/), is used as our 2020 base case anthropogenic emission input. The GAINS model provides detailed information regarding energy mix and consumption, end-use technology, and emission factors for major air pollutants in each subsector at the provincial level. Based on the ECLIPSE_V5a_CLE scenario, we design SNG allocation scenarios by deploying all planned production of SNG to replace coal in the power sector, industrial sector, and residential sector in turn in proportion to baseline gas required for coal replacement in each subsector. We estimate the provincial anthropogenic emissions under the base case and each scenario accordingly, and assume 2020 emissions within each province follow the same spatial and temporal pattern as that of the 2010 Multi-Resolution Emission Inventory for China (MEIC, http://www.meicmodel.org).

We use the weather research and forecasting model coupled with chemistry, version 3.6 (WRF-Chem v3.6), to simulate air pollutant concentrations for the 2020 base case and for each SNG scenario[43]. The study domain covers East Asia at a horizontal resolution of 27 × 27 km^2 with 31 vertical levels, from the surface to 50hPa, with a 32m thick surface layer. The global 3-D chemical transport model (MOZART-4) with a resolution of 1.9° latitude × 2.5° longitude provides the chemical initial and boundary conditions (http://www.acom.ucar.edu/wrf-chem/mozart.shtml). Meteorological data for 2015 are from the National Centers for Environmental Prediction (NCEP) Global Forecast System final gridded analysis datasets at a 6-

hour resolution (https://www.ncdc.noaa.gov/data-access/model-data/model-datasets/global-forcast-system-gfs). Biogenic emissions are calculated online using the Model of Emissions of Gases and Aerosols from Nature (MEGAN)[44].

To evaluate the health effects of the SNG allocation scenarios, we estimate the changes in premature mortality that are associated with long-term exposure to ambient $PM_{2.5}$ pollution for both adults and children based on the integrated exposure response (IER) functions developed from the Global Burden of Disease studies[27]. Detailed methods are shown in *SI Materials and Methods*.

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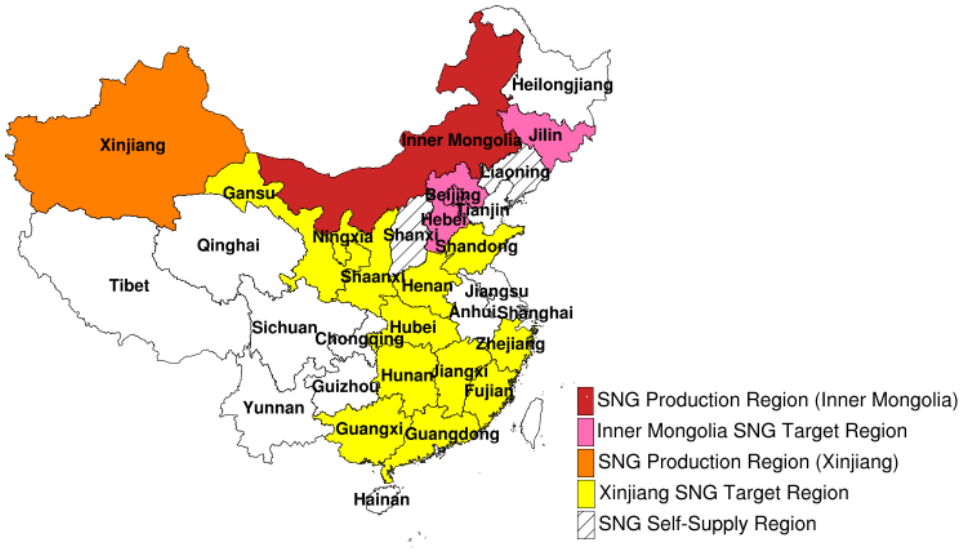


Fig. 1. Map of provinces planning to produce and consume SNG in mainland China in 2020 based on government plans and pipeline infrastructure.

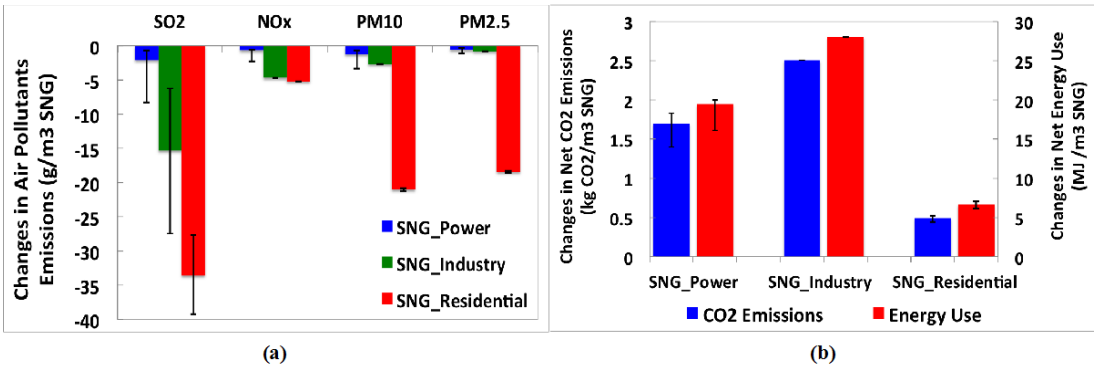


Fig. 2. (a) Decreases in air pollutant (SO_2, NO_x, PM_{10}, and $PM_{2.5}$) emissions, and (b) increases in CO_2 emissions and energy consumption due to SNG substitution for coal. Vertical lines show the range of smallest to largest potential for changes in energy use and air pollutant and CO_2 emissions due to the lower bound (SNG displaces cleanest coal first) and upper bound (SNG displaces dirtiest coal first) substitution scenarios described in Table S1. The industrial sector has no error bar for CO_2 emissions and energy intensity due to the simplifying assumption in the ECLIPSE emission scenario that industrial coal boilers have the same CO_2 emission factor and thermal efficiency (SI 1.2, Table S2).

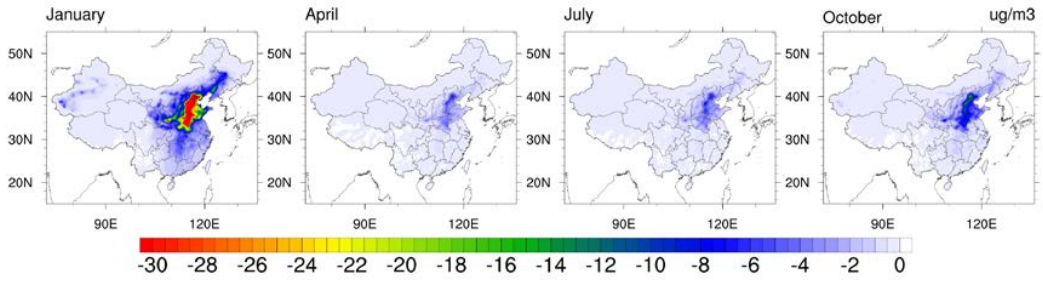
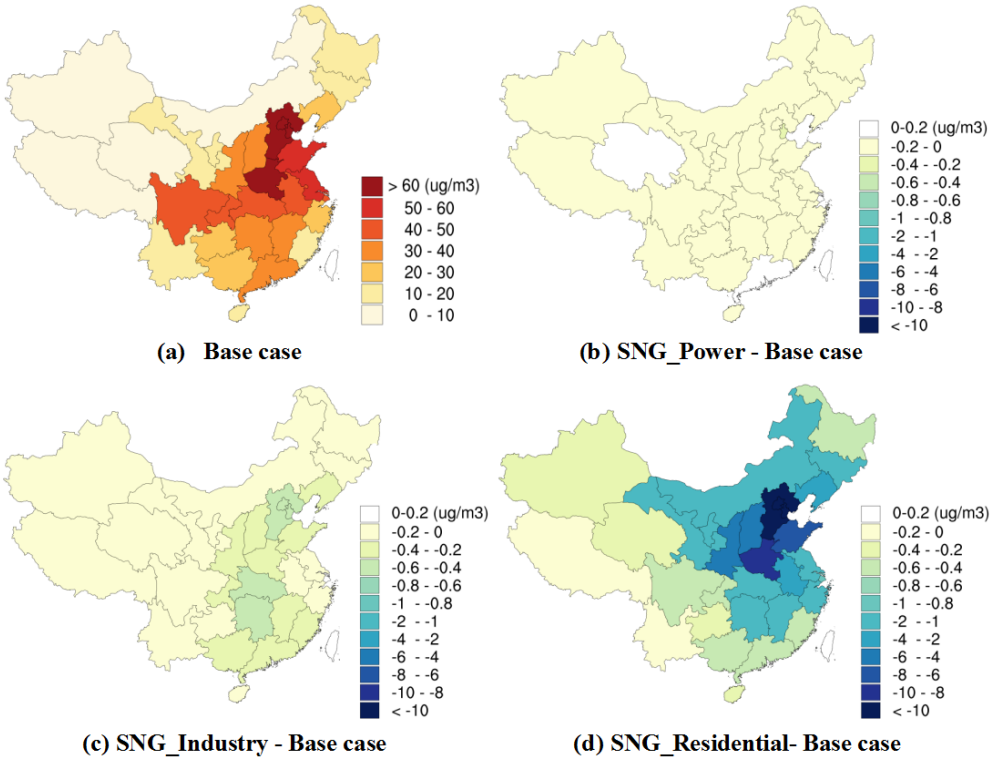


Fig.3. Reductions in simulated 2020 monthly mean surface $PM_{2.5}$ concentrations from SNG substitution for coal in the residential sector (SNG_Residential- Base Case) (Unit: μg/m^3).



(a) Base case

(b) SNG_Power - Base case

(c) SNG_Industry - Base case

(d) SNG_Residential- Base case

Fig. 4. (a) Base case annual average population-weighted (P-W) $PM_{2.5}$ concentrations at mainland China's provincial level. (b-d) $PM_{2.5}$ concentration reductions resulting from SNG deployment under each scenario. Note the non-linear color scale in (b)-(d), which is used to capture regional differences at both the low and high ends of the spectrum of reductions.

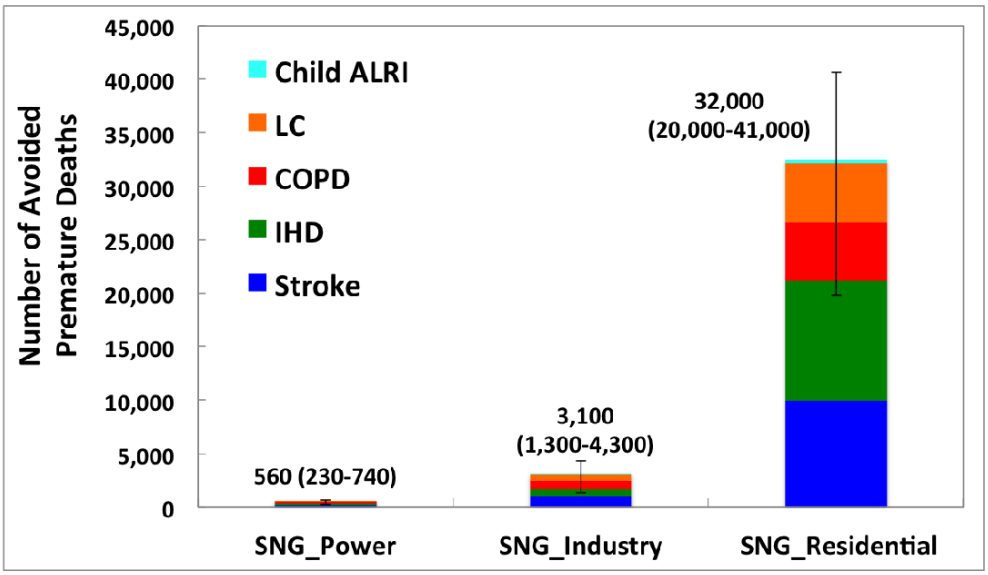


Fig. 5. National total avoided premature mortality by disease type (color) resulting from the replacement of coal with available SNG within each scenario. Child premature deaths due to ALRI are shown together with premature deaths for adults due to LC, COPD, IHD, and stroke. The first value provides the annual mean avoided premature mortality for each scenario and the range in parentheses results from uncertainties in the relative risks for $PM_{2.5}$ exposure as reported by Burnett et al. (2014).

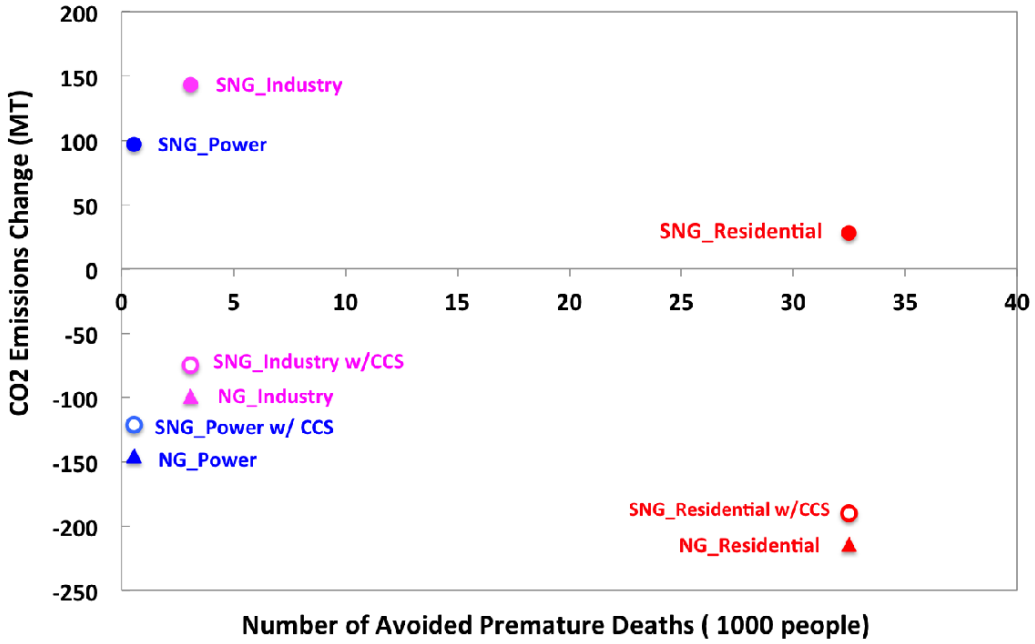


Fig. 6. Comparison of CO_2 emission changes and avoided premature mortality under various SNG allocation scenarios relative to the use of coal. Standard SNG scenarios are shown in the top figure and results that include CCS and the use of conventional natural gas (NG) are shown in the lower figure. Solid circles represent SNG without applying CCS, open circles represent SNG with CCS applied during SNG production. Solid triangles represent the outcome when conventional natural gas is used without CCS. We assume that electricity provided for SNG_CCS is from NGCC power plants using CCS. For results with other types of power plants, refer to SI Section3.

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Chapter 4. The Air, Carbon, Water Synergies and Trade-offs in China's Natural Gas Industry

1. Introduction

Most fossil energy production and combustion processes emit air pollutants and greenhouse gases (GHGs) and also consume significant quantities of freshwater[1-4]. Depending on differences in fuel types, burning conditions, cooling techniques, and existing local environmental stress, energy source choices and end-uses can lead to large variations in resulting air quality, climate, and water impacts[4-6]. Previous studies have concentrated on one or in some cases two, specific environmental impacts in the energy industry[2, 3, 7-10]. Very few analyses have evaluated the air-carbon-water interrelationships of the energy sector, and even fewer have analyzed the nexus from both supply and end-use perspectives[11]. Characterizing the interconnections of various environmental impacts resulting from energy source choices and end-use applications is critical in achieving air, carbon and water co-benefits while avoiding unintended side effects. Here we examine China's natural gas industry to systematically analyze the synergies and trade-offs among the air quality, carbon, and water impacts due to both natural gas source choices (from where natural gas originates) and deployment strategies (in which region and subsector natural gas is used to substitute for coal).

Similar to many emerging economies, China has been facing multiple environmental challenges including domestic air pollution, local water scarcity, and global climate change[12-14]. A coal-dominated energy structure (~64% of primary energy supply in 2015)[15] is partly responsible for all three environmental stresses[1, 10, 16]. In comparison, natural gas is the cleanest fossil fuel, with relatively low carbon intensity and requires less cooling water than coal in most

end uses[4, 16]. Primarily to tackle the haze issue[17], China has been actively promoting a coal-to-gas end-use energy transition[18]. Specifically, China plans to increase natural gas consumption from approximately 6% (~190 billion cubic meters, bcm) of national total primary energy consumption in 2015 to 10% (~360 bcm) in 2020[14, 19]. Until recently, China's natural gas supplies are primarily from domestic conventional gas production (~70%), imported liquefied natural gas (LNG) (~15%), and imported Central Asian pipeline gas (~15%)[20]. To further increase gas supplies, China has planned to develop domestic unconventional natural gas. For instance, China's latest government plans aim to have an annual production of approximately 20 and 30 bcm of domestic coal-based synthetic natural gas (SNG) and shale gas, respectively, by 2020[19, 21]. Meanwhile, China also plans to increase the annual capacity of imported LNG by 38 bcm, as well as increasing pipeline gas from Russia and Central Asia by 38 and 30 bcm, respectively, around 2020[22].

Though substituting conventional natural gas for coal is likely to bring multiple environmental benefits, the air quality, carbon, and water impacts, and their interactions at both aggregated and spatially resolved scales, can vary depending on gas sources[9, 23, 24]. In addition, the magnitude and interdependencies of various environmental impacts of all gas sources can be affected by different end-use deployment strategies. Thus, in this work, we explore the air-carbon-water synergies and trade-offs of both natural gas source choices and end-uses. Our objective is to identify the impacts and the relative importance of gas supply and end-use choices in affecting the air, carbon and water impacts and their interactions.

We use an integrated assessment approach in conjunction with lifecycle analysis to quantify net changes in China's air quality, carbon, and water impacts resulting from a fixed quantity (30 bcm, *SI 1.1*) of gas substitution for coal for each of China's six major gas sources under three

different deployment strategies. Based on government and industrial plans[9, 15, 19, 25, 26], Fig.1 shows, for each gas source, the spatial distribution of China's gas production and the provinces in which each gas source will likely be consumed. For each gas source, we design three coal-to-gas end-use deployment strategies to reflect three different environmental priorities, including 1) air quality-focused substitution (AS), 2) carbon-focused substitution (CS), and 3) water-focused substitution (WS), respectively (Table S1). We first estimate changes in air pollutant emissions, CO_2 emissions, and water stress index (ranging from 0 to 1)[27, 28] weighted water consumption (referred hereafter as 'weighted water consumption') resulting from end-use substitution for each gas source under each deployment strategy. End-use gas substitution for coal consequently requires more natural gas but fewer coal supplies. Both upstream natural gas and coal processes (i.e., production, processing, transmission, and distribution) emit air pollutants and GHGs (CO_{2eq}, including both CO_2 and CH_4) due to energy combustion and methane leakage[23], and consume freshwater for dust suppression, coal washing, well drilling and other purposes[29]. Thus, we further quantify the corresponding environmental impacts due to increases in upstream gas processes and decreases in coal processes. Integrating upstream and end-use processes, we estimate net changes in air pollutant emissions, GHG emissions, and weighted water consumption for 18 combinations of gas source choices and deployment strategies. Using the Weather Research and Forecasting model coupled with chemistry (WRF-Chem v3.6), we further simulate the resulting changes in $PM_{2.5}$ surface concentrations for each gas source under each deployment strategy.

2. Results

2.1 Aggregated air-carbon-water impacts resulting from various natural gas sources

We estimate net changes in lifecycle air pollutant emissions, GHG emissions, and weighted water consumption for upstream and downstream stages of various gas sources. Fig.2 shows results for the air quality-focused substitution, which is China's current primary focus. We observe striking air-carbon/water trade-offs with SNG, but most likely air-carbon-water synergies for all other gas sources.

As shown in Fig.2, end-use gas substitution for coal dominates net changes in air pollutant emissions regardless of gas sources. Despite net air pollutant emission reductions for all gas sources, upstream gas processes lead to substantial net increases in lifecycle GHG emissions and weighted water consumption for SNG. Upstream SNG processes emit roughly 4-7 times more CO_{2eq} than other gas sources. Consequently, SNG substitution for coal increases lifecycle CO_{2eq} emissions by ~20 (40) million tonnes per year (Mt/yr) under the 100-year (20-year) global warming potential (GWP). However, depending on the gas source, substituting coal with the same amount of other gas sources leads to approximately 60 to 120 (70 to 140) Mt/yr of CO_{2eq} emission reductions under GWP_{100} (GWP_{20}) assuming a mean methane leakage rate. Similarly, weighted water consumption from upstream SNG processes is roughly 20-190 times greater than other gas sources, varying depending on which gas source is compared. As a result, SNG leads to annual increases of ~200 million cubic meters (Mm^3/yr) of lifecycle weighted water consumption, while other gas sources result in ~20 to 60 Mm^3/yr of net reductions. In comparison, water consumption due to upstream SNG processes is only ~10-30 times higher than other gas sources (Fig.S2), indicating that SNG production generally occurs in places that are comparatively more water scarce than other gas producing regions. We find gas source choice matters for national carbon and water concerns, primarily because SNG results in

substantial net carbon and water penalties while having similar air quality benefits as other gas sources.

Other than SNG, all gas sources, by substituting for coal, bring net reductions in lifecycle air pollutant emissions, weighted water consumption, and GHG emissions (assuming a mean methane leakage rate). GHG emissions from upstream gas processes (except for SNG) are largely offset by decreases in GHG emissions due to less coal production. This is partly because China's coal industry has high GHG emission intensities due to substantial underground coal mining associated with high methane emissions and a low methane recovery rate[23]. However, without proper methane leakage control from the natural gas industry, coal substitution with gas sources other than SNG, particularly with shale gas, can also result in net increases in lifecycle GHG emissions under both GWPs (Fig.2, Fig.S3, and Fig.S4). Also, our estimated upstream weighted water consumption from shale gas processes is relatively small despite consuming twice as much water as upstream conventional natural gas processes (Table S8). This is because China's existing shale gas development is primarily concentrated in water-abundant Sichuan basin, the location of roughly half of China's total shale gas resources[30]. Nevertheless, as a quarter of China's shale gas resources are located in northern water scarce regions[30], further geographic expansion of shale gas development would likely worsen water stress there.

2.2 Spatially resolved air-carbon-water impacts resulting from various natural gas sources

In addition to evaluating the aggregated environmental impacts, we also explore the spatial characteristics of China's air-carbon-water nexus to characterize the unintended redistributive effects. Fig.3 shows the spatial distribution of net changes in SO_2 emissions, simulated $PM_{2.5}$ surface concentrations, GHG emissions, and weighted water consumption within mainland China for each gas source under the air quality-focused substitution. At the regional level, we find that

all gas sources generally bring air-carbon-water co-benefits in developed eastern China. However, promoting SNG results in substantial carbon-water losses in northwestern China, which already suffer from severe water stress and high per capita carbon intensity[16, 31].

As shown in Fig.3, all gas sources bring net reductions in SO_2 emissions and $PM_{2.5}$ surface concentrations in well-developed eastern China, though there are slight increases in air pollutant emissions and $PM_{2.5}$ surface concentrations in northwestern provinces primarily due to SNG or conventional gas production. For each gas source, the largest $PM_{2.5}$ concentration reductions are primarily concentrated in regions where substitution occurs. Although SNG results in only slight increases in $PM_{2.5}$ surface concentrations in northwestern provinces, it leads to substantial increases in GHG emissions and weighted water consumption in northwestern SNG producing provinces. Notably, these regions also suffer from severe water scarcity and have high per capita carbon emissions, due to their coal-dominated energy mix and substantial export of electricity to eastern China, such as Beijing and Tianjin[16, 31].

2.3 Comparison of the air-carbon-water impacts resulting from natural gas source choices and end-uses

Besides gas source choices, sectoral and regional gas deployment strategies can also lead to large variations in the air quality, carbon and water impacts of gas substitution for coal. To fully capture the synergies and trade-offs in the natural gas industry, here we integrate six major gas source choices and three end-use deployment strategies that affect the air-carbon-water interdependencies.

We find that gas source choice is the single most important factor in shaping the air-carbon-water nexus of the natural gas system, primarily because SNG clearly stands out because, unlike other gas sources, it worsens carbon emissions and water stress regardless of end-use deployment

strategies (Fig.4). That is, SNG causes net increases in GHG emissions and weighted water consumption even under the deployment strategies that aim at achieving the largest reductions in GHG emissions (CS) or weighted water consumption (WS), respectively (Fig.S3 and Fig.S4).

However, for gas sources other than SNG, end-uses determine the magnitude of the air, carbon, and water impacts. We find that within the same gas source (except for SNG), different *deployment* strategies can result in approximately 10-50 times, 1.5-1.6 times, and 2-9 times differences in China's population-weighted (P-W) $PM_{2.5}$ surface concentrations (*SI 1.3*), lifecycle GHG emissions (GWP_{20}), and China's weighted water consumption (Fig.4). In comparison, different gas *sources* lead to only roughly 1-4 times, 1-1.9 times, and 1-3 times differences, respectively, within the same deployment strategy. Thus, gas deployment strategies usually play a more important role than gas source choices in determining the environmental impacts of non-SNG natural gas, particularly on air quality.

Additionally, gas deployment strategies are the key in determining the air-carbon-water interconnections for gas sources other than SNG. We note substantial air-carbon co-benefits but air-water trade-offs due to end-use gas substitution for coal. Specifically, AS leads to ~ 0.6 to 1.8 $\mu g/m^3$ annual average P-W $PM_{2.5}$ surface concentration reductions, but merely ~20 to 60 Mm^3/yr of weighted water consumption reductions. In comparison, depending on the gas source, WS results in ~ -0.004 to 0.08 $\mu g/m^3$ P-W $PM_{2.5}$ mean surface concentration reductions, but ~90 to 200 Mm^3/yr of weighted water consumption reductions. That is, for the same gas source, AS results in over an order of magnitude greater P-W $PM_{2.5}$ surface concentration reductions than WS. However, WS results in approximately 2-8 times as much weighted water consumption reductions as AS. In comparison, CS generally leads to similar levels of air quality, water stress and carbon emission changes as AS. We find CS actually results in slightly higher net reductions

in P-W $PM_{2.5}$ surface concentrations than AS. This is mainly because the least efficient coal combustion happens to be the dirtiest, indicating potential air-carbon co-benefits. Thus, when natural gas is deployed primarily to tackle the air pollution issue, China's current major focus, it will in most cases bring substantial carbon reduction co-benefits, but have negligible water benefits. However, if natural gas were to be allocated to mainly address water scarcity concerns, it will in most cases only slightly improve air quality, though it will still bring notable carbon reductions. Therefore, there is a fundamental air-water trade-off due to end-use gas-for-coal substitution.

To put our estimated environmental impacts in perspective, we also show the percent changes for each impact resulting from each gas source and end-use choice. As shown in Fig.S9, 30 bcm of natural gas, by substituting ~1.5% to 2.2% of total coal consumption, can lead to -4.8% to 0.02%, -1.8% to 0.4%, and -0.07% to 0.08% of net changes in China's P-W $PM_{2.5}$ surface concentrations, lifecycle GHG emissions (GWP_{20}), and China's weighted water consumption. Our estimated percent contributions may be scaled up or down depending on actual increases in natural gas supplies. However, the relative trends across gas sources and end-uses in affecting various environmental impacts can be illustrative. Fig.S9 again demonstrates the determining role of SNG in causing the air-carbon/water trade-offs, as well as substantial variations in the resulting environmental impacts (air quality in particular), primarily due to end-uses.

3. Discussion

Our study demonstrates that with careful natural gas source choices and end-use designs, switching from coal to natural gas is most likely to bring air quality, carbon, and water co-benefits, though with notable air-water competing effects. However, gas source choices can be a determining factor in changing this picture due to coal-based synthetic natural gas. Upstream

SNG processes, in particular SNG production, significantly increase both water stress and carbon intensity in regions (northwestern China) already suffering from severe water scarcity and high per capita carbon intensity due to energy export to eastern China[16, 27]. Therefore, despite that end-use SNG substitution for coal reduces air pollutant and CO_2 emissions and usually decreases water consumption, SNG not only leads to lifecycle carbon and water increases for China as a whole, but also exacerbates existing environmental injustice caused by the energy industry[16]. Therefore, our results clearly show a spillover effect of China's "Clean Air Act": focusing on improving air quality in the well-developed eastern provinces may unavoidably impose environmental pressure on the less-developed northwestern provinces when substituting SNG for coal. Hence, rich regions may increase technology investment in less-developed regions to eliminate their outsourcing effects. In addition, an earlier study found that conducting carbon capture and storage (CCS) during SNG production could bring air quality and carbon co-benefits when substituting SNG for coal[9]. However, development of CCS will further increase water demands in northwestern regions due to water consumption for CO_2 scrubbers and to compensate for energy efficiency losses[32, 33]. Considering that energy infrastructure typically operates for multiple decades, our finding indicates that it is highly valuable to develop energy infrastructure with an understanding of the air-carbon-water interconnections to avoid unintended side effects at both regional and global levels.

For coal substitution with natural gas sources other than SNG, we find that regional and sectoral end-use gas deployment usually plays a far more important role in determining the magnitude of resulting local air quality (~10-50 times VS. ~1-4 times of variations in China's P-W $PM_{2.5}$ surface concentration reductions) and water (~2-9 times VS. ~1-3 times of variations in China's weighted water consumption reductions) impacts in particular, and carbon impacts

(~1.5-1.6 times VS. ~1-1.9 times of variations in lifecycle GHG emission reductions under GWP_{20}) in most cases, than energy source choices. Existing discussions have largely focused on clean energy source choices[8, 34]. However, this study shows that more attention is needed on designing clean energy deployment strategies, as end-use choices are highly influential in determining net environmental impacts.

Our study also illustrates notable air-water trade-offs due to end-use sectoral and regional natural gas deployment. This trade-off is caused by both the geographic mismatch of air pollution and water stress regions, as well as the sectoral differences in affecting various environmental impacts (Fig.S7). Particularly, under WS, natural gas is primarily distributed to the power sector to substitute for coal (Fig. S8a). This allocation can significantly reduce water consumption but bring only tiny air quality benefits due to widely employed end-of-pipe control technologies in coal-fired power plants[9]. Likewise, when natural gas substitution for coal primarily occurs in the residential sector to tackle the air pollution issue, where coal burning has the highest emissions[9], it does not reduce water consumption. Besides, when a higher portion of gas is allocated to highly water-stressed provinces under WS, it does not necessarily bring large air pollutant reductions because high water stress and severe air pollution regions do not always overlap (Fig.S8b). Notably, this intrinsic air-water trade-off identified in our study not only exists for natural gas in China, but also for other clean energy sources (i.e. deploying wind or solar in a clean power sector will bring only limited air quality benefits) and in other countries (i.e. a geographical mismatch between air pollution and water stress exists in many other countries such as India). We find it necessary to deploy a combination of energy technologies to simultaneously significantly address air quality, climate, and water concerns.

Globally, there are large uncertainties in methane leakage rates from upstream natural gas processes. This can potentially make gas source choices a more important factor in affecting net carbon impacts than what we identify here. Field measurements of methane leakage along the whole lifecycle chain of the natural gas industry both within and outside China will improve understanding of the carbon impacts of China's natural gas industry and the relative importance of gas source choices and deployment strategies.

The air-carbon-water nexus discussed in this study focuses on China's natural gas industry. Due to its enormous economy and population, China's energy plans have both important global and domestic implications. The framework described here and its qualitative conclusions could be applied to other countries and regions as they design energy transition pathways.

4. Methods

This work uses an integrated assessment approach coupled with lifecycle analysis to evaluate the air-carbon-water nexus of China's natural gas industry to prioritize both gas source choices and end-use deployment strategies. The air quality, carbon, and water impacts are reflected as changes in lifecycle air pollutant emissions and simulated $PM_{2.5}$ surface concentrations, in lifecycle greenhouse gas (GHG) emissions, and in water stress index (WSI) (the ratio of total annual fresh water withdrawal to hydrological availability, ranging from 0 to 1)[27, 28] weighted water consumption (regional actual water consumption × region-specific water stress index, referred hereafter as 'weighted water consumption').

We use the ECLIPSE_V5a_CLE (Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants) emission scenario developed by the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) model as our 2020 base case anthropogenic emissions input[35]. The ECLIPSE scenario is designed to reflect provincial energy uses and emissions in

China's 12th five-year-plan[35], and it provides detailed subsector technology information, energy consumption data, and emissions of major air pollutants and CO_2 at China's provincial level. In addition, we integrate China's provincial-level cooling technology information with end-use technology data provided by the GAINS model to evaluate water impacts (*SI Methods*).

We focus on six of China's major natural gas sources, including domestic conventional natural gas, domestic coal-based synthetic natural gas (SNG), domestic shale gas, imported liquefied natural gas (LNG), imported pipeline gas from Russia, and imported pipeline gas from Central Asia. For each gas source, the regional deployment is determined by governmental and industrial plans as shown in Fig.1. At the sectoral level, we focus on natural gas substitution for coal in three major sectors: industry, residential, and power[9].

Based on different environmental priorities, we design three natural gas end-use deployment strategies for each gas source: 1) air quality-focused substitution (AS), designed to achieve the largest reductions in SO_2 emissions during end-use substitution; 2) carbon-focused substitution (CS), designed to achieve the largest reductions in CO_2 emissions; and 3) water-focused substitution (WS), designed to achieve the largest reductions in weighted water consumption (Table S1).

As a thought experiment, with each combination of gas source and end-use, we assume an additional gas supply of 30 bcm above the baseline to replace coal. This is roughly the quantity of China's currently planned increase for most gas sources in 2020[19, 21, 22]. We then estimate the resulting changes in air pollutant emissions, CO_2 emissions, and weighted water consumption due to end-use gas substitution for coal. Additional gas supply leads to an increase in upstream natural gas production, and a decrease in upstream coal production. This results in emission and water consumption changes from upstream natural gas and coal processes (i.e., production,

processing, transmission, and distribution) due to energy combustion, methane leakage, and water uses for coal washing, well drilling and so forth[23, 29]. We quantify upstream emissions and weighted water consumption changes using stage-level energy consumption data and methane leakage rates summarized in Qin et al. (2017)[36], country-specific emission factors from the GAINS model, and fuel-specific water consumption rates (Table S9) and regional water stress indexes provided by Pfister et al. (2009)[28] (country-level WSI for regions outside China) and Feng et al. (2014)[27] (provincial-level WSI for regions within China). We assume upstream emissions and water consumption for gas processes occur in places where natural gas is produced. In comparison, the spatial distribution of reduced emissions and water consumption due to avoided upstream coal processes are identified according to where end-use coal reduction occurs and the corresponding source-receptor matrix of coal production and consumption[37].

Combining both upstream and end-use processes, we estimate net changes in air pollutant emissions, GHG emissions, and weighted water consumption for each gas source under each coal-to-gas deployment strategy. We then use the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem v3.6) to simulate resulting changes in 2020 $PM_{2.5}$ surface concentrations. Method details are summarized in the supplementary information (*SI Methods*).

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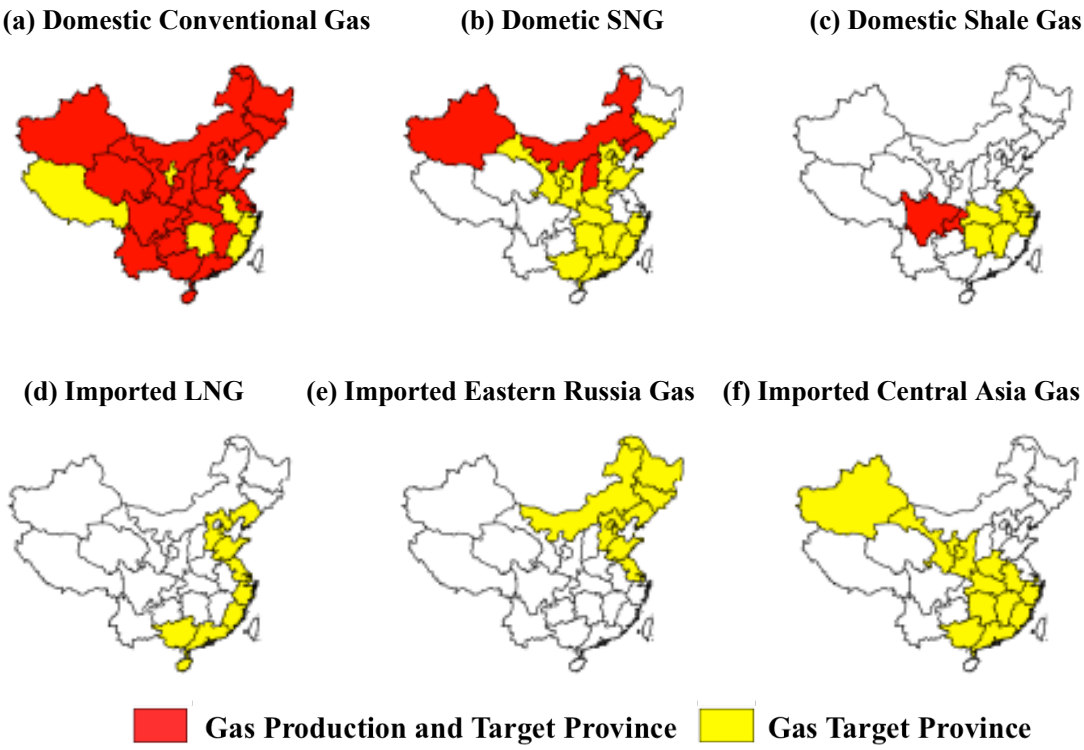


Fig. 1 Production and potential consumption regions for mainland China's six major natural gas sources based on government and industrial plans[9, 15, 19, 25, 26]. Imported LNG are mainly produced in Qatar, Australia, Indonesia, and Malaysia. Imported Eastern Russia Gas and Central Asia Gas are produced in Russia and Central Asian countries (Turkmenistan, Kazakhstan, and Uzbekistan), respectively.

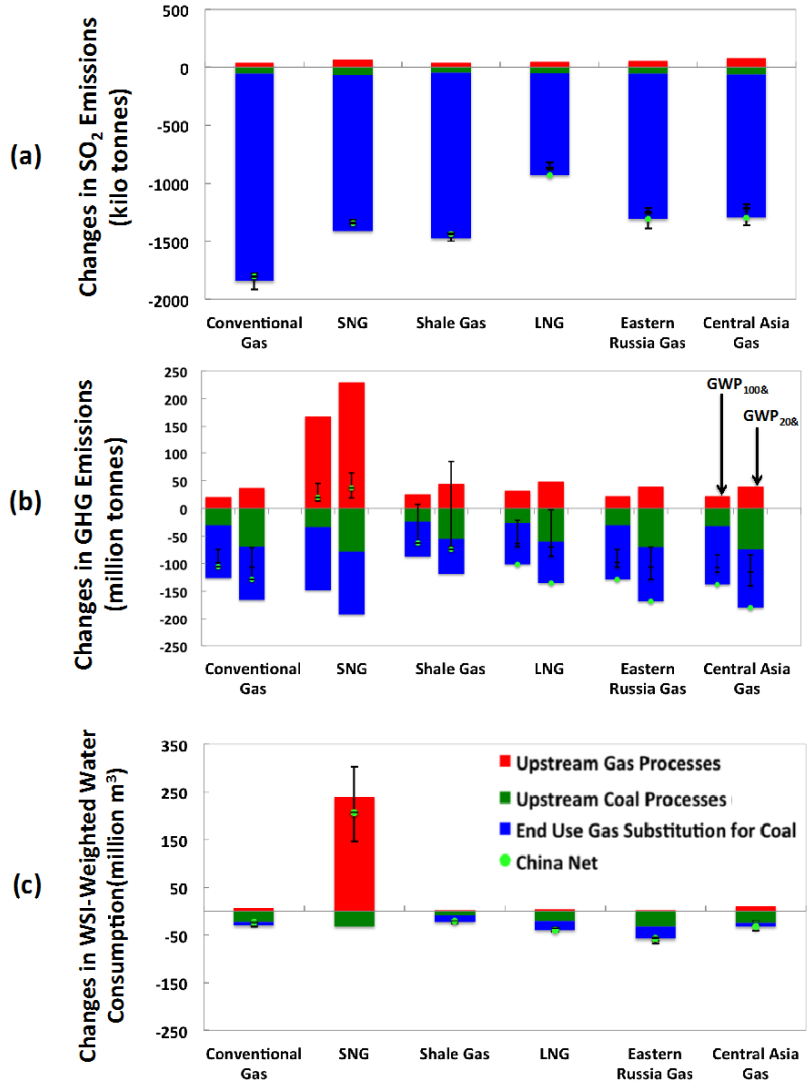


Fig. 2 Air quality-focused substitution (AS). Changes in upstream and downstream SO_2 emissions, greenhouse gas (CO_{2eq}, including both CO_2 and CH_4) emissions, and weighted water consumption for coal substitution by 30 bcm of gas from various sources. China net values are obtained by considering emission or weighted water consumption changes occurring within China's border from upstream gas processes (upstream gas production, processing, transmission and distribution), upstream coal processes (upstream coal production, processing and transport) and end-use gas substitution for coal. Error bars represent emission or weighted water consumption changes resulting from mean, lower bound, and upper bound estimates (mainly due to methane leakage rates) of coal and each gas source occurring both within and outside of China. Figures for carbon-focused (CS) and water-focused (WS) coal-to-gas substitution are included in the SI (Fig. S3 and Fig. S4).

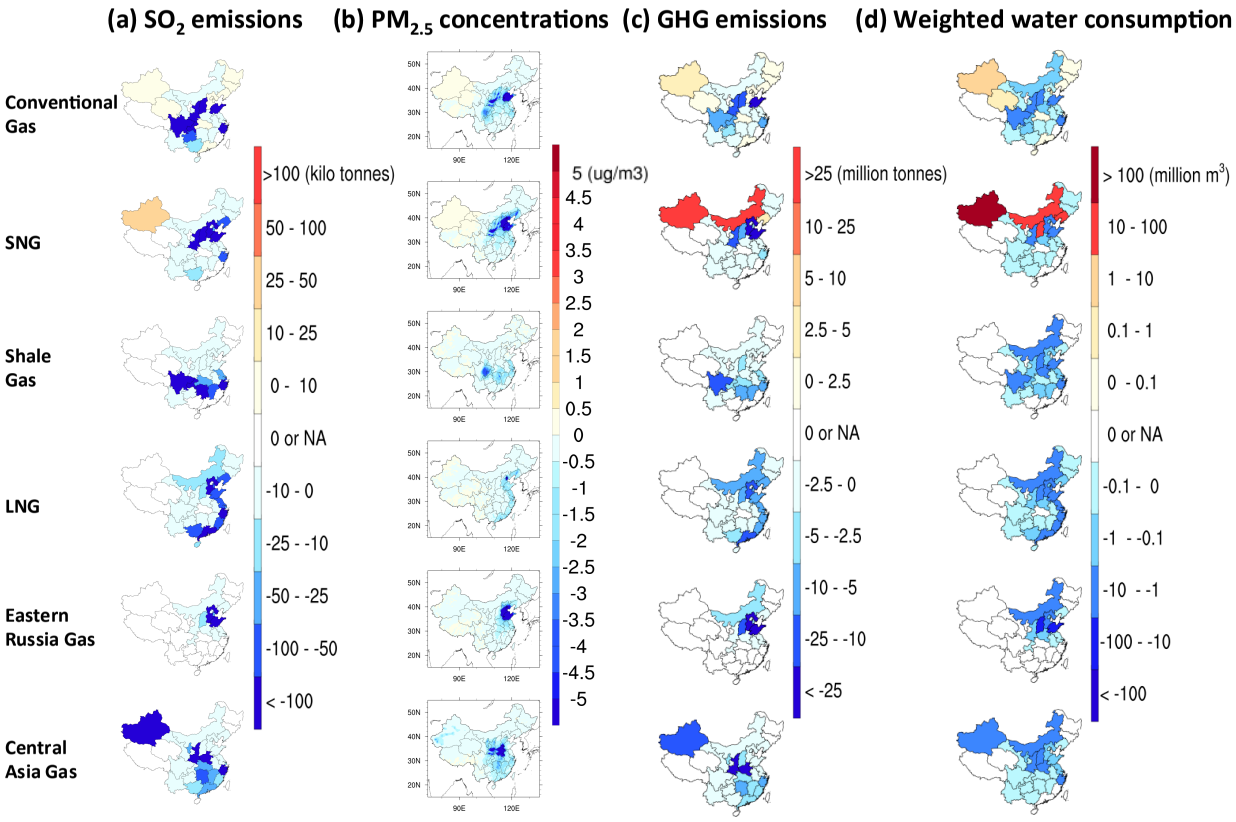


Fig. 3 Air quality-focused substitution (AS). Mainland China's spatially resolved changes in air (SO_2 emissions and population-weighted $PM_{2.5}$ surface concentrations), carbon (lifecycle greenhouse gas emissions under GWP_{20}, including CO_2 and CH_4, assuming mean methane leakage rates), and water (weighted water consumption) impacts of coal substitution by 30 bcm of gas from various sources. Changes in SO_2 emissions, GHG emissions, and weighted water consumption are shown at the provincial level; changes in $PM_{2.5}$ concentrations are shown at the grid level (27 km×27 km). Results for the carbon-focused (CS) and water-focused (WS) coal-to-gas substitution are shown in the SI (Fig.S5 and Fig.S6)

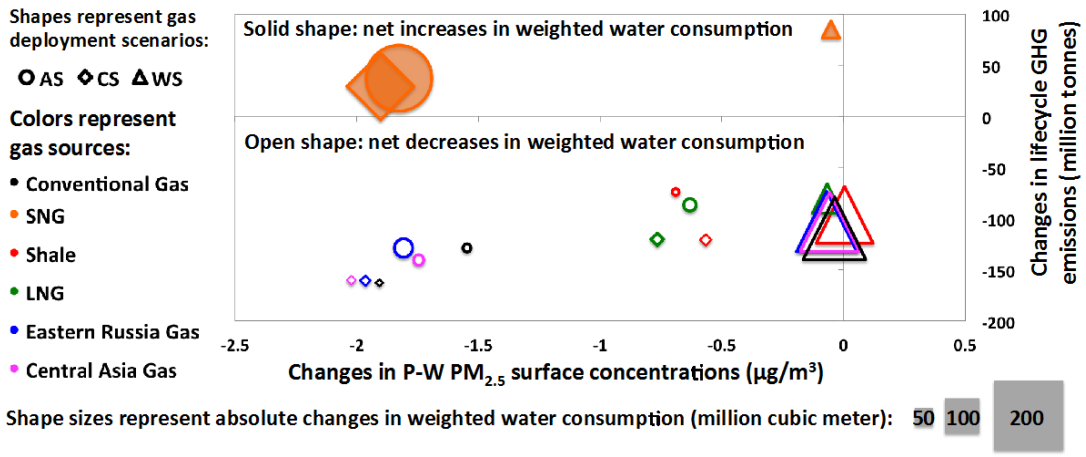


Fig. 4 Comparison of net changes in air quality (China's population-weighted $PM_{2.5}$ surface concentrations), carbon (lifecycle greenhouse gas emissions under GWP_{20}, assuming mean methane leakage rates), and water impacts (China's weighted water consumption) of coal substitution by 30 bcm of gas from various sources under three deployment strategies. Negative changes in $PM_{2.5}$ surface concentrations, lifecycle GHG emissions, and weighted water consumption represent an improvement in air quality, carbon, and water impacts, and vice versa. AS: air quality-focused substitution, CS: carbon-focused substitution, WS: water-focused substitution. Solid shapes represent net increases in weighted water consumption, and open shapes represent net decreases in weighted water consumption. Detailed breakdowns can be found in additional figures in the SI (Fig.S10).

Chapter 5. Conclusion

China's coal-dominated energy structure has been partly responsible for the multiple environmental challenges it is facing, such as air pollution, water stress, and climate change. Designing a suitable energy transition pathway that can tackle these environmental problems is critical. Natural gas is likely to play an important role in China's energy transition at least in the following several decades before renewables can massively replace coal. However, depending on natural gas source choices and deployment strategies, substituting coal with natural gas may not be able to significantly alleviate all environmental stresses we are concerned about. For instance, in Chapter 2, we show that without adequate methane leakage control, replacing coal with shale gas can potentially result in higher upper bound lifecycle greenhouse gas emissions than coal. However, government and industries can take actions to ensure net carbon reductions via green completions, complete flaring of flowback gas, and so forth. In addition, in Chapter 3, we show that deploying natural gas to the residential sector, where coal consumption is dirty and inefficient, brings the largest air quality and associated human health benefits with the smallest carbon penalties. In Chapter 4, we further integrate all major gas sources and deployment strategies focusing on three environmental objectives to identify the air-carbon-water interactions. We find that various natural gas sources bring similar degrees of air quality improvement. However, climate and water impacts are mainly determined by gas source choices, primarily due to coal-based synthetic natural gas (SNG). Our study suggests that China may consider avoiding large-scale SNG projects development concerning the overall air quality-climate-water impacts. For gas sources other than SNG, end-use deployment strategy is the key in determining the magnitude of air, carbon, and water impacts and the associated interactions. However, the intrinsic air-carbon co-benefits but air-water trade-offs suggest that switching from coal to gas

alone will not be able to substantially address multiple environmental challenges simultaneously. Concerning China's current major focus is tackling air pollution, other technologies, dry cooling, for instance, are needed to compensate for the air-water trade-offs.

Notably, after U.S. withdrawal from the Paris agreement, high expectations developed that China would take the lead in addressing climate change. As I have already shown in this dissertation, substituting natural gas for coal in China may provide a good opportunity to reduce emissions of greenhouse gases. However, to realize the intended climate benefits, government and industry must resolve the fundamental price dilemma in China's natural gas market, balance seasonal gas supply and demand, and minimize gas leakage. Some of the challenges are also applicable to the other two environmental issues.

China's rapid economic growth in the past three decades has come along with substantial increases in national energy consumption and CO_2 emissions. Currently, China, with 19% of global population, contributes 16% of global gross domestic product (GDP) and 30% of global CO_2 emissions[1]. China, the world's largest CO_2 emitter, pledged to peak national CO_2 emissions by 2030 using strategies such as fuel switching, energy efficiency improvements, and increases in carbon sinks[2]. Substituting natural gas for coal is identified as a promising carbon mitigation option[2]. In China's Nationally Determined Contributions (NDC), natural gas will account for over 10% of national primary energy consumption by 2020[2]. However, we find large uncertainties in realizing the desired climate benefits of the coal-to-natural gas transition due to challenges in expanding natural gas supplies, encouraging coal-to-gas substitution, balancing seasonal gas supply and demand, and ensuring a lower lifecycle carbon footprint for natural gas than coal.

China's natural gas supply and demand

Geologically, China has abundant coal reserves but is poor in gas. In 2015, natural gas use reached just 6% of national total primary energy consumption, compared with ~65% from coal[3]. Natural gas and coal can both be used in the industrial, residential, and power sectors[3]. To increase gas supplies, the Chinese government has actively promoted exploration and production of domestic unconventional natural gas. For instance, the central government has planned for an annual production by 2020 of approximately 30-100, 30-40, and 20-60 billion cubic meters (bcm) of shale gas, coalbed methane (CBM), and coal-based synthetic natural gas (SNG), respectively[2,4]. Meanwhile, China has been actively seeking natural gas imports from neighboring countries. According to existing contracts, China's natural gas import capacity in 2020 will increase to 85 bcm/yr (via pipelines from Central Asia), 38 bcm/yr (via pipelines from eastern Russia), and 88 bcm/yr (via liquefied natural gas from Qatar, Australia, Malaysia, and Indonesia) [5,6].

In Figure 1, we summarize historical gas supplies and near-term production or import goals for China's major natural gas sources. We find that despite strong political interest in increasing natural gas supplies, domestic unconventional natural gas production and realized natural gas imports are consistently below the government's targets[2-5]. Furthermore, the Chinese government has continually decreased the production targets for all types of unconventional gas. For instance, the central government initially planned for an annual shale gas production of 60-100 bcm by 2020, which was reduced to 30 bcm four years later (in 2016)[2,4].

Slower-than-expected domestic unconventional natural gas development can partly be explained by constraints such as unfavorable geology (i.e., insufficient water availability and great reservoir depth) and immature technology[7,8]. However, the market barriers caused by government-regulated natural gas prices may play a more important role in affecting both

unconventional gas development and gas imports. Specifically, as the government fears that high gas prices may increase residents' cost of living and decrease domestic industries' competitiveness, natural gas prices, particularly in the residential sector, are usually set lower than the cost of gas imports and unconventional gas production. This, in turn, has caused substantial economic losses to the three national oil companies (NOCs) that essentially control the production, import, and transport of China's natural gas[2]. Annual subsidies provided by the government to the NOCs only partly compensate for the economic losses. Thus, this has unequivocally dis-incentivized industrial investments in expanding natural gas imports or unconventional gas development. Without strong economic motivation, a reliable increase in natural gas supplies from the oligopoly market may not be forthcoming.

On the other hand, regulated natural gas prices are still not competitive with cheap coal, which has hindered large-scale coal-to-gas end use transitions. Essentially, China is facing a fundamental price dilemma in the natural gas market: whether to increase gas prices to encourage natural gas supplies or to decrease gas prices to expand natural gas end uses. Facing this dilemma, China's National Development and Reform Commission (NDRC) has frequently adjusted natural gas prices both up and down in recent years[2]. Inconsistent price signals further delay and reduce industrial investments on the much-needed gas pipeline infrastructure, which plays an important role in expanding China' natural gas market due to the geographic mismatch between major gas demands (southern and eastern China) and major gas supplies (western China).

Additionally, contrary to the relatively constant year-round natural gas supply, we note an increasing variation in seasonal gas demands primarily driven by winter heating gas needs. For instance, Beijing's gas demand was 6-8 times higher in winter than in summer 2016[4]. China's

current national gas storage accounts for merely 3% of annual gas consumption, far below the global average of 16%[9]. The lack of natural gas storage facilities will make it increasingly challenging to balance gas supply and demand throughout the year.

Carbon footprint of using natural gas to meet end uses

Compared to coal, when combusted, natural gas emits approximately 50% less CO_2 per unit of energy output[10]. Thus substituting natural gas for coal is considered a bridging solution to mitigate climate change[10]. However, natural gas may have a larger-than-coal lifecycle carbon footprint if substantial methane leakage occurs in the natural gas system[11]. In Fig.2, we estimate changes in CO_2 and CH_4 emissions if all natural gas produced in 2010, 2020, and 2030 (using mean government targets) is assumed to replace coal in a specific end use sector (power generation, residential cooking, and industrial heat generation). Such hypothetical allocation can indicate the sign and magnitude of emission changes within each sector resulting from a coal to gas transition.

As shown in Fig. 2, a higher natural gas consumption from 2010 to 2030 does not necessarily mean greater reductions in greenhouse gas (GHG) emissions. This is partly due to large variations in lifecycle emissions among different natural gas sources. First, if all gas consumption were SNG, it would have increased China's 2010 CO_2 emissions (~9 billion tonnes)[1] by 2-4%, and could increase China's predicted 2020 (~12 billion tonnes) and 2030 (~14 billion tonnes) CO_2 emissions[12] by 4%-10% and 4%-11%, respectively. Thus, a significant use of SNG is inconsistent with China's commitment to peak CO_2 emissions on or before 2030[2]. In contrast, assuming all gas consumption were conventional gas, 2010 CO_2 emissions would have been reduced by 1%-4%, and could lead to 4%-9% and 4%-11% reduction in 2020 and 2030 CO_2 emissions, respectively. Imported gas and shale gas may have slightly lower net CO_2

emission reductions than conventional gas due to larger energy consumption for long distance transport, liquefaction and regasification, and hydraulic fracturing[11,13]. Nevertheless, with the exception of SNG, substituting natural gas for coal facilitates China's efforts to accomplish its NDC commitment by reducing CO_2 emissions. However, this does not guarantee net climate benefits because the NDC commitment does not include methane emissions from the natural gas industry. As we show in Fig.2, only with a low methane leakage rate, can natural gas help China both achieve its NDC commitment and result in net GHG emission reductions through substituting for coal. Finally, we note substantial differences in emission impacts across end uses. Natural gas use in the residential sector is likely to bring larger GHG reductions than use in other sectors.

Policy Implications

To reduce the uncertainties in using natural gas as a carbon mitigation option in China, political and industrial efforts are needed to address the fundamental price dilemma in China's natural gas market, to increase investments in natural gas infrastructure, and to reduce the lifecycle carbon footprint of China's diversifying gas sources.

Recent efforts to gradually lift natural gas price controls, such as establishing the Shanghai natural gas hub, are positive signs for the start of a market-based pricing mechanism[2]. When gas prices reflect the actual production or import cost, this boosts industry confidence and encourages investment to expand natural gas supplies and gas-related infrastructure (especially the under-developed natural gas pipelines and storage facilities). However, natural gas price reform alone cannot fully resolve the price dilemma. A reasonably high carbon price is needed to increase the price competitiveness of natural gas over coal and to encourage natural gas end uses. We note that China plans to establish a national carbon trading market this year (2017) based on

seven provincial pilot carbon markets started in 2011[4]. Nevertheless, China's current carbon prices (US $1-15/tonne CO_2) will likely need to be increased to fully capture the carbon advantage of natural gas over coal[14].

Poor data availability on methane leakage is a major challenge in evaluating the carbon footprint of natural gas. We recommend the Chinese government consider monitoring methane leakage from natural gas systems and that they promote best practices following U.S. experiences. Natural gas sources other than SNG can facilitate the achievement of China's NDC by substituting for coal. However, from the climate change perspective, methane leakage from the natural gas system will partly offset China's CO_2 mitigation efforts. Therefore, we emphasize the need for the next round of NDC to include methane leakage from the oil and gas industry to ensure effective carbon mitigation efforts.

Nevertheless, increased use of natural gas can bring substantial co-benefits in other areas. The Chinese government has initiated nationwide efforts to replace coal with natural gas to reduce air pollution and if deployed in the residential sector would bring large air quality benefits to the North China Plain in winter [2,4,15]. Greater natural gas availability in the future would help smooth the intermittency of renewable electricity generation. Assuming the uncertainties discussed above are carefully addressed, natural gas can play an important role in facilitating China's low-carbon transition, which may boost global confidence in addressing climate change.

To sum up, the energy-environment nexus will play an essential role in determining the world's future development path. Meanwhile, both energy production and consumption can result in environmental challenges in multiple aspects. Thus, it is critical to integrate air quality, climate, and water concerns in designing energy transition pathways. In addition to direct impacts resulting from gas source choices and deployment strategies, the market and political

influences can be equally, if not more, important. Besides, the whole world is moving towards decarbonization to avoid dangerous climate change. Thus, future work in the natural gas industry should focus more on how would natural gas facilitate on-grid integration of renewables.

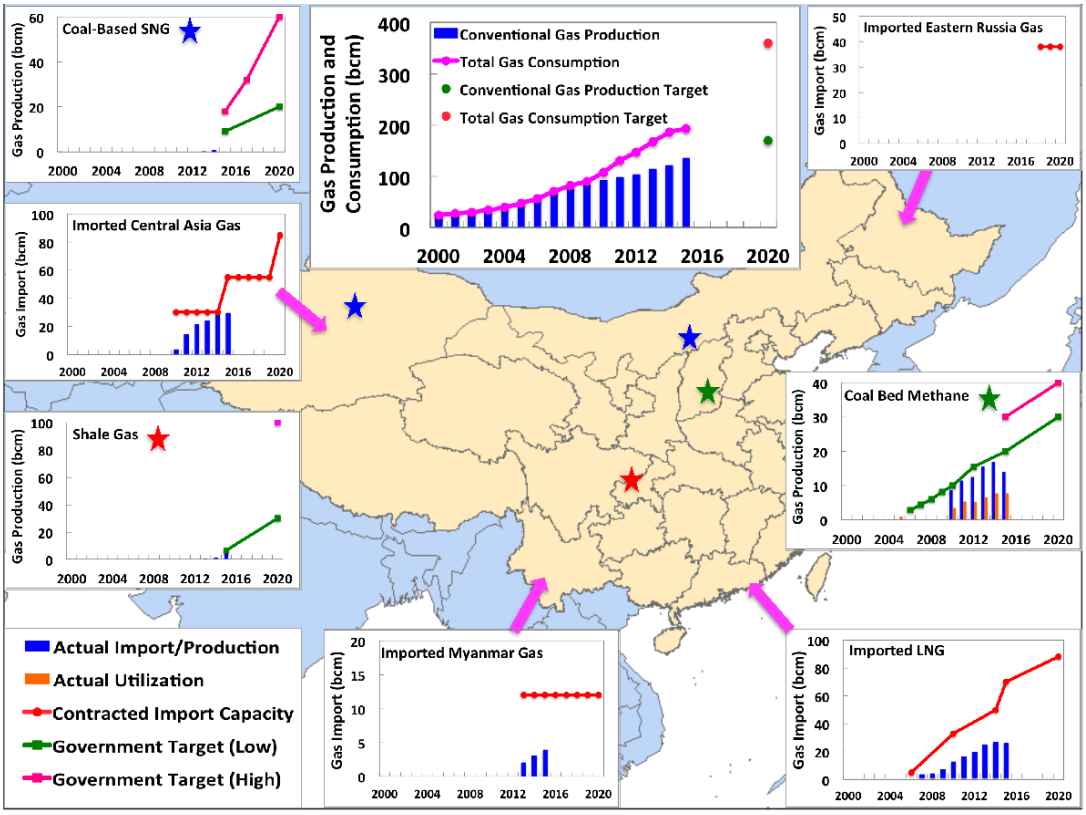


Figure 1. Historical and planned natural gas supplies from China's major natural gas sources. Information is gathered from government documents (for government targets)[2,4], statistical database (for historical gas production and consumption)[3], literature[5], and news articles (when no better information is available). Representative production regions and import locations for major gas sources are also illustrated in the map.

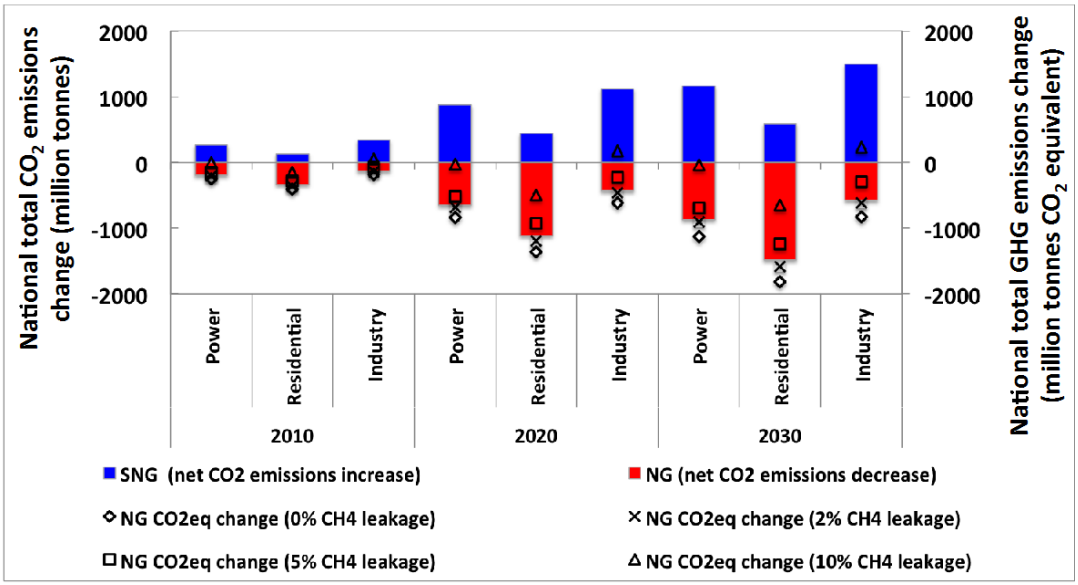


Figure 2. Changes in China's CO_2 (shown in colored bars on the left axis) and greenhouse gas (CO_2 and CH_4) (shown in black markers on the right axis) emissions when 108 billion cubic meters (bcm) (2010 gas consumption), 360 bcm (2020 consumption goal)[2], and 480 bcm (2030 consumption goal)[2] of coal-based synthetic natural gas (SNG) or conventional natural gas (NG) substitute for coal in 2010, 2020, and 2030. Here we assume all natural gas is either used for power generation (replacing supercritical coal-fired power plants), residential cooking, or industrial heat generation. We consider multiple methane leakage rates from the natural gas system and use the 100-year global warming potential. The figures are calculated based on findings in Qin et al[11,15].

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Appendix for Chapter 2.

Can Switching from Coal to Shale Gas Bring Net Carbon Reductions to China?

1. Method Details

1.1. Estimating Upstream GHG Emissions for Conventional Natural Gas in China

Within the lifetime system boundary we set (Table S1), greenhouse gas (GHG) emissions occur from all upstream stages, including natural gas 1) (pre-) production & gathering, 2) processing, 3) transmission & storage, 4) distribution, as well as from 5) combustion in end uses. In each stage, methane leakage occurs. In addition, energy is needed to run the system, thus also generates CO_2.

The most recent literatures estimating GHGs from China's natural gas industry essentially used the same methane leakage rates as reported in the 2006 IPCC guidelines (log-mean values) due to a lack of updated domestic measurements.[1-3] In contrast, U.S. researchers have significantly improved methane leakage estimates from the natural gas industry in the U.S. based on field measurements in recent years, which are much higher than reported in the 2006 IPCC guidelines.[4-8] Considering conventional natural gas industry in both China and the U.S. are long-standing businesses, we apply the conventional gas methane leakage rates reported in the latest U.S. field studies to China as the "most likely" (mean) case, to take advantage of the U.S. recent measurement and estimation efforts.

Allen et al. (2013)[5] reported CH_4 emissions from direct measurements at 190 onshore natural gas production sites in the U.S. Similarly, Marchese et al. (2015)[6] and Zimmerle et al. (2015)[7] measured CH_4 emissions from 130 natural gas gathering & processing facilities, and 2292 transmission & storage facilities, respectively. Lamb et al. (2015)[8] measured CH_4 emissions for 459 natural gas distribution pipelines, and metering & regulating facilities together. These

authors also estimated national total methane emissions for each stage of the natural gas industry. Estimates of national total CH_4 emissions in each stage from these studies are based on direct measurements of fugitive EFs and EPA reported activity data in a certain year. We report the corresponding stage-level methane leakage rates via normalizing the estimated CH_4 emissions for each stage by gross production reported in that year (Table S2).

In addition, to cover the potential ranges of methane leakage rates from China's conventional natural gas industry, we summarize the literature that reported stage-level methane leakage rates in China and in the U.S. [9-15], and set the lowest and highest estimates as the lower and upper bound methane leakage rates from China's conventional natural gas industry (Table S3). As Burnham et al. (2012)[12] reported methane leakage from transmission (T) and distribution (D) together, we allocate the leakage rates for transmission and distribution stages following Weber et al. (2012)[14] (T/(T+D)=60%). For studies that reported fugitive methane emissions in CO_2 equivalent intensity, we convert the normalized estimates in Weber et al. (2012) to methane leakage rates based on natural gas energy content, natural gas density and GWP values used for normalization.[14] IPCC (2006)[9] provided the lowest estimates of total methane leakage rates among our summarized studies. However, the methane leakage rates during distribution stage are larger than what Lamb et al. (2015) found recently.[8] As Lamb et al. (2015) only provided the mean and upper bound estimates for methane leakage rates during natural gas distribution, here we assume a conservative lower bound at 10% of their mean estimates based on limited information provided in Lamb et al. (2015) (i.e Fig. S3.12 and Fig. S4.5). Nevertheless, as robust policy making cares more about the upper bound than the lower bound, the lower bound estimates we provide here have more computational meanings than policy implications. For the upper bound estimates, Howarth et al. (2011)[13] and Weber et al. (2012)[14] provided similar

estimates of total methane leakage rates from the conventional natural gas industry. We use Howarth et al. (2011)'s estimates in our study that are slightly larger. The low, mean, and high methane leakage rates used in this study to estimate the corresponding lower bound, mean, and upper bound fugitive emissions for China's conventional natural gas are listed in Table 1 in the main text.

Tables S4 presents energy consumption for the conventional natural gas system in China. Neither Chinese nor U.S. studies reported the uncertainty ranges in the energy consumption for the natural gas industry to our knowledge. Earlier studies reported an uncertainty range of ±20% for China's industrial sector energy consumption based on expert judgment, which were applied to industries such as nonferrous metal and oil refining.[16] Without better information, here we assume this also applies to the conventional natural gas industry.

As gas leakage and energy consumption at each stage affects the amount of gas remaining in subsequent stages (which consequently influences the GHG emissions in those stages), GHG emissions are calculated following the flow chart shown in Fig. S1. Fig. S1 is used to illustrate the calculations for 100 m^3 of conventional natural gas produced at a well. Fugitive emissions in each stage are calculated based on mean methane leakage rates shown in Table 1 in the main text. Energy consumption related CO_2 emissions are calculated based on energy consumption and the energy mix shown in Table S4, as well as the corresponding emission factors from recent estimates.[17] Natural gas losses during each stage are estimated based on both average methane leakage rates (Table 1) and natural gas consumption rates at each stage (Table S4). Lifecycle GHG emissions are expressed as g CO_{2eq} per unit end use energy produced using either a 100 year (GWP_{100}) or 20 year (GWP_{20}) time horizon. For instance, in the power sector, g CO_{2eq}/Kwh electricity generated is reported for both GWP_{100} and GWP_{20}.

Table S1. Lifecycle processes included in this study.

Fuel Type	Upstream Processes				End Uses
Coal	Coal Mining	Coal Processing	Coal Transport		Power Generation
Conventional Natural Gas	Gas (Pre-) Production & Gathering	Gas Processing	Gas Transmission & Storage	Gas Distribution	Residential Cooking
Shale Gas	Gas (Pre-) Production & Gathering	Gas Processing	Gas Transmission & Storage	Gas Distribution	Industrial Boilers

Table S2. Summary of the mean estimates of CH_4 emissions from the conventional natural gas system based on the latest direct measurements in the U.S.

Processes	CH_4 Emission Sources	CH_4 Emissions (Gg/yr)	CH_4 Leakage
	Chemical pumps	68	0.015%
	Pneumatic controllers	580	0.127%
	Equipment leaks	291	0.064%
	Unloadings (nonplunger lift)	149	0.033%
	Workovers (without HF)	0.3	0.000%
	Unloadings (plunger lift)	108	0.024%
	Other sources	911	0.200%
	Gathering facilities [6]	1,697	0.357%
	Gathering pipelines [6]	178	0.037%
	Processing plants	506	0.106%
	Processing plants under routine maintenance	40	0.008%
Gas Transmission[8]	Transmission & storage	1,503	0.316%
Gas Distribution[7]	Distribution	393	0.086%

Table S3. Summary of stage-level methane leakage rates for conventional gas system reported in Chinese and the U.S.' studies.

	IPCC [9] (low)	IPCC [9] (high)	Burnham et al. [12] (low)	Burnham et al. [12] (high)	Weber et al. [14] (low)	Weber et al. [14] (high)	Howarth et al. [13] (low)	Howarth et al. [13] (high)	Harrison et al. [15] (low)	Harrison et al. [15] (high)	NETL [10]	Jiang et al. [11] (low)	Jiang et al. [11] (high)
Gas Production	0.05%	3.35%	0.62%	4.19%	0.52%	2.30%	0.31%	2.17%	0.24%	0.51%	1.78%	0.80%	1.90%
Gas Processing	0.02%	0.05%	0.06%	0.23%	0.17%	0.68%	0.00%	0.19%	0.11%	0.23%	0.25%	0.11%	0.76%
Gas Transmission	0.03%	0.27%	0.18%	0.64%	0.25%	0.53%	0.80%	2.20%	0.33%	0.70%	0.49%	0.00%	0.57%
Gas Distribution	0.15%	0.35%	0.12%	0.41%	--	--	0.60%	1.40%	0.22%	0.46%	--	--	--
Sum	0.25%	4.0%	0.97%	5.47%	1.0%	3.5%	1.7%	5.96%	0.9%	1.9%	2.56%	0.91%	3.23%

*Methane leakage rates for NETL[10] and Jiang et al. (2011)[11] are estimated based on the normalized fugitive carbon footprint summarized in Weber et al. (2012)[14]

Table S4. Energy efficiency and energy mix of the conventional natural gas system in China.

Processes	Energy Efficiency*	Energy Mix
Gas Production	96%	Electricity (40%), Natural Gas (23%), Residual Oil (20%), Diesel (8%), Coal (7%)
Gas Processing	94%	Electricity (10%), Natural Gas (28%), Residual Oil (40%), Diesel (1%), Coal (20%)
	Energy Intensity (KJ.ton^{-1}km^{-1})	
Gas Transmission & Storage	372	Natural Gas (99%), Electricity (1%)

*MJ out of Stage X/ MJ prior to Stage X
(Data Source: Ou et al. [18]; Ou et al.[19]; Ou and Zhang[20])

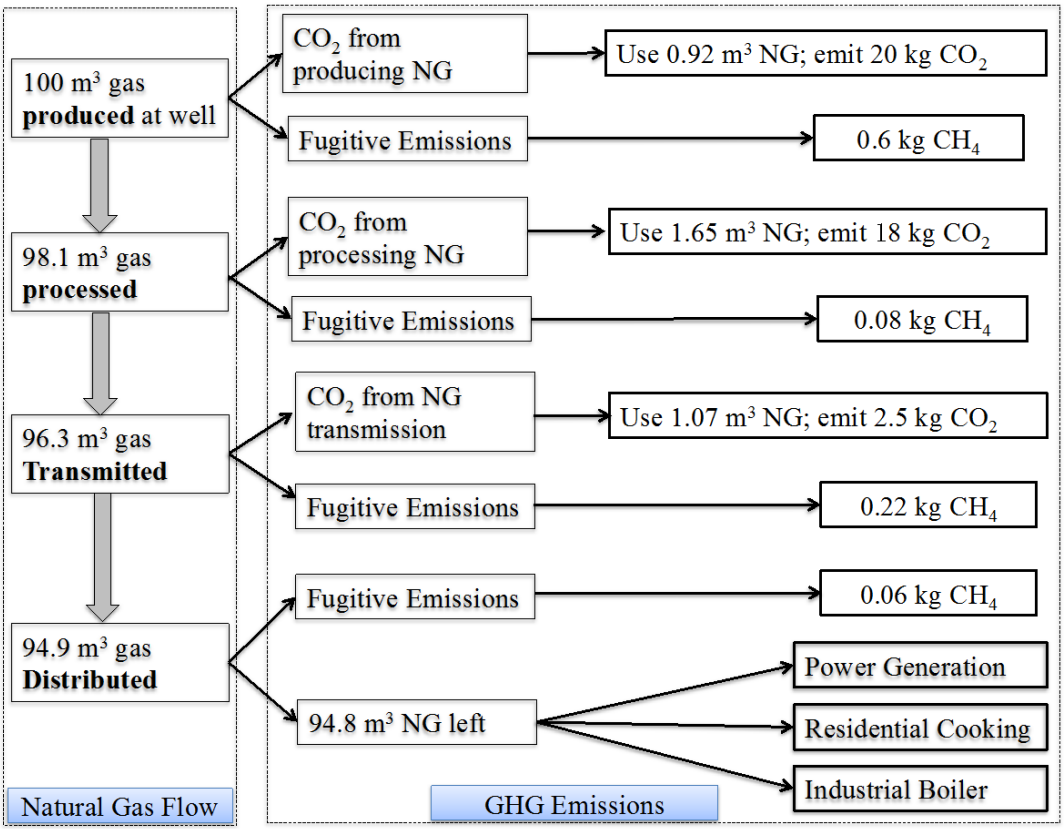


Fig. S1. Flowchart of upstream GHG emissions estimation for China's conventional natural gas (NG) system. The left side shows the natural gas flow, while the right side illustrates emissions of GHG. We illustrate the estimation process by using 100 m^3 of natural gas produced at the well under the mean methane leakage rate and upstream energy use. In each stage, there are fugitive emissions due to methane leakage and CO_2 emissions due to fossil fuel combustion. CO_2 emissions are from all fossil fuels (not only natural gas) consumed at that stage. Detailed energy consumption and energy mix in each stage are shown in Table S4. Note that CH_4 content is 85% before processing, and 95% after processing.

1.2. Estimating Additional GHG Emissions for China's Shale Gas Production

Several U.S. studies compared methane leakage between conventional natural gas and shale gas, and found that the major differences between the two lie in well completion of the pre-production stage and workover in the production stage (See Table 1 in Burnham et al. (2012)[12],

Table S1-5 in Weber and Clavin (2012)[14], Table 2 in Howarth et al. (2011)[13], Fig. 6 in Bradbury et al. (2013)[21]). NETL (2011)[10] and Burnham et al. (2012)[12] estimated lower GHG emissions for shale gas than for conventional gas as they assumed liquids unloading is conducted only for conventional gas production. However, liquids unloading was later found to be a common practice for shale gas as well.[22] Based on the U.S.' experience, we assume the major differences in fugitive emissions between conventional gas and shale gas systems in China are also from well completion and workover processes.

1.2.1 Estimating Additional Fugitive Emissions from China's Shale Gas Production

1.2.1.1 Comparison between the U.S. and Sichuan Basin Shale

China's shale development is currently concentrated in Sichuan province. In Table S5 below, we compile data on geological properties of shale in the U.S. and Sichuan basin. Reservoir pressure, which is mainly influenced by depth, is the major factor influencing shale gas production. Thus, in this study, we assume that Sichuan shale gas fields in China have similar production curves as the Haynesville Shale in the U.S., following China's Energy Research Institute National Development And Reform Commission (ERINDRC).[23]

Table S5. Comparison of geological properties between shale fields in the U.S. and China

	U.S.			**Sichuan [24]**	
	Haynesville	**Barnett**	**Marcellus**	**Qiongzhusi**	**Longmaxi**
Depth, m	3,300 – 4,000 [25]	1,200 – 2,600 [26, 27]	1,800 – 2,400 [26, 27]	2600-4600	1600-4200
Shale thickness, m	30-100 [25]	30-100 [27]	15-90 [27]	20-80	20-100

TOC, %	1-6 [25, 28]	3-6 [27, 29]	2-10 [27, 30]	2.3-4.2	1.6-3.6
Porosity, %	4-7 [31, 32]	3-6 [27, 32]	3-9 [27, 30]	1.5-2.6	1-5
RP, MPa	54-90 [31]	20-31 [29]	28-38 [33]		

TOC: Total Organic Carbon**; RP:** Reservoir Pressure

1.2.1.2 Gas Release during Well Completion

In the initial 1-2 weeks, shale gas and flowback liquids flow from underground to the surface via the wellbore.[24] At this stage, shale wells are not connected to gas pipelines, thus shale gas generated during flowback is either flared or vented. We assume a linear increase of shale gas production in Sichuan shale wells until they reach the peak production (see Fig. S2). Applying the Haynesville Shale values to Sichuan shale, the average flowback period is approximately 10 days, and the time to reach maximum production from initial gas capture is 30 days.[34] Thus, based on the calculations below, we estimate the peak production for Sichuan shale to be 96,000 m^3 /day.

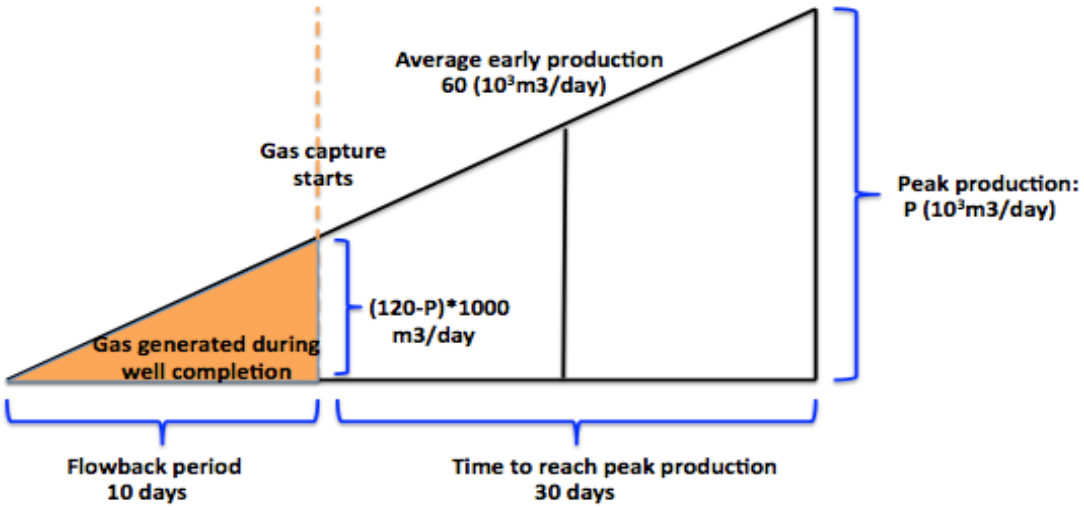


Fig. S2. Shale gas wells timeline for well completion, workover and peak production

Using theorem of similar triangles:

$$\frac{\text{Flowback period}}{\text{Flowback period+Time to reach peak production}} = \frac{2*\text{Average early production}-\text{Peak production}}{\text{Peak production}}$$

$$Peak\ production = \frac{2*\text{Average early production}*(\text{Flowback period+Time to reach peak production})}{2*\text{Flowback period+Time to reach peak production}}$$

$$\frac{10}{10+30} = \frac{2*60-P}{P} \quad \Rightarrow \quad P = \frac{2*60*40}{50}$$

$$\Rightarrow \quad P = 96{,}000\ \text{m}^3\ \text{/day}$$

Gas generated during well completion $= \frac{1}{2}$**Flowback period * (120-P)*1000*

$= \frac{1}{2}$**10 * (120-P)*1000*

$=$ *120,000* m^3

1.2.1.3 Estimation of Fugitive and Flaring Emissions during Well Completion

Gas generated during well completion is either flared at 98% combustion efficiency or vented directly to the atmosphere. Methane and CO_2 emissions from fugitive leaks and flaring emissions during well completion are calculated using equations below.[11] Average chemical composition for shale gas in China is assumed to be the same as that of conventional gas: 85% CH_4, 10% C_2H_6, 3% C_3H_8, and 2% CO_2 before processing. After processing, CH_4 content increases to 95%. In the absence of specific data, fugitive emissions from workover are generally treated the same as those from well completion.[10, 12, 35] However, this is likely to be an overestimation as the formation gas pressure (and therefore the flow rate to the well) is lower during workover than during the initial well completion. We assume that on average workover is conducted once in the lifetime of each shale well in Sichuan.

a. CO_2 Emissions:

E_{CO2} = Gas Generated during Well Completion (m^3) * Flaring Rate (%) * Methane Content (%) * Methane Density (kg/m^3) * Combustion Rate (98%) * CO_2 Emission Factor for Methane Combustion (kg CO_2 / kg CH_4) + Gas Generated during Well Completion (m^3) * Flaring Rate (%) * long chain Hydrocarbons Content (HCs) (%) * HCs Density (kg/m^3) * Combustion Rate (98%) * CO_2 Emission Factor for HCs (kg CO_2 / kg HCs) + Gas Generated during Well Completion (m^3) * Vent Rate (%)* Carbon Dioxide Content (%) * Carbon Dioxide Density (kg/m^3)

E_{CO2} = *120,000 * 0.51 * 0.85 * 0.716 * 0.98 * 2.75 + 120,000 * 0.51 * 0.1 * 1.356 * 0.98 * 88/30 + 120,000 * 0.51 * 0.03 * 2.02 * 0.98 * 3 + 120,000 * 0.49 * 0.02 *1.98*

= 137 tons

b. CH_4 Emissions:

E_{CH4} = Gas Generated during Well Completion (m^3) * Vent Rate (%) * Methane Content (%) * Methane Density (kg/m^3) + Gas Generated during Well Completion (m^3) * Flaring Rate (%) * Methane Content (%) * Methane Density (kg/m^3) * Incomplete Combustion Rate (2%)

E_{CH4} = *120,000 * 0.49 * 0.85 * 0.716 + 120,000 * 0.51 * 0.85 * 0.716 *0.02*

= 36.5 tons

1.2.1.4. Estimated Ultimate Recovery (EUR) for Sichuan Shale Wells

The first shale gas well in China was drilled in January 2011 in Sichuan. As the average lifetime of shale gas wells is likely to be more than 15 years, there is no information yet available regarding estimated ultimate recovery (EUR). Although most Haynesville wells were also drilled around 2010-2011, detailed production data from thousands of Haynesville wells exist that can

be used to estimate ultimate recovery. A national survey shows there were 322 shale wells in China in April 2014[36], and detailed production data is not available.

Table S6 shows the estimated cumulative production for an average Haynesville shale gas well based on the survey data of 1,253 Haynesville shale wells between 2008 to 2014.[34] As data is only available for 5 years, cumulative production for longer periods is obtained via model fitting of early stage observation data. Edwards et al. (2015)[37] described the model and data fitting procedure in detail. Assuming the average shale well's lifetime is 20 years, we estimate EUR for an average Haynesville shale gas well to be 4.33 bcf (0.12 bcm), with the peak daily production of 9536 Mcf/day (270,000 m^3/day). We estimate the Sichuan Shale's EUR by scaling that of Haynesville Shale based on ERINDRC's assumption that Sichuan shale has a similar production curve as the Haynesville Shale.[23] As in section 1.3.2, we estimate Sichuan Shale's peak daily production to be 96,000 m^3/day, and the EUR for average Sichuan Shale wells with a 20 year lifetime to be 0.04 billion m^3. Parameters for estimating the lower and upper bound additional fugitive emissions during well completion and workover are listed in Table S13.

$$\frac{\text{EUR}_{\text{Sichuan}}}{\text{EUR}_{\text{Haynesville}}} = \frac{\text{Peak daily production}_{\text{Sichuan}}}{\text{Peak daily production}_{\text{Haynesville}}}$$

$$\frac{\text{EUR}_{\text{Sichuan}}}{0.12} = = \frac{96{,}000}{270{,}000}$$

$\text{EUR}_{\text{Sichuan}} = 0.04\ (\text{bm}^3)$

Table S6. Modeled cumulative production for an average Haynesville Shale well

Time (years)	5	10	15	20	25	30	50
Cumulative Production (billion cubic feet)	3.47	4.00	4.22	4.33	4.39	4.43	4.48

1.2.2 Estimating Additional Energy-Related CO_2 Emissions from China's Shale Gas Production

As mentioned in the main text, Chang et al. (2014)[38] reported detailed shale-to-well energy consumption data for China's first shale well, which we use to determine the additional energy consumption for China's shale gas production compared to that of conventional natural gas. Table S7 lists our estimated additional CO_2 emissions for shale production. According to Chang et al. (2014)[38], energy use of the shale-to-well system is dominated by diesel. We estimate energy-related CO_2 emissions for hydraulic fracturing to be 456 tons/well. For a well conducting both vertical drilling and horizontal drilling, we assume that energy use for well drilling is proportional to the drilling distances. The average vertical depth and horizontal length of current shale gas wells in Sichuan are around 3000-3500 meters and 1000-1500 meters, respectively.[23] Thus, we estimate total CO_2 emissions for horizontal drilling is 111 tons/well. Additionally, total diesel consumption related to onsite water use and flowback processing (wastewater transport, treatment & disposal) is 64640 kg/well. As 95% of onsite water use is related to HF[38], additional water and wastewater related CO_2 emissions for shale gas compared with conventional gas is 192 tons/well. We make a conservative assumption that the uncertainty range for shale gas industry in total energy consumption is the same as that for the conventional natural gas industry.

Table S7. CO_2 emissions from additional energy consumption in shale gas production compared with conventional gas production in China.

Mechanical Operation	Detailed Break-down	Diesel Consumption (kg/well)	CO_2 Emissions (tons/well)
	Hydraulic fleet energy use (i.e., Pumper, Blender, Monitoring Van)	143000	
	Fracturing fluid sand transportation	2730	

	Diesel transport	64	
Horizontal Drilling	Horizontal Drilling	35640	111
	Fracturing related water transportation	58180	
	Fracturing & flowback processing	3230	

(Data source: Chang et al., 2014[38])

1.3. Estimating Upstream GHG Emissions for Coal in China

Table S8 and Table S9 present emission factors we use for coal mining in China, as well as the energy efficiency and energy mix for coal production, processing, and transport. Similar to conventional natural gas, coal industry is a long-standing business in China. Thus, we apply the same uncertainty range of ±20% reported in Zhao et al. (2011)[16] for China's typical industries' energy consumption to the coal industry.

Table S8. Fugitive emission factors for coal mining in China.

Coal Mine Types	**Fugitive Emission Factors (m^3 CH_4/ ton coal)**	
Underground mining	Coal mining	Post mining*
High-methane mine	21.83	3.02
Low-methane mine	4.53	1.13
Surface mining	2.5	0.1

* Post mining fugitive emissions refer to methane leakage after coal mining, during the subsequent handling, processing and transport processes.[9] Data Source: Zhang and Chen[2]

Table S9. Energy efficiency and energy mix for coal systems in China.

Processes	**Energy Efficiency**	**Energy Mix**

Coal Mining	97%		Coal (80%), Electricity (16%), Natural Gas (1%), Diesel (2%), Gasoline (1%)
Coal Processing	97%		
	Transport Mode Share	**Energy Intensity ($KJ.ton^{-1}km^{-1}$)**	
	Railway (75%)	240	Diesel (55%), Electricity (45%)
	Waterway (17%)	148	Residual oil (100%)
	Long-distance Road (8%)	1200	Diesel (68%), Gasoline (32%)
	Short-distance Road (100%)	1362	Diesel (68%), Gasoline (32%)

(Data source: Ou et al.[19]; Ou and Zhang[20])

1.4. Estimating GHG Emissions from End Use Combustion in China

Table S10 shows the emission factors for coal and gas burning in China's power, residential and industrial sectors. EFs used in this study are obtained from Zhao et al. (2012) and IPCC (2006).[39, 40] Table S11 shows the corresponding end use efficiencies for coal and gas combustion in China.

Table 10. Emission factors for coal and gas in various end use sectors in China.

Sector	Fuel Type	EFs (g CO_2/kg fuel): Mean (Low-High)
	Bituminous Coal [a]	2058 (1840-2342)
	Lignite [a]	1358 (1268-1447)
	Anthracite [a]	2320 (2176-2467)
	Gas [b]	2750 (2662-2858)
	Coal [a]	2170 (1276-2670)
	Gas [a]	3144 (2791-3493)
	Bituminous Coal [a]	1923 (1215-2631)
	Lignite [a]	1260 (1111-1408)
	Anthracite [a]	2320 (2176-2467)
	Gas [a]	3116 (2785-3448)

(Data source: [a] Zhao et al., 2012[39]; [b] IPCC, 2006[40])

Table S11. Energy efficiencies for coal and gas technologies in the power, residential, and industrial sectors in China.

Sector	Fleet Type	Efficiency
	High Pressure PC	30%[a]
	Very High Pressure PC	33% [a]
	Subcritical PC	37% [a]
	Supercritical PC	40%[a]
	Ultra-Supercritical PC	42%[a]
	IGCC*	45%[b]
	NGCC*	50%[c]
	Coal Cooking Stove	35%[d]
	Gas Cooking Stove	57% [c,e]
	Industrial Coal Boiler	69% [c,e]
	Industrial Gas Boiler	83% [c,e]

*NGCC: natural gas combined cycles; IGCC: integrated gasification combined cycle

(Data source: [a]Shen et al. [41]; [b]Kunze and Spliethoff (2012)[42]; [c]Ding et al. [43]; [d]Mao et al. [44]; [e]Wang et al.[45])

1.5. Sensitivity Analysis of Key Parameters in Estimating Fugitive Emissions During Well Completion and Workover for Shale Wells

Table S12 illustrates ranges of input values of the major parameters we use in section 1.2 to estimate fugitive emissions during well completion, as well as EUR for an average Sichuan Shale. Lacking of information to accurately determine the distribution of input parameters, we conduct a sensitivity study to identify the contribution of each key factor in affecting the fugitive emissions in turn by fixing all other input parameters at the mean level. To date, there is no requirement to use methane mitigation technologies for China's natural gas or shale gas production. In comparison, green completions are required in the U.S.'s shale gas industry, and

are estimated to reduce fugitive emissions per well completion by 90%.[21] Here we use a sensitivity test to analyze the effect of green completions on total emissions during well completion and workover. Similarly, we assume two extreme cases regarding the flaring ratio: all gas is either flared or vented. Flowback normally takes 1-2 weeks.[46] Workover frequency is hard to determine, as it can vary from well to well. Most U.S. studies assume it occurs between zero and twice during an average shale well's lifetime[10-12, 14, 47], so we apply the same uncertainty range for Sichuan shale. We assume average shale well has a lifetime between 15 to 50 years. Regarding the EUR for Haynesville Shale, we obtain the upper and lower bound by calculating the observed standard deviations (SD) of over 1000 Haynesville shale gas wells. Each shale gas well reports cumulative production at the 1st, 2nd and 5th year. Without better information, we assume that the SD for cumulative production at the 20th year is the same as that at the end of the 5th year (refer to Fig. 1 in the main text to see the temporal variation of the SD of cumulative production for shale wells). The major parameters for estimating the upper and lower bound well completion and workover GHG emission intensity are shown in Table S13.

Table S12. Ranges of key parameters for sensitivity study of methane leakage from shale well completion and workover. All parameters are for Sichuan Shale unless otherwise indicated.

	Mean	Low	High
Control technology	No	No	Green completion
Flaring ratio	51%	0%	100%
Flowback time	10 days	7 days	14 days
Workover frequency	1	0	2
Lifetime	20 years	15 years	50 years
Gas composition	85% CH_4 +10% C_2H_6 + 3% C_3H_8 + 2% CO_2	70% CH_4 + 30% inert components (i.e. N_2, H_2S)	90% CH_4 +10% C_3H_8
EUR-Haynesville	4.3 billion cubic feet	1.3 billion cubic feet	7.4 billion cubic feet

Table S13. Key input factors in estimating the lower bound, mean, and upper bound lifecycle carbon intensities for fossil fuels evaluated in this study.

Shale Gas				
		Mean	**Lower bound**	**Upper bound**
Methane leakage rates for the conventional gas system [a]		1.37%	0.11%	5.96%
	Control technology	No	Yes	No
	Flaring ratio	51%	100%	0%
	Flowback time	10 days	7 days	14 days
	Workoever frequency	1	0	2
	Lifetime	20 years	50 years	15 years
	EUR-Haynesville	4.3 billion cubic feet	7.4 billion cubic feet	1.3 billion cubic feet
Upstream energy consumption [c]		Energy use in Tables S4 and S7	-20% mean energy use	+20% mean energy use
Shale gas exploration success rates [d]		70%	45%	100%
End use combustion emission factors [e]	Refer to Table S10			

Conventional Natural Gas			
	Mean	**Lower bound**	**Upper bound**
Methane leakage rates for the conventional gas system [a]	1.37%	0.11%	5.96%
Upstream energy consumption [f]	Energy use summarized in Table S4	-20% mean energy use	+20% mean energy use

End use combustion emission factors [e]	Same as that for shale gas		
Coal			
	Mean	**Lower bound**	**Upper bound**
Methane recovery rates during coal mining [g]	7%	90%	0%
Upstream energy consumption [h]	Energy use in Table S9	-20% mean energy use	+20% mean energy use
End use combustion emission factors [e]	Refer to Table S10		

[a] Detailed breakdowns shown in Table 1 in the main text; [b] Refer to Table S12; [c] Refer to Tables S4 and S7 for mean energy consumption, and Section S1.1 and S1.2.2 for discussions on the uncertainty range; [d] Chang et al. (2014)[48]; [e] Zhao et al. (2012)[39]; [f] Refer to Tables S4 for mean energy consumption, and Section S1.1 for discussions on the uncertainty range; [g] Zhang et al. (2014)[3], Burnham et al. (2012)[12]; [h] Refer to Table S9 for mean energy consumption, and Section S1.1 and S1.3 for discussions on the uncertainty range.

1.6. Breakeven Methane Leakage Rates Estimation

Here we estimate the breakeven methane leakage rates to ensure net carbon reductions by switching from coal to shale gas. When estimating carbon intensity for coal, we apply the mean values for all factors other than CH_4 recovery rate during coal mining, which is likely to experience considerable changes along with China's coalbed methane development policies. Similarly, for shale gas system, we focus on lifecycle methane leakage, the factor we identify contributing the largest uncertainty in shale gas lifecycle carbon intensity, while using the mean values for all other factors (i.e., energy consumption for the shale gas system). Thus, the breakeven methane leakage rates calculated are primarily based on an average coal-to-shale gas switch. For simplification, we do not differentiate methane leakage from different stages, and assume an average CH_4 content of 90%. Also, flaring emissions during flowback, which depends on the methane leakage during well completion, are not considered due to a lack of exact information about methane leakage in each process. Below we use 1 MJ coal and shale gas production in the power sector to illustrate the calculation process. Additionally, we use the technology warming potential (TWP) metric developed by Alvarez et al. (2011)[4] to calculate the breakeven methane leakage rates using the excel model provided in their study and obtained similar results as using GWPs.

a. 1 MJ coal production:

Upstream CH_4 Emissions (g) =

Coal production (MJ) * CH_4 EFs during coal mining (g/MJ) * [1- CH_4 recovery rates (R)] + Coal production (MJ) * CH_4 EFs post coal mining (g/MJ) = 0.42 (1-R)+0.067 (g)

Upstream CO_2 Emissions (g) =

Energy consumption for coal production (MJ) * ΣFuel type $_i$ (%) * EFs $_i$ (g/MJ) + Energy consumption for coal processing (MJ) * ΣFuel type $_i$ (%) * EFs $_i$ (g/MJ) + Energy consumption for coal transmission (MJ) * ΣFuel type $_i$ (%)* EFs $_i$ (g/MJ) = 6.83 (g)

End-Use CO_2 Emissions (g) =

Coal delivered to end users (MJ) * EFs (coal) $_{sector}$ (g/MJ) = 92.8 (g)

Lifecycle CO_{2eq} Emissions (g) =

[0.42 (1-R)+0.067] * GWP + 6.83 + 92.8 (g)

Electricity Generated (kWh) =

Coal delivered to end users (MJ) * Average PC fleet efficiency * (1- electricity transmission loss rate) /3.6 = 0.1 (kWh)

Lifecycle carbon intensity for Coal (g CO_{2eq} /kWh) =

Lifecycle CO_{2eq} Emissions (g) / *Electricity Generated* (kWh) =

10*[(0.42 (1-R)+0.067])* GWP+99.63] (g CO_{2eq} /kWh)

b. 1 MJ shale gas production:

Upstream CH_4 Emissions (g) =

Shale gas production (MJ) * Lifecycle methane leakage rates (r) / CH_4 energy content (MJ/m^3) * CH_4 density (g/m^3) = 20.1*r (g)

Upstream CO_2 Emissions (g) =

Energy consumption for shale gas production (MJ) * ΣFuel type $_i$ (%) * EFs $_i$ (g/MJ) + Additional diesel consumption (kg) * EFs $_{diesel}$ (g/kg) + Energy consumption for shale gas processing (MJ) * ΣFuel type $_i$ (%) * EFs $_i$ (g/MJ) + Energy consumption for shale gas transmission (MJ) * ΣFuel type $_i$ (%)* EFs $_i$ (g/MJ) = 14.45 (g)

End-Use CO_2 Emissions (g) =

Shale gas delivered to end users (MJ) * EFs (gas) $_{sector}$ (g/MJ) =

Shale gas production (MJ) * [1- Lifecycle methane leakage rates (r)/Shale gas CH_4 content] * EFs (gas)

$_{sector}$ (g/MJ) = 56.1 * (1-r/0.9) (g)

Lifecycle CO_{2eq} Emissions (g) =

20.1 r * GWP + 8.82 + 56.1 * (1-r/0.9) (g)

Electricity Generated (kWh) =

Shale gas production (MJ) * [1- Lifecycle methane leakage rates (r)/ Shale gas CH_4 content] * NGCC efficiency * (1- electricity transmission loss rate) /3.6 = 0.13 * (1-r/0.9) (kWh)

Lifecycle carbon intensity for shale gas (g CO_{2eq} /kWh) =

Lifecycle CO_{2eq} Emissions (g) / *Electricity Generated* (kWh) =

[20.1 r * GWP + 14.45+ 56.1 * (1-r/0.9)] / [0.13 * (1-r/0.9)] (g CO_{2eq} /kWh)

To ensure net carbon reductions by switching from coal to shale gas in the power sector:

Lifecycle carbon intensity for shale gas < Lifecycle carbon intensity for Coal

[20.1 r * GWP + 14.45+ 56.1 * (1-r/0.9)] / [0.13 * (1-r/0.9)] < 10*[(0.42 (1-R)+0.067])* GWP+99.63].

Thus: r < 6.0%

Likewise, the breakeven r is approximately 4.2% and 7.7% in the industrial and residential sector, respectively.

2. Detailed Results

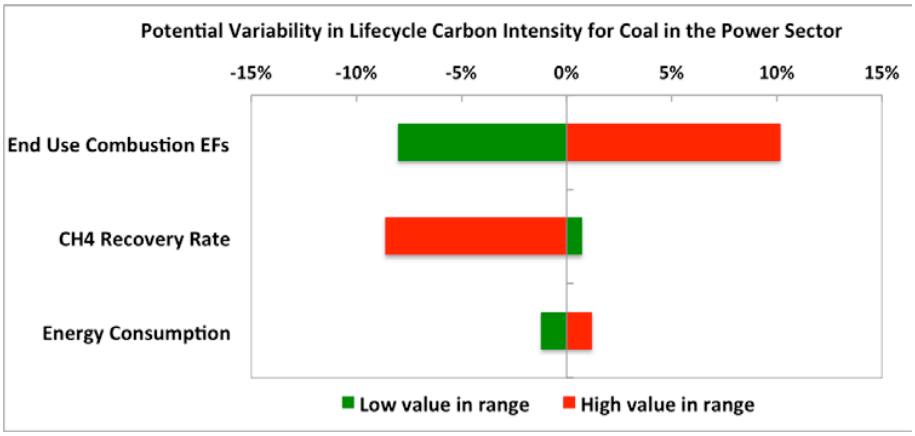
Table S14. Upper bounds of lifecycle GHG emission intensities for shale gas under each industrial practice. [a]

	Green Completions	Complete Flaring of Flowback Gas	Lowest Flowback Time	No Workover	High EUR	All Five Practices	Shale Gas[b]	Coal[c]
GWP_{100}	268	273	281	285	262	234	335	355
GWP_{20}	431	429	470	479	432	383	626	432
GWP_{100}	241	245	252	254	236	213	296	387
GWP_{20}	374	372	406	414	375	335	534	461
GWP_{100}	164	167	172	174	161	145	202	189
GWP_{20}	256	255	278	282	256	229	366	226

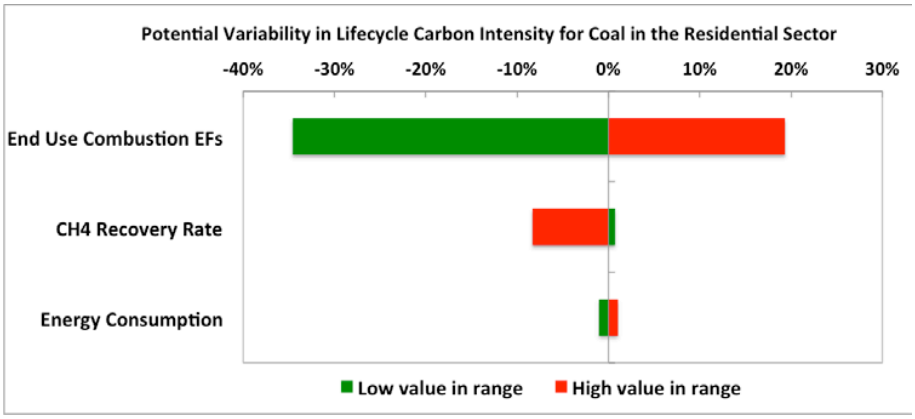
[a] The upper bound estimate of GHG emission intensity under each industrial practice is estimated by assuming that all other factors, rather than the industrial practice evaluated, take the value in Table S13 that causes the largest lifecycle GHG emission intensity for shale gas wells. For the industrial practice evaluated, it takes the value in Table S13 that causes the lowest lifecycle GHG emission intensity.

[b] The upper bound estimate of GHG emission intensity for shale gas in China.

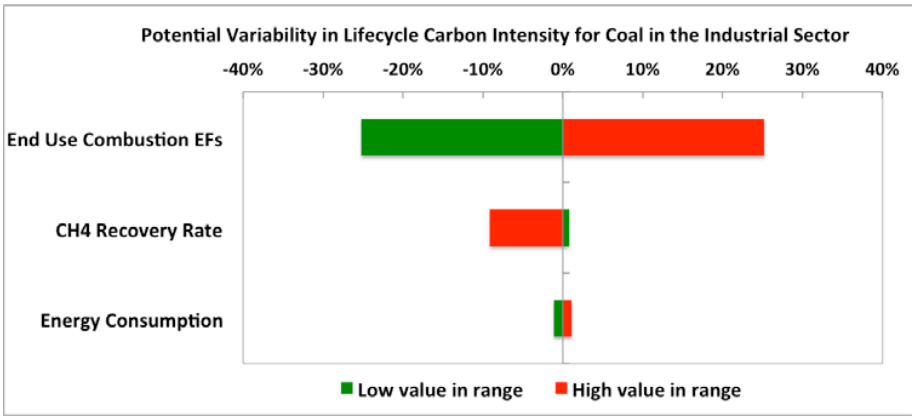
[c] The upper bound estimate of GHG emission intensity for coal in China.


Potential Variability in Lifecycle Carbon Intensity for Coal in the Power Sector
-15%
-10%
-5%
0%
5%
10%
15%
End Use Combustion EFs
CH4 Recovery Rate
Energy Consumption
Low value in range
High value in range

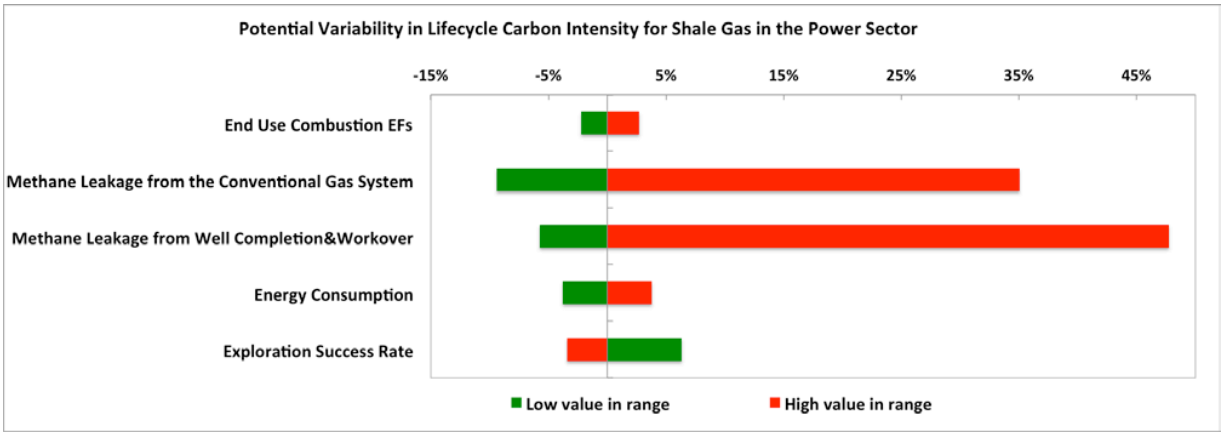
(a)


Potential Variability in Lifecycle Carbon Intensity for Coal in the Residential Sector
-40%
-30%
-20%
-10%
0%
10%
20%
30%
End Use Combustion EFs
CH4 Recovery Rate
Energy Consumption
Low value in range
High value in range

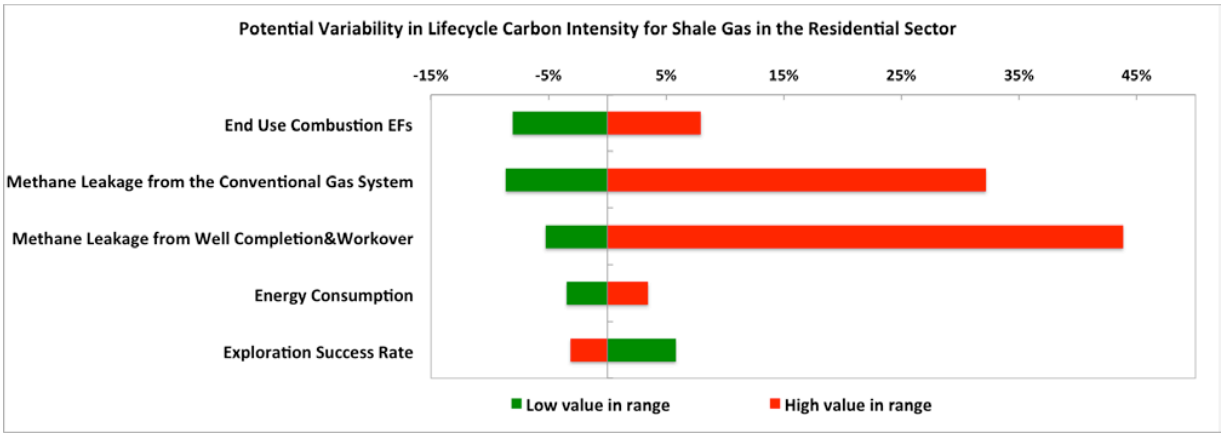
(b)


Potential Variability in Lifecycle Carbon Intensity for Coal in the Industrial Sector
-40%
-30%
-20%
-10%
0%
10%
20%
30%
40%
End Use Combustion EFs
CH4 Recovery Rate
Energy Consumption
Low value in range
High value in range

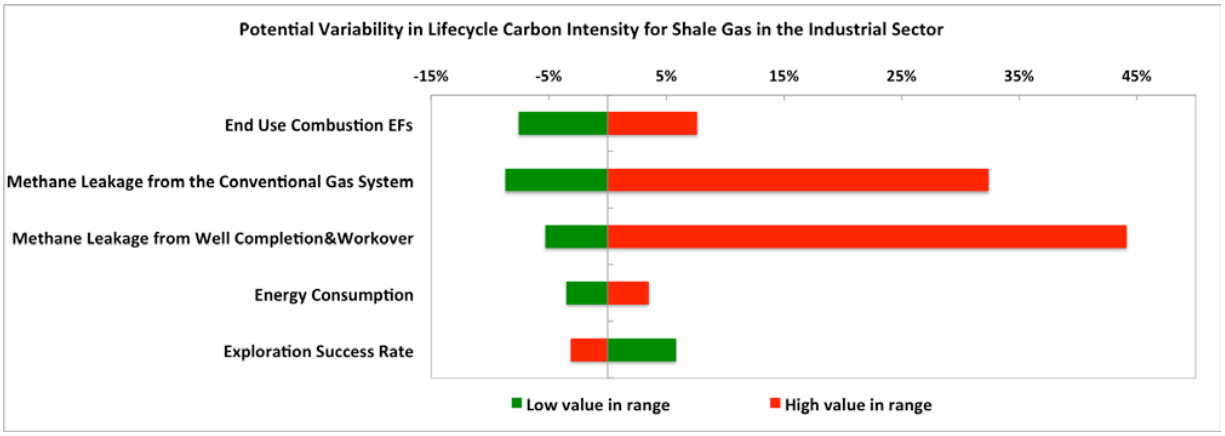
(c)


Potential Variability in Lifecycle Carbon Intensity for Shale Gas in the Power Sector
-15%
-5%
5%
15%
25%
35%
45%
End Use Combustion EFs
Methane Leakage from the Conventional Gas System
Methane Leakage from Well Completion&Workover
Energy Consumption
Exploration Success Rate
Low value in range
High value in range

(d)

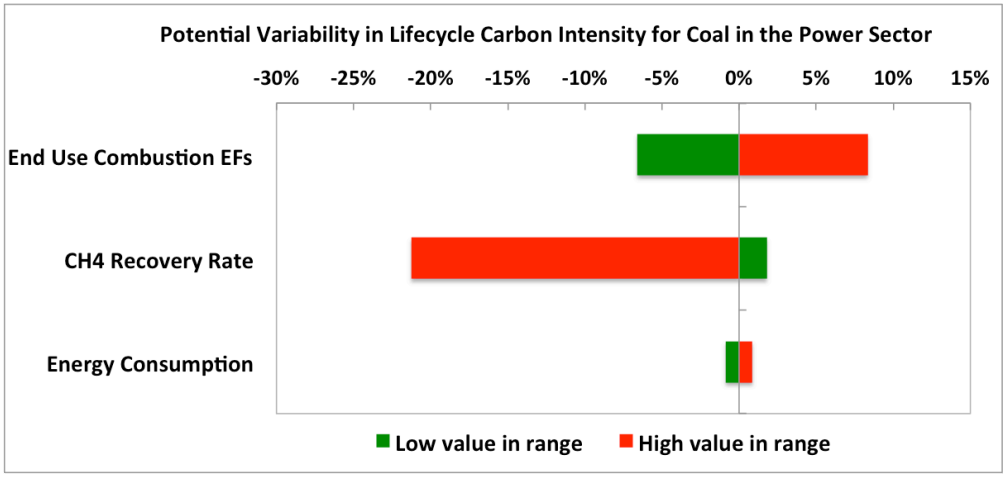

Potential Variability in Lifecycle Carbon Intensity for Shale Gas in the Residential Sector
-15%
-5%
5%
15%
25%
35%
45%
End Use Combustion EFs
Methane Leakage from the Conventional Gas System
Methane Leakage from Well Completion&Workover
Energy Consumption
Exploration Success Rate
Low value in range
High value in range

(e)

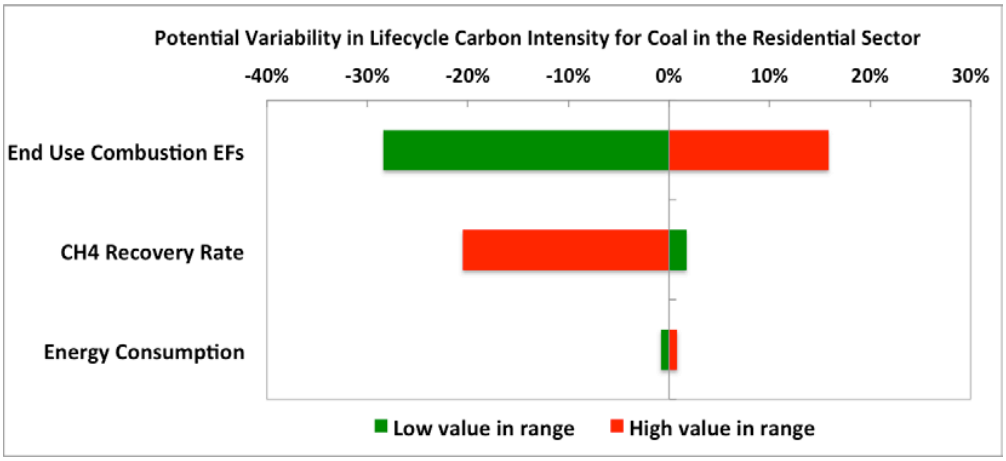


(f)

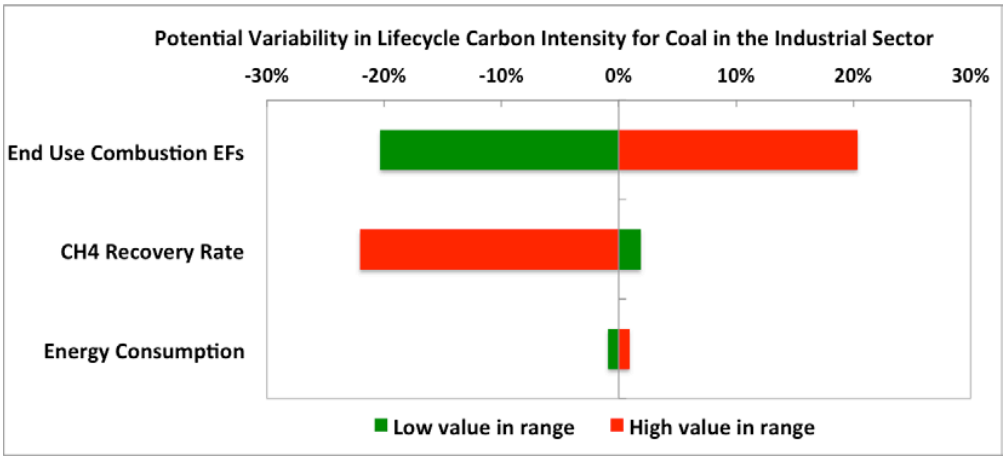
Fig. S3. Sensitivity analysis for lifecycle GHG emission intensity for coal (a, b, c) and shale gas (d, e, f) burned in the residential and industrial sectors over a 100-year time horizon (0% indicates the mean estimate).



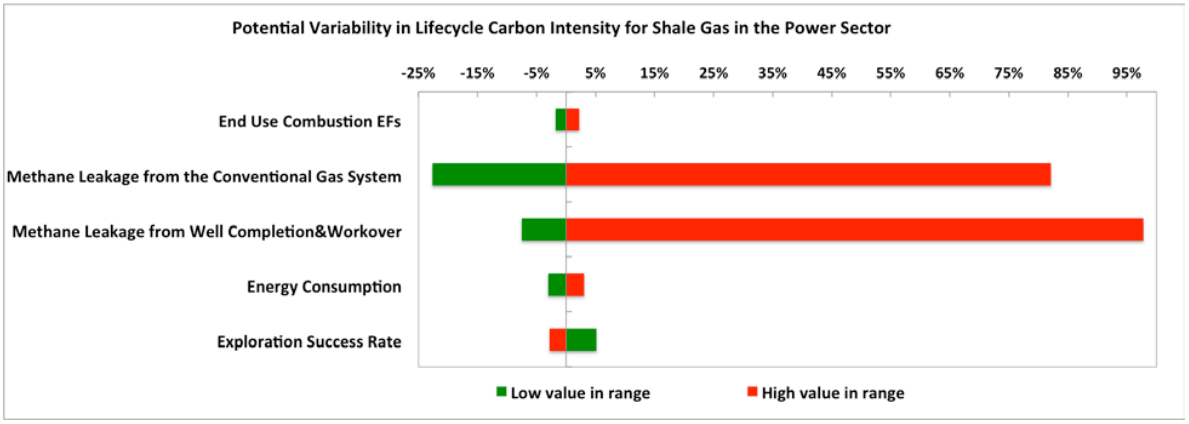
(a)


Potential Variability in Lifecycle Carbon Intensity for Coal in the Residential Sector
-40%
-30%
-20%
-10%
0%
10%
20%
30%
End Use Combustion EFs
CH4 Recovery Rate
Energy Consumption
Low value in range
High value in range

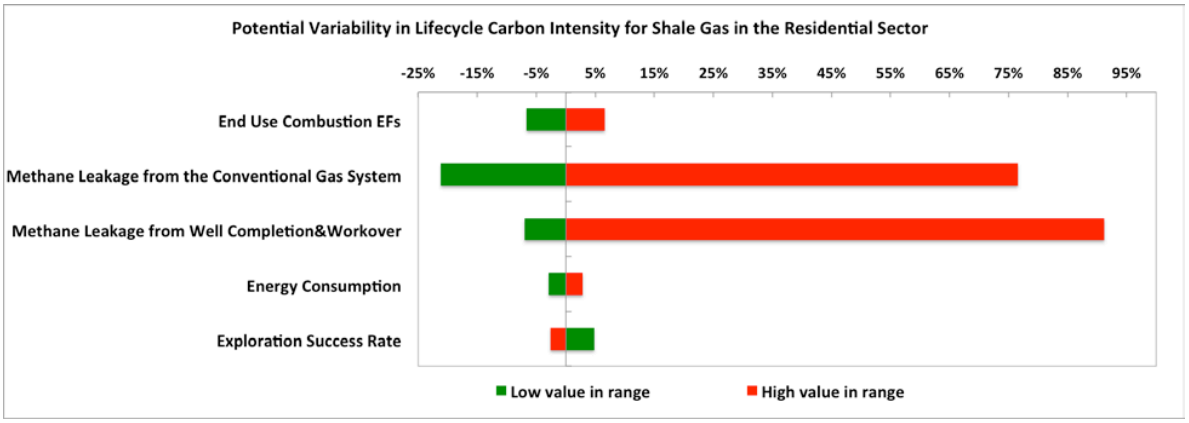
(b)


Potential Variability in Lifecycle Carbon Intensity for Coal in the Industrial Sector
-30%
-20%
-10%
0%
10%
20%
30%
End Use Combustion EFs
CH4 Recovery Rate
Energy Consumption
Low value in range
High value in range

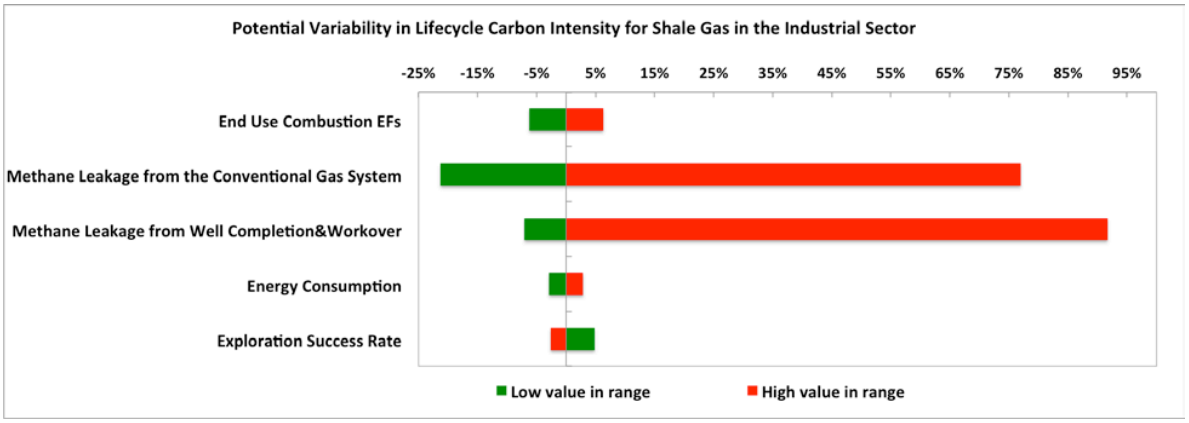
(c)


Potential Variability in Lifecycle Carbon Intensity for Shale Gas in the Power Sector
-25%
-15%
-5%
5%
15%
25%
35%
45%
55%
65%
75%
85%
95%
End Use Combustion EFs
Methane Leakage from the Conventional Gas System
Methane Leakage from Well Completion&Workover
Energy Consumption
Exploration Success Rate
Low value in range
High value in range

(d)



(e)



(f)

Fig. S4. Sensitivity analysis for lifecycle GHG emission intensity for coal (a, b, c) and shale gas (d, e, f) burned in the power, residential, and industrial sectors over a 20-year time horizon (0% indicates the mean estimate).

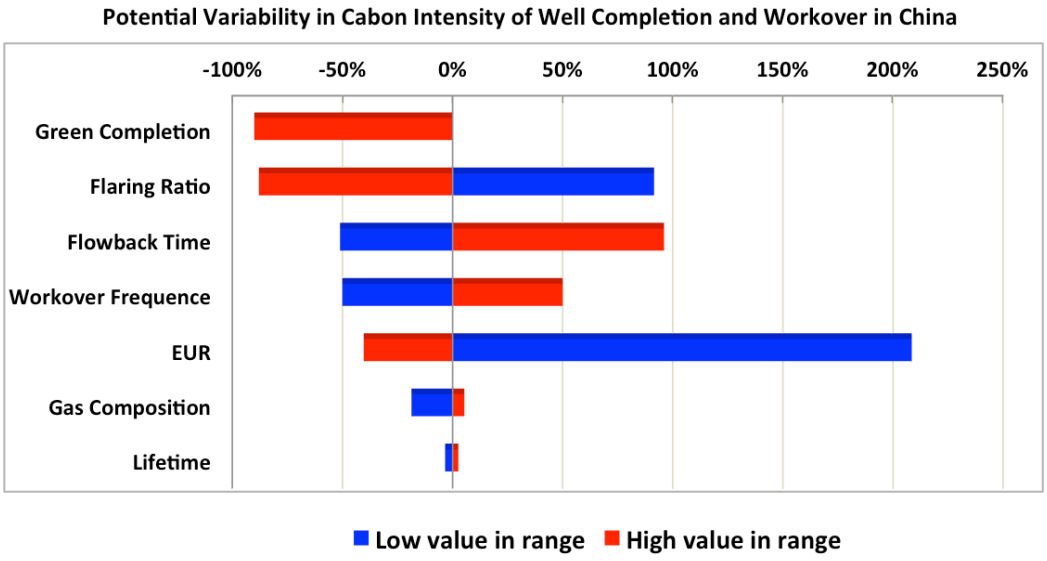


Fig. S5. Sensitivity analysis for GHG emission intensity during well completion and workover over a 20-year time horizon (0% indicates the average estimate).

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Appendix for Chapter 3.

Air Quality, Human Health and Climate Implications of China's Synthetic Natural Gas Development

1. Materials and Methods

We describe the details of the integrated assessment tools used in this study to estimate the environmental and human health impacts of China's coal-based synthetic natural gas (SNG) projects.

1.1 Constructing SNG Allocation Scenarios

In this study, we follow the 2010-2050 energy projections of the International Energy Agency's Energy Technology Perspectives (ETP)[1]. The Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) model (http://gains.iiasa.ac.at/models/), developed by the International Institute for Applied Systems Analysis (IIASA) (http://www.iiasa.ac.at), implements the ETP energy scenario as the ECLIPSE_V5a_CLE scenario (2010-2050)[2-4] to reflect and predict air pollution policies and regulations based on China's 12th Five-year plan (FYP). Policies and regulations are represented as penetration rates of a variety of control technologies with various removal efficiencies. This study uses the ECLIPSE_V5a_CLE scenario for 2020 as its base case. We then estimate the impacts of a switch to SNG from coal for various end use applications in China on air pollutant emissions, associated premature mortality, and CO_2 emissions. The focus of our study is the year 2020 for which development plans are relatively clear as multiple large-scale SNG projects have been approved[5]. Beyond that year, the further development of SNG is

more uncertain due to the negative environmental implications on global climate, local water stress, technology lock-in and so forth[5, 6].

As we mentioned in the main text, through 2013, the Chinese government had approved a total capacity of 37.1 billion cubic meters (bcm) of SNG, with another ~200 bcm proposed by the industry[5]. Government plans for annual SNG production is about 15-18 bcm by 2015 and 32 bcm by 2017, with a potential production of approximately 57 bcm by 2020[7-10]. Although the government has continuously changed SNG production plans, in this study we use the potential production of 57 bcm as a sensitivity analysis to identify a superior SNG allocation strategy that brings large air quality and health benefits, while small climate penalties. Discussions of uncertainties in actual SNG production are included in the main text. Of total SNG projected to be produced nationally in 2020, we assume Xinjiang, Inner Mongolia, Liaoning and Shanxi each has an annual production of about 29 bcm, 20 bcm, 4 bcm, and 4 bcm, respectively, based on SNG development plans[5, 11]. According to official plans, Inner Mongolia's SNG is designed mainly to serve gas needs in the Beijing-Tianjin-Hebei (BTH) region, Inner Mongolia and Jilin province[12, 13]. Similarly, Xinjiang's SNG targets Xinjiang, Gansu, Ningxia, Shaanxi, Shandong, Henan, Hubei, Hunan, Jiangxi, Fujian, Zhejiang, Guangdong and Guangxi provinces[7, 14]. SNG produced in Liaoning and Shanxi provinces is expected to be primarily consumed locally, as these two provinces traditionally have imported gas[15].

About 85% of natural gas in China is consumed in the power, industrial, and residential sectors[15]. Thus, this study focuses on the environmental impacts of natural gas substitution for coal in the three dominant sectors. We build three main SNG sectoral allocation scenarios. Each allocation scenario has 3 sub-scenarios that use various criteria for coal substitution (Table S1).

These allocation scenarios examine the impacts of SNG substitution for coal in the power, industrial, and residential sectors in turn.

1.2 Estimating Air Pollutant and Carbon Dioxide Emission Changes from SNG Substitution for Coal

The primary difference in air pollutant emissions between SNG and coal lie in end use combustion as gas made from coal can be burned much more cleanly and efficiently than the coal used to make it[16, 17] and relatively few emissions are associated with the centralized gasification process[18]. In contrast, SNG substitution for coal leads to a net increase in CO_2 emissions over direct coal use even though the end use efficiencies are higher with gas[11].

CO_2 emissions due to SNG production from coal are calculated by multiplying the SNG CO_2 emission factors (4.25 kg CO_2 emitted/m^3 SNG produced)[19] by total SNG produced. The GAINS model provides hundreds of detailed breakdowns of provincial energy uses and mixes, unabated emission factors for various air pollutants and fuel types, combustion technologies, and control technologies with varying removal efficiencies, though with no spatial information within each province. For each combustion situation (unabated EFs for each fuel type, consumption province, combustion technology and control technology), it provides a specific EF for each air pollutant. Fig. S1 shows the unabated and abated emission factors (EFs) for major air pollutants due to coal combustion in the power, industrial, and residential sectors to illustrate the differences across the three sectors. Power and industrial sectors generally have higher unabated air pollutants EFs than those in the residential sector, suggesting a higher quality of coal allocated to the residential sector. This is also identified in earlier emission inventory studies[20, 21]. However, primarily due to increasing penetration of more efficient end-of-pipe control technologies based on China's 12^{th} five-year plan, particularly in the power and industrial sectors, the abated EFs in these two

sectors are significantly reduced in year 2020. On the contrary, the abated residential EFs are marginally lower than the corresponding unabated EFs, as few clean-coal technologies can be applied for small and scattered residential coal users (i.e. improved coal stoves)[17]. Thus, a large proportion of coal burned in the power and industrial sectors have low abated air pollutants emission factors, resulting in lower air pollutant emission reductions when SNG proportionally substitutes coal in these two sectors than in the residential sector. Table S2 shows CO_2 emission factors and thermal efficiencies for coal and natural gas combustion in the three sectors used in the ECLIPSE scenario. The ECLIPSE scenario uses the same CO_2 emission factor for the same type of fuel combustion, consistent with the fact that energy-related CO_2 EFs are primarily determined by fuel types[22]. In addition, energy efficiencies for coal and gas combustion listed in Table S2 are generally comparable to China's domestic studies[11, 23]. The ECLIPSE scenario assumes the same energy efficiency for industrial coal boilers. This is supported by State Administration for Quality Supervision and Inspection and Quarantine (AQSIQ)'s finding that over 95% of China's industrial boilers have an operation capacity of lower than 20 t/h, with an average thermal efficiency of 69% (55%-75%) and 83% (80%-85%) for coal and gas fired industry boilers, respectively[11, 19]. Thus, the relative energy efficiency improvement from a coal-to-gas switch for industrial boilers identified in the ECLIPSE scenario is marginally lower than that estimated using the average efficiency values from the AQSIQ survey. Also, larger thermal efficiency improvement of a coal-to-gas switch in the residential sector than in the industrial and power sectors was also identified in earlier studies[17]. Thus, at the national average level, the relative thermal efficiency improvement due to switching from coal to gas identified in Table S2 is consistent with other studies. However, more specific information (i.e. operating conditions, equipment maintenance) is needed to conduct case-by-case comparisons, which is beyond the

scope of this study. Please refer to Klimont et al. (2016)[24], Stohl A, et al. (2015)[3], and the publicly available GAINS model (http://gains.iiasa.ac.at/models/) for more details.

Following SNG allocation sub-scenarios in Table S1, we use SNG to substitute coal used in each of the key demand sectors across provinces targeted to receive SNG using the GAINS model. For instance, SNG_Power medium scenario means SNG proportionally substitutes coal in the power sector across provinces targeted to receive SNG. Integrated gasification combined cycle (IGCC), the cleanest current available coal-fired power plants, are not considered for substitution with SNG as they have comparable air pollutants emission factors with natural gas combined cycle (NGCC). Air pollutants and CO_2 emission reductions from end use combustion resulting from SNG substitution for coal under each sub-scenario are calculated using equation 1.

$$\Delta E_{a,p,s} = \Delta SNG_{p,s} * \left(\frac{HV_{SNG}}{HV_{Coal}} * \frac{\eta_{SNG,s}}{\eta_{Coal,s}} * \sum EF_{Coal,a,p,s,t} * P_{Coal,a,p,s,t} - \sum EF_{SNG,a,p,s,t} * P_{SNG,a,p,s,t}\right) \quad (1)$$

$\Delta E_{a,p,s}$: emission reductions for pollutant type a, in province p and subsector s;

$\Delta SNG_{p,s}$: SNG allocation to province p and subsector s;

HV : heating value;

η : energy efficiency;

EF : control technology (t)-specific abated emission factors;

P : penetration rate of a particular control technology (t).

1.3 Evaluating Air Quality Impacts

Weather research and forecasting model coupled with chemistry, version 3.6 (WRF-Chem v3.6) simulates atmospheric transport, chemistry and meteorology simultaneously, and is designed for regional-scale air quality simulations[25]. Our study domain covers East Asia at a horizontal resolution of 27 × 27 km^2 with 31 vertical levels, from the surface to 50hPa, with a 32m thick surface layer. The global 3-D chemical transport model (MOZART-4) with a resolution of 1.9°

latitude × 2.5° longitude provides the chemical initial and boundary conditions (http://www.acom.ucar.edu/wrf-chem/mozart.shtml). Meteorological data for 2015 are from the National Centers for Environmental Prediction (NCEP) Global Forecast System final gridded analysis datasets at a 6-hour resolution (https://www.ncdc.noaa.gov/data-access/model-data/model-datasets/global-forcast-system-gfs). Biogenic emissions are calculated online using the Model of Emissions of Gases and Aerosols from Nature (MEGAN)[26]. Forest and savanna fire emissions are from the Global Fire Data (GFEDV3) (http://www.globalfiredata.org/). Anthropogenic emissions are either from the 2020 ECLIPSE_V5a_CLE scenario (our base case), or our calculated air pollutant emissions under each SNG scenario (Section 1.2), with detailed spatial and temporal allocation described below. The Regional Acid Deposition version 2 (RADM2) atmospheric chemical mechanisms and Modal Aerosol Dynamics model for Europe with the Secondary Organic Aerosol Model (MADE/SORGAM) are used for gas-phase chemistry and aerosol chemistry, respectively.

We estimate provincial total anthropogenic emissions under each medium replacement scenario (Table S1) according to calculations in section 1.2. We assume that, within each province, 2020 emissions follow the same spatial and temporal pattern as that of the 2010 Multi-Resolution Emission Inventory for China (MEIC, http://www.meicmodel.org). National total emissions for regions outside of China in 2020 are also from the ECLIPSE_V5a_CLE scenario, and we assume its spatial and temporal allocation is the same as that of the 2006 INTEX-B Asian emission inventories[27]. Due to data limitations, all anthropogenic emissions are treated in the model surface layer. Spatial distribution of air pollutant emission reductions under various SNG medium scenarios is shown in Figures S2. Due to high computational demands of these simulations, we simulate January, April, July and October as representative months for each

season, with one week at the end of December, March, June and September used for spin-up. The mean $PM_{2.5}$ surface concentrations for the year is assumed to be the average of these four months. For both the base case and SNG medium scenario simulations, we use the same 2015 meteorology. We evaluate the differences of air quality impacts due to various SNG sectoral allocation strategies by comparing the changes of air pollutant concentrations from the base case among SNG medium scenarios.

Based on our simulated grid-level annual average $PM_{2.5}$ surface concentrations and the spatial distribution of China's population[28], we estimate the population-weighted (P-W) ($C_{PW}(P) = \frac{\sum_{i=1}^{n} Pop_i(P) * C_i(P)}{\sum_{i=1}^{n} Pop_i(P)}$) annual average $PM_{2.5}$ surface concentrations at the provincial level. $C_{PW}(P)$ refers to population-weighted $PM_{2.5}$ concentrations in province P, Pop_i refers to population in each grid i within province P, and C_i refers to model simulated $PM_{2.5}$ concentrations in grid i in province P.

1.4 Evaluating Health Impacts

To evaluate the health effects of the SNG allocation scenarios, we estimate the changes in premature mortality that is associated with long-term exposure to ambient $PM_{2.5}$ pollution for both adults (≥ 25 years old) and children (< 5 years old)[29]. In China's many cities, annual average $PM_{2.5}$ concentrations can be as high as 5 times that of the WHO guidelines for annual mean $PM_{2.5}$ concentrations (10 μg/m^3)[30, 31]. Burnett et al. (2014) developed integrated exposure response (IER) functions for $PM_{2.5}$ that cover the global range of exposures, and are suitable for China's high $PM_{2.5}$ concentrations. We use Burnett et al. (2014) relative risk (RR) estimates for ischemic heart disease (IHD), cerebrovascular disease (stroke), chronic obstructive pulmonary disease (COPD), lung cancer (LC), and child acute lower respiratory infection (ALRI). Based on

the IER functions and P-W provincial $PM_{2.5}$ concentrations, we estimate the disease-specific population attributable fraction (PAF) from exposure to ambient $PM_{2.5}$ for each province ($PAF_{i,P}$=1-1/$RR_{i,P}$, where $RR_{i,P}$ is the RR_{IER} values corresponding to the population-weighted $PM_{2.5}$ concentrations for disease i in province P either resulting from the base case or a SNG medium scenario). Thus, premature mortality in each province for disease i due to $PM_{2.5}$ exposure are calculated using:

$$Mort_{i,P} = POP_{j,P} * Mortbase_{i,P} * (1 - 1/RR_{i,P}) \quad (2)$$

where $Mort_{i,P}$ is the number of premature mortality in province P from disease i, $POP_{j,P}$ is the number of exposed targeted population in province P, either adults (≥ 25 years old) for IHD, stroke, COPD and LC, or children (< 5 years old) for ALRI, $Mortbase_{i,P}$ is the baseline mortality rate for either adults or children in province P for disease i. Differences in premature mortality between a medium SNG scenario and the base case are the number of avoided premature mortality due to the specific SNG scenario. Disease-specific mortality rates for adults and children at the provincial level are obtained from the 2013 GBD study (http://www.healthdata.org/gbd). China's projected provincial population in 2020 is from the ECLIPSE_V5a_CLE scenario, and we use the provincial-level age distribution in the 2013 GBD study to estimate the population of adults and children in 2020. We assume that the disease-specific mortality rates remain the same between 2013-2020. Our method conducts PAF calculations separately for baseline and intervention conditions and reports the difference, which differs from some related approaches but produces more conservative estimates of avoided mortality burdens.

2. Model Evaluation

Earlier studies have evaluated WRF-Chem over the U.S. and East Asian regions[25, 32, 33]. Here we evaluate our 2015 simulations with $PM_{2.5}$ monitoring data across China. 2015 model configurations are the same as that described in SI Section 1.3. For regions within China, we use the 2015 ECLIPSE_V5a_CLE anthropogenic emissions at the provincial level, and use the spatial and temporal distribution of emissions from the 2010 Multi-Resolution Emission Inventory for China to distribute the emissions. For regions outside of China, we use the total regional anthropogenic emissions from the 2015 ECLIPSE_V5a_CLE, and use the same spatial and temporal distribution as in the 2006 INTEX-B Asian emission inventory[27]. Simulations are run for January, April, July, and October to represent air quality in winter, spring, summer and fall.

We calculate the monthly mean $PM_{2.5}$ concentrations from each of the four months of 2015 model simulations, and compare them with observations in China's 31 provinces in 2015. Observation data for Beijing is from the Beijing municipal environmental monitoring center (http://www.bjmemc.com.cn); data for other provinces are from the PM25.in project (http://pm25.in). Table S3 describes the monitoring sites. We also compare the observed and simulated daily mean $PM_{2.5}$ concentrations in China's seven regions (Figure S3). Our simulations generally capture the seasonal and monthly trends in $PM_{2.5}$ concentrations in most parts of China. However, noticeable model underestimation is observed in northeastern and northwestern provinces. This underestimation may occur for a variety of reasons. One possibility is that the GAINS model estimates lower anthropogenic emissions than what actually occurred in these provinces in 2015. Additionally, China's current $PM_{2.5}$ monitoring sites are mostly located in city centers. This is particularly the case in northwestern provinces where population and emission distribution is sparse. Thus the model resolution of our simulations may miss urban hot spots in

these regions. Moreover, our model simulation does not include dust that, particularly in winter and spring, likely contributes to the underestimation of northwestern $PM_{2.5}$ concentrations. Overall, however, our model does a reasonable job when compared with observation data, particularly in eastern China, which is the major focus of this study (Figure S3).

3. Synthetic Natural Gas Production -- Energy Penalty of Carbon Capture and Storage

During SNG production, coal is first converted to syngas in the gasifier generally at a high pressure. CO_2 emitted from this process is at a relatively high partial pressure thus requires low energy for CO_2 separation[18]. In addition, CO_2 is separated from syngas before methanation regardless of conducting CCS as the cost for capturing CO_2 is more than paid back by cost savings from better methanation efficiency (due to higher CO and H_2 concentrations) and lower volume of input syngas (smaller reactor volume and costs)[18, 34]. Thus, the energy penalty for conducting CCS at SNG plants is substantially lower than that at coal combustion power plants[18]. Liu et al. (2011) estimated that the energy penalty is 91 kWh per tonne CO_2 captured for coal-based synthetic fuels production (energy needed for compressing CO_2 to 150 bar for pipeline transport to underground storage)[34]. We assume this penalty is representative of SNG plants. Electricity needed for conducting CCS along with SNG production can either be generated from gas- or coal- fired power plants. In China, over 98% of existing gas-fired power generation units were natural gas combined cycle (NGCC) in 2012[11]. Thus, we use NGCC to represent gas-fired power generation in 2020. For coal-fired power plants, we use supercritical, ultra-supercritical and integrated gasification combined cycle (IGCC) power plants to represent the 2020 fleets. For each type of power plant we examine the implications of including/excluding CCS. We assume CCS can capture 90% of CO_2 emissions during SNG production or power plants operation[35]. Thus, when CCS is applied during SNG production, only 10% of CO_2 produced by this process

will be emitted to the atmosphere. Power plants providing electricity also emit CO_2 to the atmosphere: either 100% of CO_2 produced from electricity generation (power plants without CCS) or 10% of CO_2 produced from electricity generation (power plants with CCS).

Below we estimate the energy penalties and CO_2 emissions for each type of power plant (with and without CCS), and illustrate the calculation processes for 1 MJ coal as energy input using equations 1-6. Energy efficiencies for various power plant types, with and without CCS, and the calculated net energy penalties and CO_2 removal efficiencies are listed in Table S4.

1) SNG production and associated CO_2 emissions from 1MJ coal input:

$$\mathbf{V_{SNG} = 1\ MJ * \eta_1 / HV_{SNG}} \qquad \mathbf{(1)}$$

$$\mathbf{E1_{CO2} = V_{SNG} * EF1_{CO2}} \qquad \mathbf{(2)}$$

$\mathbf{V_{SNG}}$ refers to the volume (m^3) of SNG produced from 1 MJ coal input; $\boldsymbol{\eta_1}$ refers to coal-to-SNG conversion efficiency (51%)[11]; $\mathbf{HV_{SNG}}$ refers to SNG heating value, 34 MJ/m^3; $\mathbf{E1_{CO2}}$ refers to CO_2 emissions from SNG production processes (kg); $\mathbf{EF1_{CO2}}$ refers to CO_2 emission factors for SNG production processes not using CCS, 4.25 kg CO_2/m^3 SNG.

2) Electricity needed to capture CO_2 generated from SNG production:

$$\mathbf{ELE = EP*E1_{CO2}/1000} \qquad \mathbf{(3)}$$

ELE refers to total electricity needed to conduct CCS during SNG production (kWh), **EP** refers to the energy penalty for applying CCS for synthetic fuels production, 91 kWh/tonne CO_2.

3) Energy consumption to provide electricity for applying CCS during SNG production:

$$\mathbf{ENE = ELE * 3.6\ (MJ/kWh) / \eta_2} \qquad \mathbf{(4)}$$

ENE refers to energy consumption to provide electricity for applying CCS during SNG production (MJ); η_2 refers to power plant efficiency (%).

4) Net energy penalty for conducting CCS during SNG production:

$$NEP = \eta_1 / [(1\ MJ * \eta_1 - ENE) / 1MJ] - 1 \quad (5)$$

NEP refers to net energy penalty (%). This uses the equation from Metz et al. (2005)'s study: $\Delta E = (\eta_{w/o\ ccs} / \eta_{ccs}) - 1$.

5) Net CO_2 emissions from each system in Table S4:

$$E2_{CO2} = E1_{CO2} * (1-90\%) + ENE * EF2 * CCS_{y/n} \quad (6)$$

$$RMF = (1- E2_{CO2} / E1_{CO2}) * 100\% \quad (7)$$

$E2_{CO2}$ refers to total CO_2 emissions from each power plant-SNG system in Table S4 (kg). **EF2** refers to CO_2 emission factors for fossil fuel (coal or gas) combustion in the power plants without CCS. We use the values from the GAINS model, which are 94.3 g CO_2/MJ coal and 55.8 g CO_2/MJ natural gas (Table S4). For unit consistency, we convert the CO_2 EFs to kg CO_2/MJ for use in Eq. 6. $CCS_{y/n}$ refers to whether CCS is applied in the power plant, it equals 0.1 if applied, 1 if not. **RMF** refers to net CO_2 removal efficiency from each power plant-SNG system shown in Table S4.

4. Regional SNG Allocation

In addition to the three major sectoral SNG allocation scenarios, we also build two regional allocation scenarios (each with 3 sub-scenarios) to evaluate the impacts of regional SNG distribution on air quality, human health and CO_2 emissions (Table S1). These allocation scenarios examine the benefits of SNG substitution for coal by giving all SNG-receiving provinces equal priority in SNG allocation, or giving provinces with political or economic importance a higher priority.

Following the same processes described from Section 1.2 to 1.4, we use the WRF-Chem model to estimate the net changes in monthly mean $PM_{2.5}$ surface concentrations for the two medium regional SNG scenarios (Fig.S4 d-e). Comparing the two medium regional allocation scenarios, grid-level $PM_{2.5}$ concentration reductions are generally below 5 $\mu g/m^3$ when available SNG is allocated proportionally to each province and each sector's original gas needs for coal replacement (SNG_Region_Sector). However, the maximum grid-level $PM_{2.5}$ concentration reductions in Beijing are over 5 $\mu g/m^3$ year-round and can reach ~60 ug/m^3 in winter when emphasizing gas allocation to affluent provinces (SNG_Province). Notably, differences in $PM_{2.5}$ concentration reductions outside Beijing between the two scenarios are negligible.

We estimate the national total avoided premature deaths under the two regional allocation scenarios (SNG_Region_Sector and SNG_Province) are 2,700 (1,300-3,500) and 3,100 (1,800-3,600), respectively, varying depending on the low and high estimates of relative risks. The national total health impacts of the two regional allocation scenarios are similar to that of the SNG_industry scenario. In addition, the impacts of these two regional allocation scenarios on CO_2 emissions are nearly identical, with an additional ~40% increases in CO_2 emissions for SNG without conducting CCS compared with direct combustion of coal (Table S5).

To sum up, prioritizing SNG allocation to affluent provinces significantly improves the air quality in Beijing in comparison to an equal-regional-priority SNG allocation. However, the national total air quality and human health benefits and carbon penalties are similar between these two regional allocation scenarios.

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Appendix for Chapter 4.

The Air, Carbon, Water Synergies and Trade-offs in China's Natural Gas Industry

Supporting Method and Data

1. Method

This study uses an integrated assessment approach coupled with lifecycle analysis to evaluate the air quality, carbon, and water impacts for six natural gas sources under three different end-use coal-to-gas deployment strategies (Table S1).

1.1 Environmental Impacts from End-Use Coal Substitution with Natural Gas

We use the ECLIPSE_V5a_CLE (Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants) emission scenario developed by the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) model (http://gains.iiasa.ac.at/models/) as our 2020 baseline anthropogenic emissions input[1]. As described in Qin et al. (2017)[2], the ECLIPSE scenario provides detailed subsector technology information (i.e. power plant fleet types), end-use energy consumption data, and emissions for major air pollutants and CO_2 at the provincial level, though without cooling technology information for end-use combustion. We obtain China's provincial-level cooling technology information for coal-fired power plants from the World Electric Power Plants (PLATTS-WEPP) database in 2013 June version (https://www.platts.com/products/). We assume coal-fired power plants in China that are currently under operation, construction or planned before 2018 that are summarized in the database represent China's coal-fired power plant fleet in 2020. We calculate the percent penetration of various cooling technologies used on coal-fired power plants within each province from the database when cooling technologies information are available and apply the penetration rates to coal-fired power plants in the GAINS model. Table S2 shows provincial-level penetration rates of various cooling technologies used in this study in 2020.

Among various cooling technologies, both once-through and recirculating cooling are mature technologies. In particular, once-through cooling, with the lowest capital cost, has been

employed for decades[3]. In comparison, recirculating cooling, with higher capital cost but substantially lower water withdrawal than once-through cooling, is a standard choice for most newer plants[4]. Dry cooling, on the other hand, which has the lowest water consumption but requires significantly higher capital cost and power consumption, has only been widely used recently[4]. Thus, the oldest plants typically use once-through cooling, newer plants use recirculating cooling and the newest plants, particularly in regions of high water stress typically employ dry cooling. Similarly, in a certain province, more advanced coal-fired power plants are usually built more recently. Therefore, for each province with a certain penetration rate of dry cooling as shown in Table S2, we assume that dry cooling technology is allocated to coal-fired power plants in the order of PP_MOD (ultra-supercritical and supercritical), PP_NEW_L (subcritical), and PP_EX_L (very high pressure and high pressure) as categorized in the GAINS model. After allocating all dry cooling technologies in each province, we then allocate recirculating cooling following the same order. Additionally, the same type of coal-fired power plants with more advanced air pollution control technologies within each province are also usually built more recently due to technology improvement and increasingly stricter environmental regulations. Thus, when allocating dry cooling or recirculating cooling technology among the same type of coal-fired power plants in each province, we first allocate it to power plants with more advanced air pollution control technologies. The rest of coal-fired power plants in each province are equipped with either once-through-fresh, or once-through-brackish, or once-through-saline cooling technologies. As the choice of these three once-through cooling technologies are not directly determined by power fleet types, we proportionally allocate once-through-fresh cooling technology among the rest of coal-fired power plants based on its penetration rate in each province shown in Table S2. Following these assumptions, we integrate provincial-level cooling technology information with coal-fired power plants information in the GAINS model in each province accordingly. This study focuses on freshwater use, thus we only evaluate water consumption changes due to a coal-to-gas substitution when accompanied by dry cooling, recirculating cooling, or once-through-fresh cooling (referred to as 'once-through cooling hereafter') technologies. Thus, water consumption changes due to once-through saline and once-through brackish cooling technologies are not included. In addition, following earlier studies, we assume industrial boilers generally use recirculating cooling[5]. Table S3 shows water

consumption coefficients for coal- and gas- fired power plants and industrial boilers. Notably, when evaluating the water impacts of each coal-to-gas scenario, we assume only the fuel changes while the cooling technology stays the same.

As a thought experiment, we assume an additional gas supply of 30 billion cubic meters (bcm) above the baseline for each gas source in turn, to replace coal under each scenario. This is roughly the quantity of China's currently planned increase for most gas sources in 2020[6-8]. For each gas source, Table S4 summarizes the provinces from which the gas originates and in which it is consumed. Natural gas is used to substitute for coal in the residential, industrial, and power sectors, which are China's major natural gas end-uses[9]. Following the regional and sectoral natural gas deployment requirements, 30 bcm of each gas source is then deployed to the region and subsector that will either achieve the largest reductions in SO_2 emissions, or CO_2 emissions, or water stress index (WSI)-weighted water consumption (referred hereafter as 'weighted water consumption') under the three deployment strategies (Table S1). Here we assume that China's provincial-level WSI in 2020 stays the same as that reported in Feng et al. (2014)[10]. We next quantify the quantity of coal substituted by natural gas in each subsector within each province for each gas source under each scenario using the GAINS model. End-use air pollutants and CO_2 emission reductions in each province are calculated using equation 1. End-use weighted water consumption reductions in each province are calculated using equation 2.

$$\Delta E_{a,p,s} = \sum[\Delta Gas_{p,s,t} * (\frac{HV_{Gas}}{HV_{Coal}} * \frac{\eta_{Gas,s}}{\eta_{Coal,s}} * EF_{Coal,a,p,s,t} - EF_{Gas,a,p,s,t})] \quad (1)$$

$\Delta E_{a,p,s}$: Emission reductions for pollutant type a, in province p and subsector s;

$\Delta Gas_{p,s,t}$: Additional natural gas allocation to province p in subsector s under control technology t;

HV: Heating value in the GAINS model[2];

η: Energy efficiency for various end-use technologies[2];

EF: Control technology (t)-specific abated emission factors.

$$\Delta WWC_{p,s} = WSI_p * \sum[\Delta Gas_{p,s,t} * (\frac{HV_{Gas}}{HV_{Coal}} * \frac{\eta_{Gas,s}}{\eta_{Coal,s}} * WCR_{Coal,s,c} - WCR_{Gas,s,c})] \quad (2)$$

$\Delta WWC_{p,s}$: Weighted water consumption reductions in province p and subsector s;

$\Delta Gas_{p,s,c}$: Additional natural gas allocation to province p in subsector s under cooling technology c;

WSI_p: Water stress index at the provincial level, ranging from 0 (no stress) to 1 (maximum stress)[10];

$WCR_{Coal,s,c}$: Cooling technology (c)-specific water consumption rates.

1.2 Environmental Impacts of Upstream Processes

Table S5 shows the upstream stages considered in this study for coal and for each of the six natural gas sources. We use the lifecycle assessment framework built in Qin et al. (2017)[11] to estimate upstream greenhouse gas (GHG) (CO_2 and CH_4) emission intensity, upstream air pollutant emission intensity, and upstream water consumption intensity, based on stage-level energy consumption, methane leakage rates, and the corresponding fuel-specific emission factors and water consumption coefficients.

Qin et al. (2017)[11] estimated lifecycle GHG emissions for China's conventional natural gas, shale gas, and coal industries, which are used in this study. In addition, Qin et al. (2017)[11] described efforts of U.S. researchers to directly measure methane leakage rates from natural gas systems in recent years[12-16]. However, most other countries still rely on the outdated 2006 IPCC guidelines to indirectly estimate fugitive methane emissions[17-19]. Thus, due to data deficiency, we apply the latest stage-level U.S. average methane leakage rates and the uncertainty ranges for the conventional natural gas industry summarized in Qin et al. (2017)[11] (total leakage: 1.4%, 0.1%-6.0%) to other natural gas producing countries from which China imports natural gas (Table S4). Similarly, we assume upstream stage-level energy consumption for the natural gas industry in these developing and developed countries are the same as that in China[11] and the U.S.[20], respectively.

Imported pipeline gas must go through long-distance international pipelines before reaching China's border. We obtain the average international gas transmission distances from Kong et al. (2006)[21]. Country-specific fugitive emission factors of international gas pipelines reported in Hoglund-Isaksson et al. (2012)[19] are used in this study. As eastern Russian gas is transported via newly-built pipelines[22], we assume its fugitive emission factors are the same as those in developed countries: 10 kg CH_4/(bcm*km)[19]. After reaching China, imported pipeline gas will go through the same transmission and distribution processes as China's conventional natural gas. Table S6 summarizes the average international and domestic gas transmission distances for each gas source.

Compared to imported pipeline gas, imported LNG goes through additional steps including liquefaction, tanker transport, and regasification. Jaramillo et al. (2007)[23] reported CO_2 and CH_4 emission factors from LNG liquefaction based on earlier measurements[24]. These emission factors are adjusted by the latest global warming potential (GWP) values reported in the IPCC AR5 report to obtain an updated CO_{2eq} emission factor (Table S7)[25]. Overseas LNG is transported via large tankers with capacities ranging from 75,000 to 265,000 m^3 of LNG[26]. Tanker capacity is the single most important factor influencing the amount of GHG emissions during LNG transport with smaller tanks emitting more per unit LNG transported[26]. Thus, we estimate GHG emissions from LNG transport using both the smallest and largest tanker size to obtain a full range of shipping GHG emissions. Following earlier studies, we assume the tanker travels round trip. It uses a 0.15% boil-off rate to power steam engines with fuel oil to supplement the boil-off gas if needed when transporting LNG to China, and exclusively uses fuel oil for the returning trip[23, 26]. Additionally, we follow the assumption that either 3% of gas, or 0.15% of gas together with electricity, is consumed to run regasification equipment[23]. For the latter case, Jaramillo et al. (2007)[23] assumed additional electricity was generated from clean energy. Here we update his estimate with reported China's GHG emission intensity for electricity generation, which is dominated by coal[27]. After regasification, LNG is usually distributed locally via low-pressure distribution pipelines, the same as the distribution process for China's conventional gas.

Upstream SNG processes are a combination of coal and gas processes (Table S5). Stage-level emissions for coal processes are from Qin et al. (2017)[11]. As SNG projects are located close to coalmines, only short-distance (~50 km) coal transport occurs[28]. Emission factors for SNG production are 0.0016 kg CH_4/m^3 and 4.25 kg CO_2/m^3 [28]. Table S8 summarizes the upstream GHG emission intensities for each gas source and coal.

Similarly, upstream air pollutant emission intensity for coal and natural gas industries is calculated based on stage-level fuel consumption and methane leakage rates summarized in Qin et al. (2017)[11], as well as fuel-specific air pollutant emission factors (EFs) reported in the GAINS model[1]. We use the activity data (AD)-weighted air pollutant emission factors in the industrial sector in each gas producing country as the mean EFs, with the minimum and maximum EFs in the industrial sector in each country as the lower and upper bound estimates of the fuel-specific air pollutants EFs (Fig. S1).

Upstream water consumption intensity is calculated with fuel-specific water consumption rates reported in earlier studies (Fig. S2)[4, 29-31] and stage-level methane leakage rates[11]. As we only focus on freshwater uses, water use during imported LNG liquefaction, tanker transport and regasification, which primarily consumes seawater, are not considered[32]. Upstream air pollutant emission intensities and water consumption intensities for various gas sources and coal are also shown in Table S8. Notably, as China's LNG is primarily from Qatar (34%), Australia (19%), Malaysia (15%), and Indonesia (12%)[33], we use the activity data weighted emission and water consumption intensities of LNG produced in these four countries to represent the emission and water consumption intensities of China's imported LNG.

We then calculate the changes in upstream air pollutant emissions, GHG emissions, and water consumption for various gas sources and coal using the corresponding emission and water consumption intensities (Table S8), region-specific gas production (Table S4), and coal reduction quantity (Section 1.1). For simplification, we assume upstream air pollutant emissions, GHG emissions, and water consumption for gas processes occur in places where natural gas is produced. In comparison, the spatial distribution of reduced emissions and water consumption due to avoided upstream coal processes are identified according to where end-use coal reduction occurs and the corresponding source-receptor matrix of provincial coal production and consumption[34]. Natural gas and coal losses during upstream stages are reflected in our calculated upstream emission and water consumption intensities. In addition, to reflect regional water impacts, we use water stress index (WSI) in both China (at provincial level) and other gas-producing countries (at country level) reported in Feng et al. (2014)[10] and Pfister et al. (2009)[35] to estimate changes in weighted water consumption. We assume the water stress index in 2020 for countries outside China and provinces within China remain the same as that reported in Feng et al. (2014)[10] and Pfister et al. (2009)[35].

Combing upstream and end-use processes, we obtain net changes in air pollutant emissions, GHG emissions, and weighted water consumption for each gas source under each coal-to-gas scenario.

1.3 Simulated Air Quality Impacts

To reflect regional air pollution exposure, we further evaluate changes in population-weighted (P-W) $PM_{2.5}$ surface concentrations ($C_{PW} = \frac{\sum_{i=1}^{n} Pop_i * C_i}{\sum_{i=1}^{n} Pop_i}$) for each gas source under each end-use scenario using the weather research and forecasting model coupled with chemistry (WRF-Chem v3.6)[36]. C_{PW} refers to China's P-W $PM_{2.5}$ concentrations, Pop_i refers to population in each grid i, and C_i refers to model simulated $PM_{2.5}$ concentrations in grid i. We use the same model configurations as that in Peng et al. (2017)[37]. Our model domain is set to cover East Asia, with a horizontal resolution of 27 × 27 km^2 and a vertical resolution of 31 vertical levels (~30 m thick surface layer). Chemical initial and boundary conditions (1.9° latitude × 2.5° longitude) are provided by the global 3-D chemical transport model for Ozone and Related Tracers (MOZART-4). 2014 meteorological data (https://www.ncdc.noaa.gov/data-access/model-data/model-datasets/global-forcast-system-gfs), a non-El Nino year, is used for all of our 2020 model simulations. We use the Model of Emissions of Gases and Aerosols from Nature (MEGAN) to calculate biogenic emissions online[38]. The Global Fire Emissions Database (GFED) v4 (http://dx.doi.org/10.3334/ORNLDAAC/1293) provides fire emissions.

ECLIPSE_V5a_CLE emission scenario provides 2020 baseline anthropogenic emissions at the provincial level. We also estimate total anthropogenic air pollutant emissions in each province for 18 combinations of gas sources and end-use scenarios based on upstream and end-use emission changes occurring within our model domain. Within each province, we assume that the 2020 spatial and temporal emission patterns are the same as that in the 2012 Multi-Resolution Emission Inventory for China (MEIC, http://www.meicmodel.org). Anthropogenic emissions outside of China in 2020 are also calculated from the ECLIPSE_V5a_CLE emission scenario. We assume its spatial and temporal pattern follows the 2010 HTAP emission inventories (http://edgar.jrc.ec.europa.eu/htap_v2/). For each scenario, we simulate $PM_{2.5}$ surface concentrations in January, April, July and October, and use the average to represent annual mean $PM_{2.5}$ surface concentrations. Refer to Peng et al. (2017)[37] for model evaluation.

Table S1. Natural gas source choices and end-use deployment strategies for coal replacement by 30 billion cubic meter of gas.

Deployment Strategies	Gas Sources					
	Domestic Conventional Gas	Domestic SNG	Domestic Shale Gas	Imported LNG	Imported Eastern Russia Gas	Imported Central Asia Gas
Air quality-focused substitution (AS)	Each source of natural gas is allocated to targeted provinces[1] and subsectors[2] to replace coal in a way that achieves the largest SO_2 emission reductions possible with end-use substitution[3]					
Carbon-focused substitution (CS)	Each source of natural gas is allocated to targeted provinces[1] and subsectors[2] to replace coal in a way that achieves the largest CO_2 emission reductions possible with end-use substitution[3]					
Water-focused substitution (WS)	Each source of natural gas is allocated to targeted provinces[1] and subsectors[2] to replace coal in a way that achieves the largest weighted water consumption reductions possible with end-use substitution[3,4]					

[1]Targeted provinces for each gas are illustrated in Fig. 1 and summarized in Table S4.

[2] End-use coal-to-gas substitution from three major gas consumption sectors, including the power, residential, and industrial sectors, is included in the analysis. Each major sector includes various subsector technologies, which are described in the GAINS model in detail (http://gains.iiasa.ac.at/models/).

[3]In order to achieve the largest possible SO_2 (CO_2 or weighted water consumption) reductions, both SO_2 emission factors (CO_2 emission factors or weighted water consumption coefficients) and energy efficiencies of coal and natural gas combustion in various subsectors are considered. Energy efficiencies for various end-use technologies in the GAINS model are summarized in Qin et al. (2017)[2].

[4]End-use coal-to-gas substitution occurs within China. End-use cooling water consumption is weighted by China's provincial-level water stress index (WSI)[10] to reflect region-specific water impacts, which are referred as weighted water consumption.

Table S2. Provincial-level penetration rates of various cooling technologies in 2020.

Province	Dry	Recirculating	Once-through-fresh	Once-through-brackish	Once-through-saline
Anhui	0	0.97	0.03	0	0
Beijing	0	1	0	0	0
Chongqing	0	0.47	0.53	0	0
Fujian	0	0.07	0	0	0.93
Gansu	0.85	0.15	0	0	0
Guangdong	0	0.06	0	0	0.94
Guangxi	0	0.55	0.15	0	0.3
Guizhou	0	1	0	0	0
Hainan	0	0	0	0	1
Hebei	0.31	0.43	0.08	0	0.17
Heilongjiang	0	1	0	0	0
Henan	0	0.99	0.01	0	0
Hong Kong	0	0	0	0	1
Hubei	0	0.81	0.19	0	0
Hunan	0.4	0.2	0.4	0	0
Inner Mongolia	0.77	0.23	0	0	0
Jiangsu	0.15	0.44	0.01	0.4	0
Jiangxi	0.27	0.53	0.2	0	0
Jilin	0.23	0.77	0	0	0
Liaoning	0.06	0.55	0	0	0.39
Ningxia Hui	0.56	0.44	0	0	0
Qinghai	0.72	0.28	0	0	0
Shaanxi	0.71	0.29	0	0	0
Shandong	0	0.71	0	0.07	0.22
Shanghai	0	1	0	0	0

Shanxi	0.58	0.42	0	0	0
Sichuan	0	1	0	0	0
Tianjin	0	1	0	0	0
Xinjiang	0.8	0.2	0	0	0
Yunnan	0	1	0	0	0
Zhejiang	0	0.17	0	0	0.83

Data source: https://www.platts.com/products/. Our study focuses on freshwater impacts, thus we include only dry, recirculating, and once-through-fresh (referred as once-through in this study) cooling technologies in our analysis on weighted water consumption.

Table S3. Water consumption coefficients for end-use combustion technologies using various cooling technologies (Unit: g/MJ).

Technology Type*	Cooling Technology	Water Consumption (mean-low-high)
Gas technologies		
	Once-through	105 (21-242) [a]
	Recirculating	221 (49-316) [a]
	Dry	2 (0-4) [b]
Industrial Gas Boiler	Recirculating	127 (29-182) [c]
Coal technologies		
PP_MOD	Once-through	74 (51-107) [a]
PP_NEW_L	Once-through	95 (47-136) [a]
PP_EX_L	Once-through	154 (75-258) [a]
PP_MOD	Recirculating	461 (383-509) [a]
PP_NEW_L	Recirculating	525 (436-631) [a]
PP_EX_L	Recirculating	634 (475-862) [a]
PP_MOD	Dry	91 (65-117) [a]
PP_NEW_L	Dry	116 (69-164) [a]
PP_EX_L	Dry	164 (139-189) [a]
Industrial Coal Boiler	Recirculating	245 (204-294) [c]

* End-use technology types defined in the GAINS model. PP_MOD, PP_NEW_L, and PP_EX_L correspond to ultra-supercritical and supercritical (MOD), subcritical (NEW_L), and very high pressure and high pressure (EX_L) coal-fired power plants, respectively. Relative ratios of ultra-supercritical, supercritical, very high pressure and high pressure fleets are taken from Shen et al. (2017)[39].

[#] NGCC: Natural Gas Combined Cycles.

Data source: [a]Zhang et al. (2016)[40], [b]Macknick et al. (2011)[41]. [c] Water consumption rates for industrial coal (gas) boilers are estimated based on water consumption rates for PP_NEW_L (NGCC) and their relative thermal efficiency reported in Qin et al. (2017)[2].

Table S4. Provinces in which natural gas is produced and potentially consumed for each gas source*.

Gas Sources		Producing Provinces	Target Provinces
	Conventional Gas[1]	Beijing (12.8), Hebei (17.5), Tianjin (21.15), Shanxi (31.6), Heilongjiang (35.39), Jilin (8.11), Liaoning (15.45), Inner Mongolia (22.28), Shandong (2.12), Jiangsu (0.52), and Shanghai (4.92), Jiangxi (0.43), Henan (4.87), Hubei (1.45), Guangdong (83.66), Guangxi (0.16), Hainan (1.58), Chongqing (7.78), Sichuan (253.53), Guizhou (0.4), Yunnan (0.02), Shaanxi (410.11), Gansu (0.15), Qinghai (68.9), Xinjiang (296.7)	Across Mainland China
	SNG[2]	Xinjiang (13), Inner Mongolia (11), Shanxi (3), and Liaoning (3)	Xinjiang SNG: Xinjiang, Gansu, Ningxia, Shaanxi, Shandong, Henan, Hubei, Hunan, Jiangxi, Fujian, Zhejiang, Guangdong and Guangxi provinces Inner Mongolia SNG: Inner Mongolia, Beijing, Hebei, Tianjin, Jilin Shanxi and Liaoning SNG: consumed within each province
	Shale Gas[3]	Sichuan (15), and Chongqing (15)	Sichuan, Chongqing, Hunan, Hubei, Jiangxi, Anhui, Jiangsu, Zhejiang, and Shanghai
	LNG[4]	Qatar, Australia, Malaysia, and Indonesia	Guangdong (8), Fujian (4), Zhejiang (3), Jiangsu (3), Hebei (3), Tianjin (3), Liaoning (2), Guangxi (2), Shanghai (1), Shandong (1), and Hainan (1)

	Eastern Russia Gas[5]	Eastern Russia	Heilongjiang, Jilin, Inner Mongolia, Liaoning, Beijing, Hebei, Tianjin, Shandong, Jiangsu, and Shanghai
	Central Asia Gas[6]	Central Asia	Xinjiang, Gansu, Ningxia, Shaanxi, Henan, Hubei, Hunan, Jiangxi, Fujian, Anhui, Zhejiang, Shanghai, Guangdong, and Guangxi

*Numbers in the parentheses indicate the quantity of gas production or consumption at the unit of billion cubic meters (bcm).

[1] For conventional natural gas, we assume the provincial production ratio in 2020 is the same as that reported in 2015[9]. Conventional natural gas is consumed across Mainland China[9].

[2] For SNG, we assume the provincial production ratio in 2020 is the same as that of government-approved SNG projects as of 2013[42, 43]. SNG potential consumption regions are summarized in Qin et al. (2017)[2].

[3] Shale gas development is primarily concentrated in Sichuan and Chongqing provinces, thus we assume a 15 bcm of gas production in each province. Shale gas can potentially be consumed in our listing targeting provinces through the "Chuan Qi Chu Chuan" and "Chuan Qi Dong Song" gas transmission projects.

[4] Imported LNG is mainly produced in Qatar (34%), Australia (19%), Malaysia (15%), and Indonesia (12%) in 2014[33], which we assume is also true in 2020. LNG receiving terminals are built along China's coastal provinces that are usually far away from domestic gas fields. After receiving LNG, it is mainly consumed within the province. We assume the provincial allocation of LNG in 2020 is proportional to China's LNG importing facilities as of 2014[44].

[5] Imported Eastern Russia gas, after reaching China, is mainly consumed in northern and northeastern provinces[22].

[6] Imported Central Asia gas, after reaching China, is mainly transported to eastern and southeastern China via the 2nd and 3rd West-East Gas Pipelines[8].

Table S5. Upstream stages included in this study for each gas source.

Fuel Types	Gas Sources	Upstream Stages			
	Conventional Gas	Gas production & gathering	Gas processing	Gas transmission & storage	Gas distribution
	SNG	Coal mining & processing & transport	Coal gasification & methanation	Gas transmission & storage	Gas distribution
	Shale Gas	Gas (pre-) production & gathering	Gas processing	Gas transmission & storage	Gas distribution
	Eastern Russia Gas	Gas production & gathering	Gas processing	Gas transmission & storage	Gas distribution
	Central Asia Gas	Gas production & gathering	Gas processing	Gas transmission & storage	Gas distribution
Coal	China Coal	Coal mining	Coal processing	Coal transport	

Table S6. Average international and domestic transport distances for each gas source (km).

	International Transport	Domestic Transport
Conventional Gas	-	1500[c]
SNG	-	2000[d]
Shale Gas	-	900[d]
Central Asia	2006[a]	2000[d]
Eastern Russia Pipeline Gas	2680[b]	1500[d]
Australia LNG	4372 [a]	-
Qatar LNG	8136 [a]	-
Indonesia LNG	1866 [a]	-
Malaysia LNG	1866 [a]	-

[a]Kong et al. (2016)[21]; [c]Ou et al. (2013)[45], [d]Author summary; imported LNG is assumed to be mainly consumed within the importing province after regasification

[b]http://www.cnpc.com.cn/cnpc/jtxw/201409/462ab7e154ec4bd1b1a2878e4bd7b0d7.shtml

Table S7. Greenhouse gas emission factors for LNG liquefaction (mean, min-max) (Units: g CO_2eq/MJ). Adapted from Jaramillo et al. (2007)[23].

CO_2 from fuel combustion	5.2 (4.7-5.6)
CO_2 from flare combustion	0.3 (0-0.6)
CH_4 leakage from vent	0.6 (0-4.7)
CO_2 from raw gas	1.7 (0-2.8)

Table S8. Upstream emission (major air pollutants and CO_{2eq}) and water consumption intensities for coal and each gas source sources (Unit: g/MJ)* (Mean, Low-High)

Fuel Types	SO_2	NO_x	$PM_{2.5}$	CO_{2eq} (GWP_{100})	CO_{2eq} (GWP_{20})	Water Consumption
Coal	0.036 (0.005-0.16)	0.027 (0.02-0.04)	0.003 (0.001-0.02)	19 (9-19)	43 (15-67)	20 (14-29)
Conventional Natural Gas	0.036 (0.014-0.12)	0.036 (0.02-0.06)	0.003(0.001-0.03)	20 (13-52)	36 (14-131)	11 (10-11)
SNG	0.066 (0.007-0.30)	0.042 (0.03-0.06)	0.006 (0.002-0.03)	164 (141-190)	225 (157-294)	287 (203-418)
Shale Gas	0.037 (0.015-0.12)	0.038 (0.02-0.06)	0.003 (0.001-0.03)	25 (12-94)	44 (14-229)	23 (20-30)
Imported LNG	0.048 (0.02-0.28)	0.056 (0.03-0.15)	0.003 (0.001-0.08)	32 (24-79)	48 (25-163)	9 (8-14)
Imported Eastern Russia Gas	0.058 (0.04-0.11)	0.041 (0.03-0.06)	0.003 (0.002-0.03)	22 (14-55)	39 (16-135)	11 (10-12)
Imported Central Asia Gas	0.078 (0.06-0.14)	0.043 (0.03-0.06)	0.003 (0.001-0.03)	22 (14-55)	39 (16-135)	11 (10-12)

*Upstream emission intensity is calculated as the total emissions or water consumption during fuel production, processing, transmission, and distribution stages divided by total net energy produced that can be used for end-use combustion. Energy produced at the well minus energy losses during upstream processes provides the net energy produced for end-use combustion. Values for imported LNG is the quantity-weighted average for LNG produced in Qatar, Australia, Indonesia, and Malaysia.

Table S9. Water consumption coefficients for energy extraction, processing and transport (Unit: g/MJ)*.

	Coal	Conventional Gas	SNG	Shale Gas
Production	**17 (12-24)[46]**	**1.3 (1-1.6)[29, 47]**	**250 (176-353)[42]**	**13 (11-17)[31, 48]**
Processing	**2.2 (1.5-3.1)[4, 47]**	**6[3, 30]**	**6[3, 30]**	**6[3, 30]**
Transport	**-**	**3[3, 30]**	**3[3, 30]**	**3[3, 30]**

*Here we summarize stage-level water consumption coefficients based on earlier studies. About 43% of coal is washed during coal processing[47]. Also, water consumption intensity for China's shale gas wells are estimated to be about 50% higher than that in the U.S[48].

Upstream water consumption intensities in Table S8 are obtained based on this information and stage-level methane leakage rates. As we assume China's imported natural gas is primarily conventional natural gas, imported gas has the same stage-level water consumption coefficients as that for conventional gas. However, the total upstream water consumption intensities are different due to extra methane leakage or gas consumption during long distance natural gas transmission.

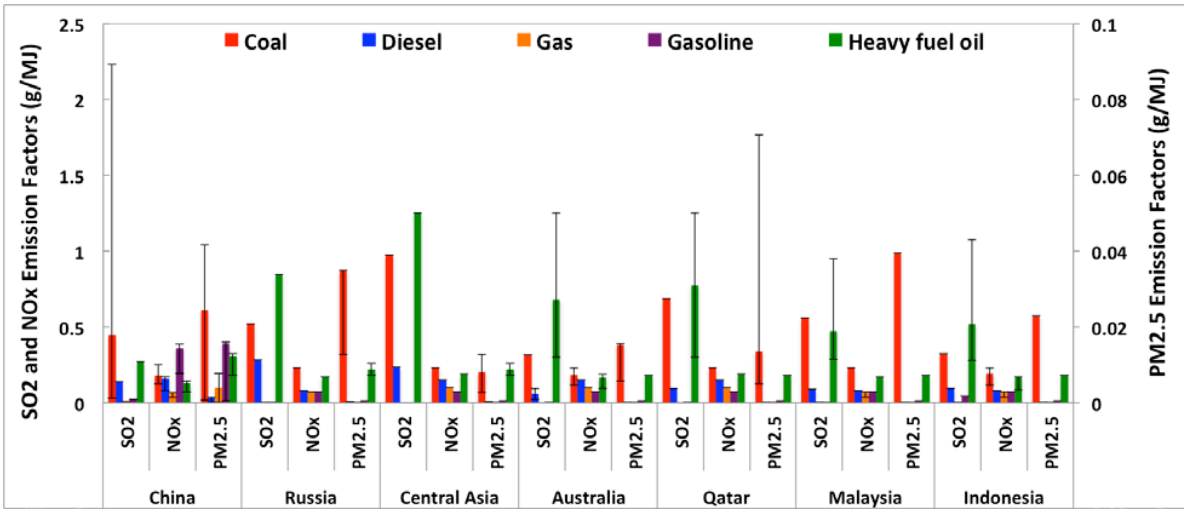


Fig. S1. Region-specific major air pollutants (SO_2 and NO_x, left axis; $PM_{2.5}$, right axis) emission factors for energy consumption during upstream natural gas and coal processes (production, processing, transmission, and distribution) in the GAINS model. Stage-level energy consumption and energy mix are summarized in Qin et al. (2017)[11].

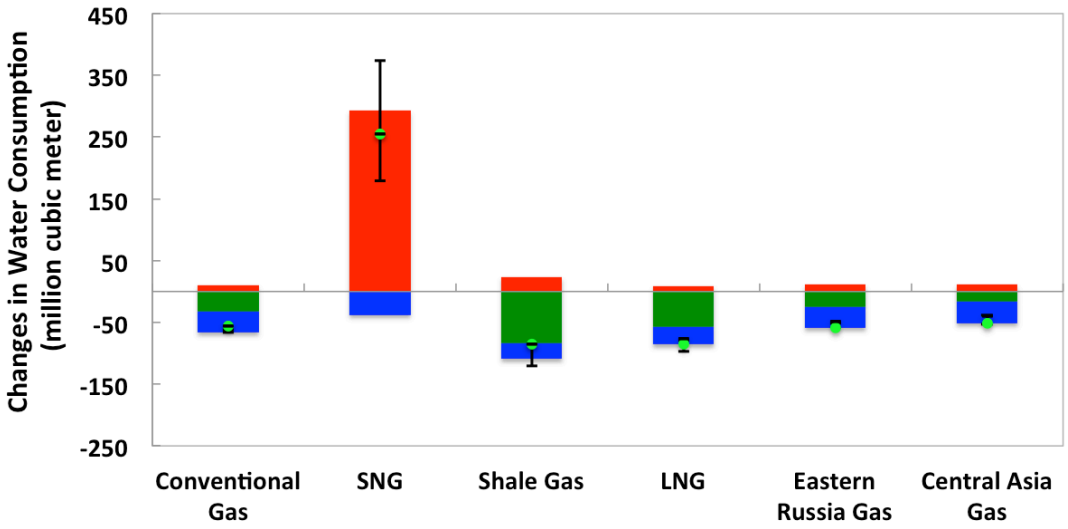


Fig. S2. Air quality-focused substitution (AS). Changes in stage-level water consumption for each gas source under the air quality focused coal-to-gas substitution scenario. China net values are obtained by considering water consumption changes occurring within China's border from upstream gas processes (upstream gas production, processing, transmission and distribution), upstream coal processes (upstream coal production, processing and transport) and end-use gas substitution for coal. Error bars represent water consumption changes resulting from mean, lower bound, and upper bound estimates of coal and each gas source occurring both within and outside of China.

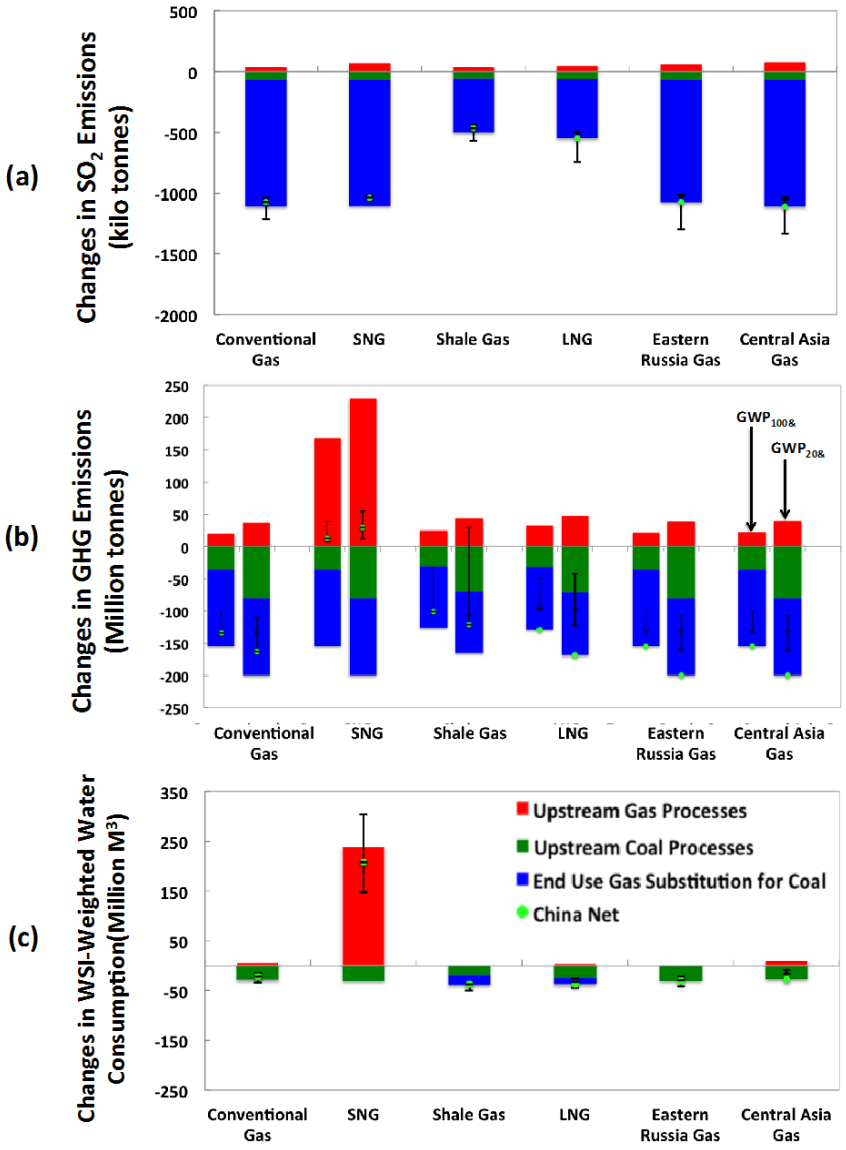


Fig. S3. Carbon-focused substitution (CS). Changes in upstream and downstream SO_2 emissions, greenhouse gas (CO_{2eq}, including both CO_2 and CH_4) emissions, and weighted water consumption for coal substitution by 30 bcm of gas from various sources. China net values are obtained by considering emission or weighted water consumption changes occurring within China's border from upstream gas processes (upstream gas production, processing, transmission and distribution), upstream coal processes (upstream coal production, processing and transport) and end-use gas substitution for coal. Error bars represent emission or weighted water consumption changes resulting from mean, lower bound, and upper bound estimates (mainly due to methane leakage rates) of coal and each gas source occurring both within and outside of China.

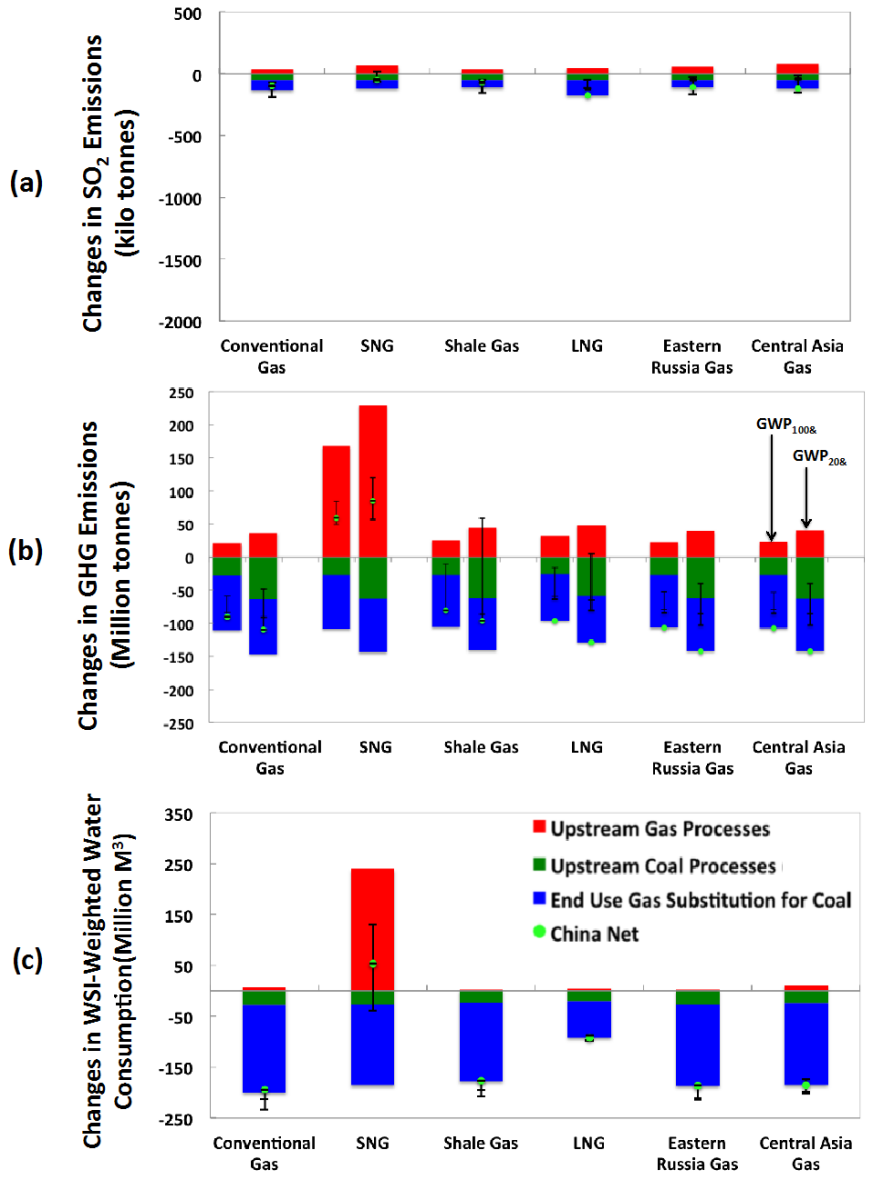


Fig. S4. Water-focused substitution (WS). Changes in upstream and downstream SO_2 emissions, greenhouse gas (CO_{2eq}, including both CO_2 and CH_4) emissions, and weighted water consumption for coal substitution by 30 bcm of gas from various sources. China net values are obtained by considering emission or weighted water consumption changes occurring within China's border from upstream gas processes (upstream gas production, processing, transmission and distribution), upstream coal processes (upstream coal production, processing and transport) and end-use gas substitution for coal. Error bars represent emission or weighted water consumption changes resulting from mean, lower bound, and upper bound estimates (mainly due to methane leakage rates) of coal and each gas source occurring both within and outside of China.

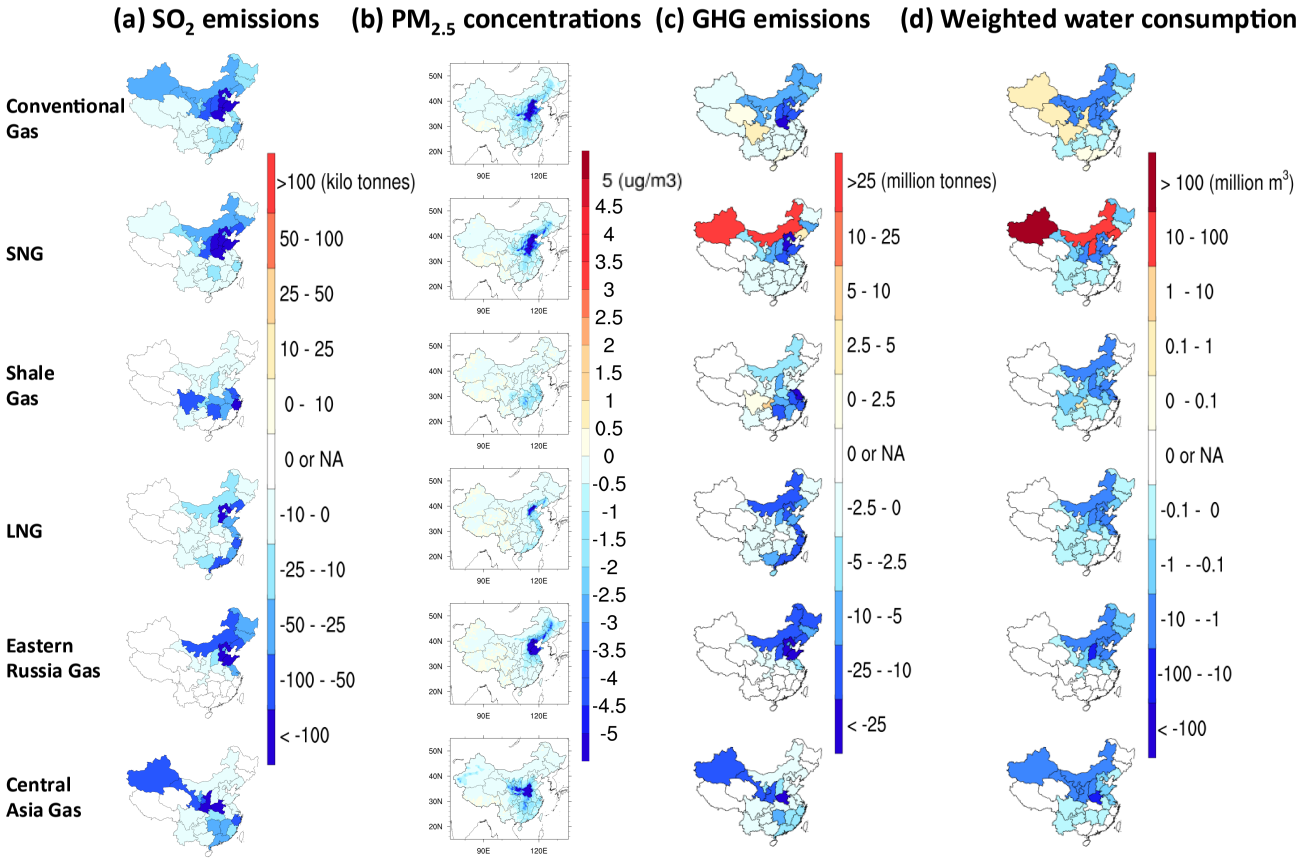


Fig. S5. Carbon-focused substitution (CS). Mainland China's spatially resolved changes in air (SO_2 emissions and population-weighted $PM_{2.5}$ surface concentrations), carbon (lifecycle greenhouse gas emissions under GWP_{20}, including CO_2 and CH_4, assuming mean methane leakage rates), and water (weighted water consumption) impacts of coal substitution by 30 bcm of gas from various sources. Changes in SO_2 emissions, GHG emissions, and weighted water consumption are shown at the provincial level; changes in $PM_{2.5}$ concentrations are shown at the grid level (27 km×27 km).

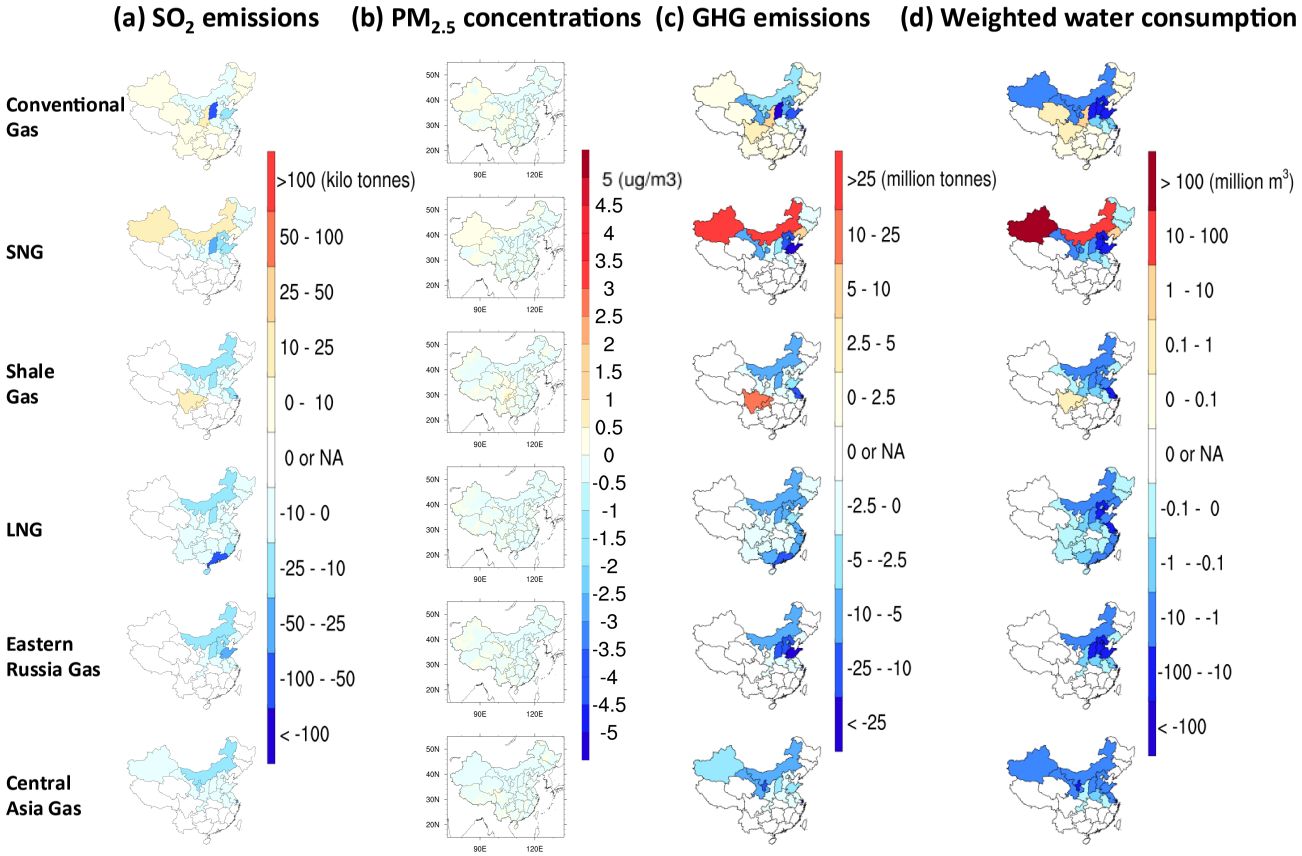


Fig. S6 Water-focused substitution (WS). Mainland China's spatially resolved changes in air (SO_2 emissions and population-weighted $PM_{2.5}$ surface concentrations), carbon (lifecycle greenhouse gas emissions under GWP_{20}, including CO_2 and CH_4, assuming mean methane leakage rates), and water (weighted water consumption) impacts of coal substitution by 30 bcm of gas from various sources. Changes in SO_2 emissions, GHG emissions, and weighted water consumption are shown at the provincial level; changes in $PM_{2.5}$ concentrations are shown at the grid level (27 km×27 km).

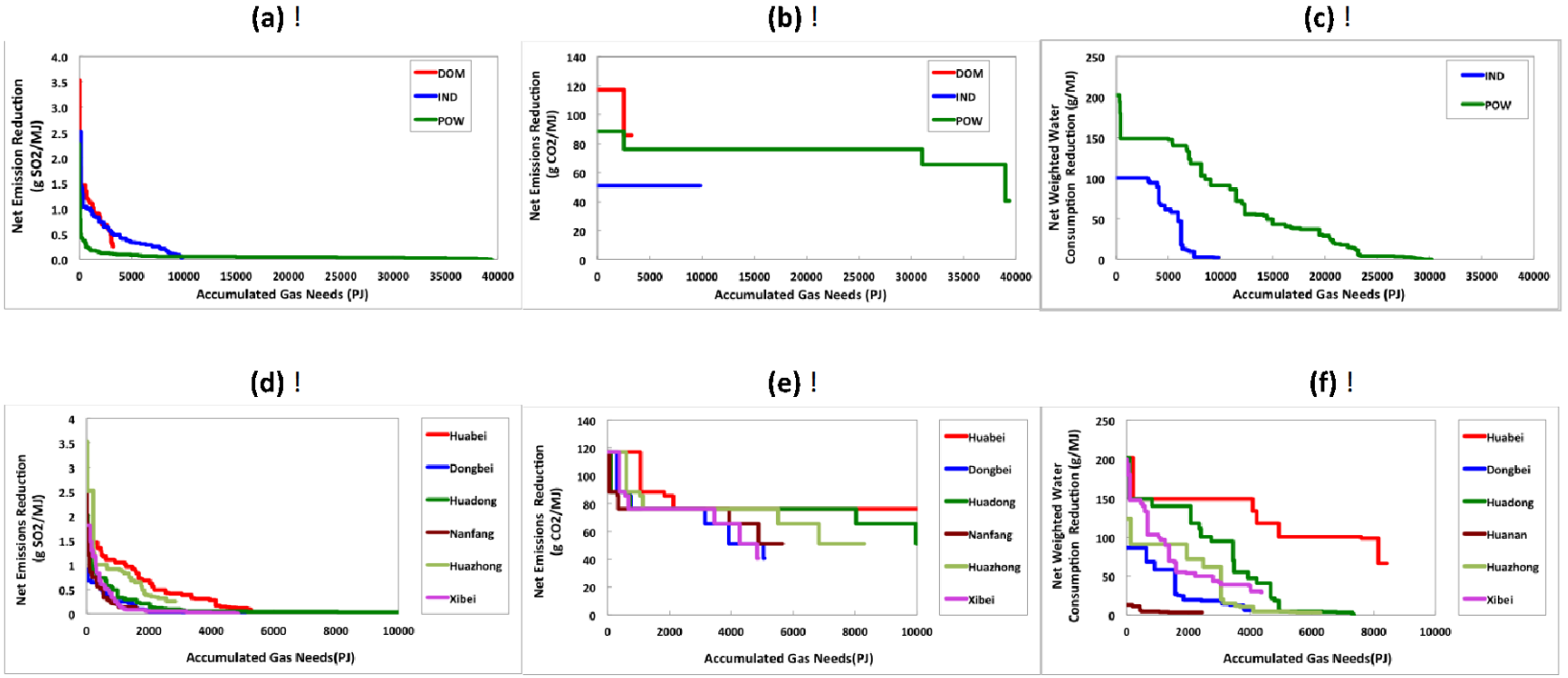
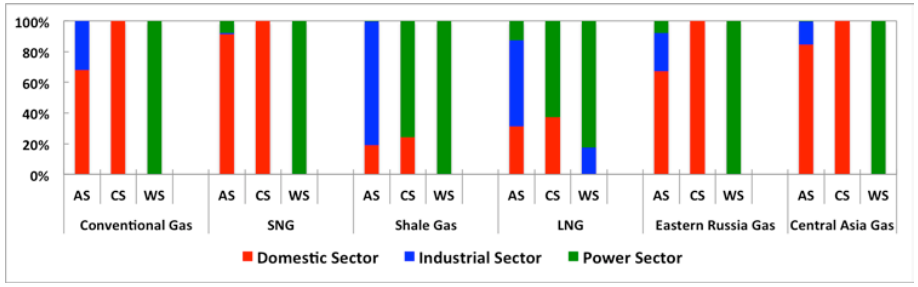
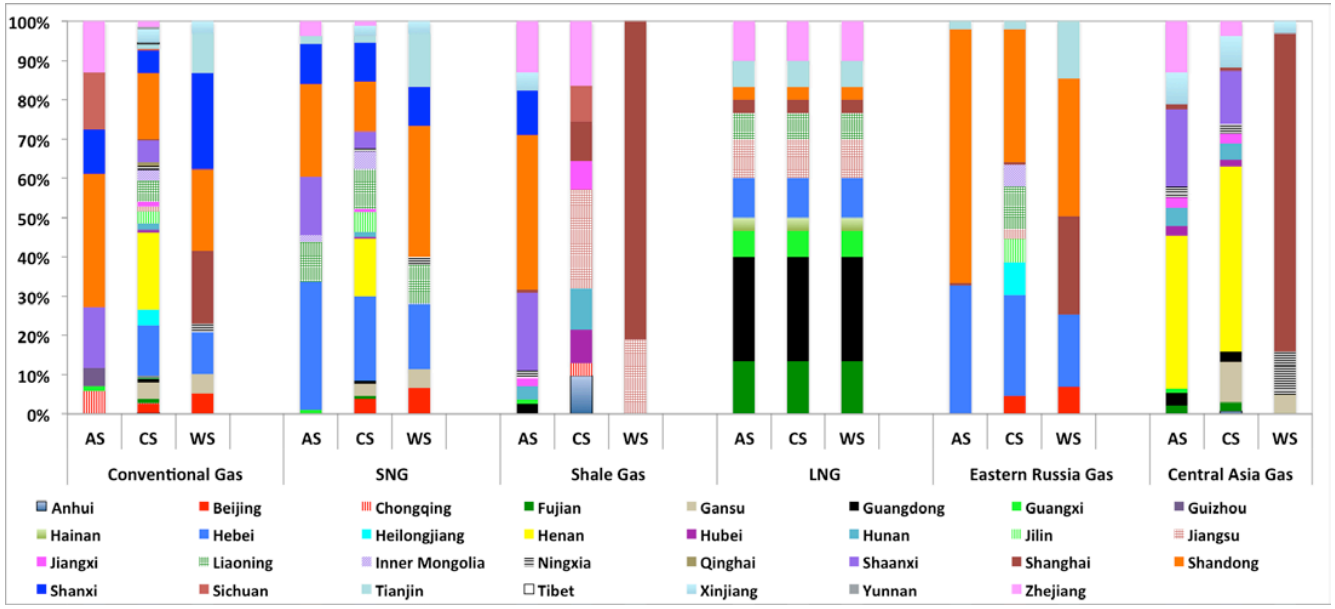


Fig. S7. Sectoral (a, b, c) and regional (d, e, f) marginal environmental benefits due to China's end-use coal-to-gas substitution in SO_2 emission reductions (a, d), CO_2 emission reductions (b, e), and weighted water consumption reductions (c, f) per MJ natural gas supply based on the Eclipse_V5a scenario. DOM, POW, and IND refers to the domestic, power, and industrial sectors, respectively, to represent sectoral differences. Huabei, Dongbei, Huadong, Nanfang, Huazhong, and Xibei represent China's six major power grids, respectively, to represent regional differences. Maginal benefits are sorted from highest to lowest for each sector or region and presented as total accumulated gas needs for coal substitution on the x axis.



(a)



(b)

Fig. S8. (a) Sectoral and (b) provincial gas allocation for each gas source under air quality focused substitution (AS), carbon focused substitution (CS), and water focused substitution (WS).

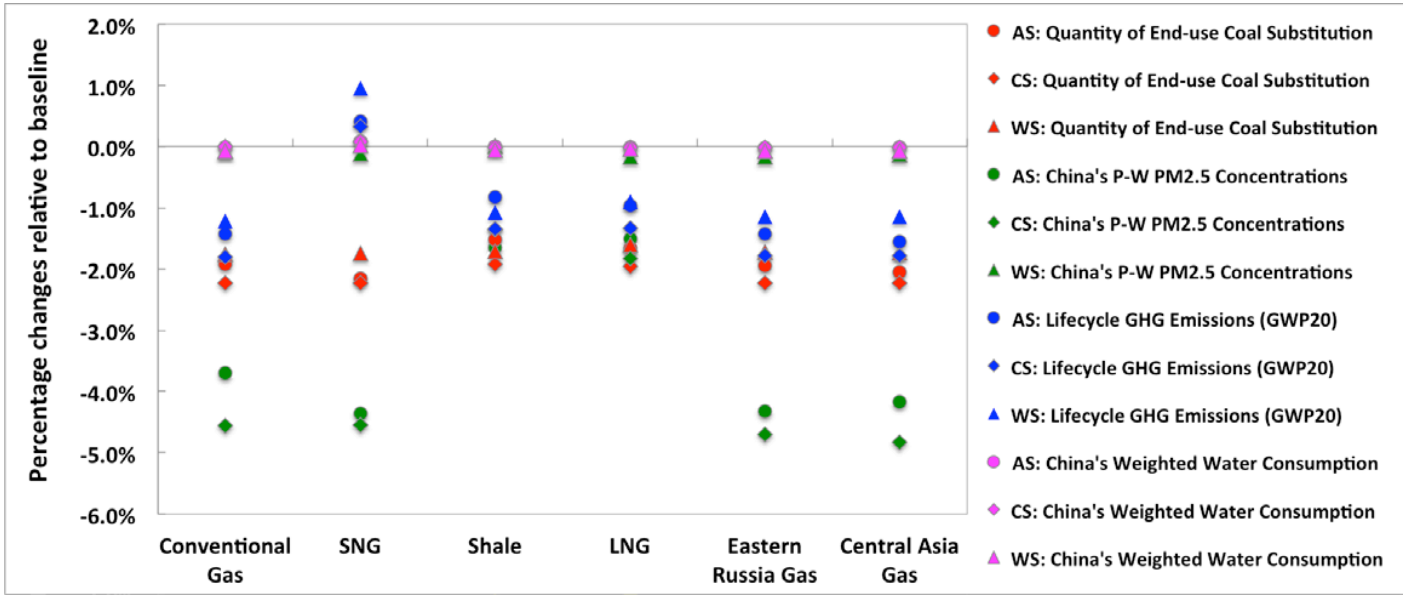


Fig. S9. Average percent changes in end-use coal consumption (quantity of end-use coal substitution) by natural gas, air quality (China's population-weighted $PM_{2.5}$ surface concentrations), carbon (lifecycle greenhouse gas emissions under GWP_{20}, assuming mean methane leakage rates), and water (China's weighted water consumption) impacts resulting from coal substitution by 30 bcm of natural gas from each source under three deployment strategies. AS: air quality-focused substitution, CS: carbon-focused substitution, WS: water-focused substitution. Baseline air quality, carbon and water impacts are represented by China's 2020 baseline population-weighted $PM_{2.5}$ surface concentrations obtained from our 2020 baseline WRF-Chem model simulations, predicted China's 2020 CO_2 emissions (~9000 million tonnes) reported in Guan et al. (2012)[49], and China's latest available national baseline weighted water consumption[10, 50]. As GHG emission changes are predominantly determined by changes in CO_2 emissions under a mean methane leakage, we have only slightly overestimated the percent changes in carbon impacts by using 2020 CO_2 emissions as the baseline.

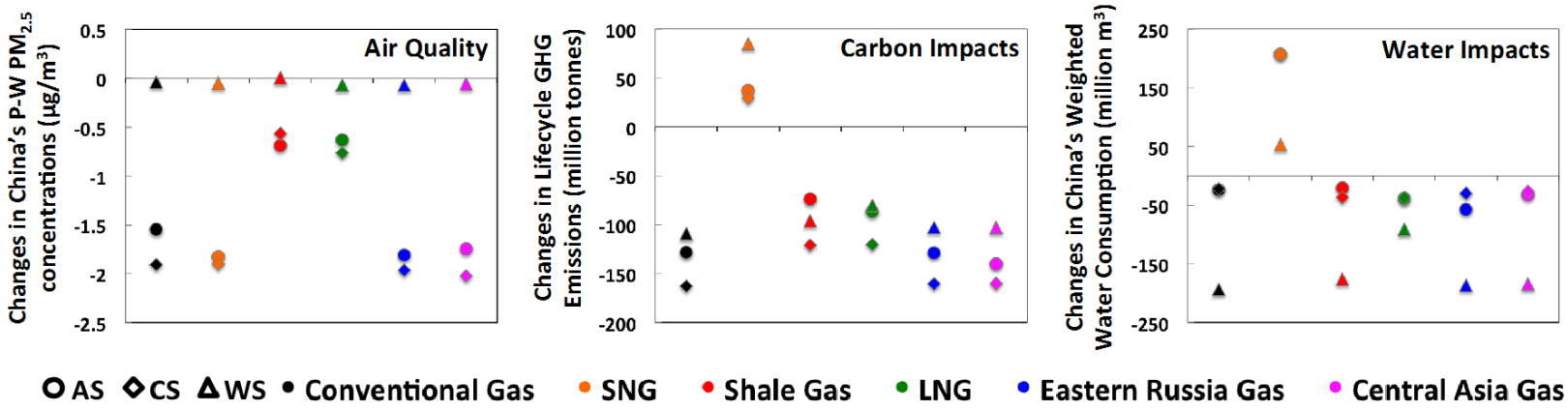


Fig. S10. Detailed breakdowns of comparison of net changes in air quality (China's population-weighted $PM_{2.5}$ surface concentrations), carbon (lifecycle greenhouse gas emissions under GWP_{20}, assuming mean methane leakage rates), and water impacts (China's weighted water consumption) of coal substitution by 30 bcm of gas from various sources under three deployment strategies. AS: air quality-focused substitution, CS: carbon-focused substitution, WS: water-focused substitution.

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