Air pollutant emissions from Chinese households: A major and underappreciated ambient pollution source

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As part of the 12th Five-Year Plan, the Chinese government has developed air pollution prevention and control plans for key regions with a focus on the power, transport, and industrial sectors. Here, we investigate the contribution of residential emissions to regional air pollution in highly polluted eastern China during the heating season, and find that dramatic improvements in air quality would also result from reduction in residential emissions. We use the Weather Research and Forecasting model coupled with Chemistry to evaluate potential residential emission controls in Beijing and in the Beijing, Tianjin, and Hebei (BTH) region. In January and February 2010, relative to the base case, eliminating residential emissions in Beijing reduced daily average surface $PM_{2.5}$ (particulate mater with aerodynamic diameter equal or smaller than 2.5 micrometer) concentrations by 14 ± 7 $\mu g \cdot m^{-3}$ (22 ± 6% of a baseline concentration of 67 ± 41 $\mu g \cdot m^{-3}$; mean ± SD). Eliminating residential emissions in the BTH region reduced concentrations by 28 ± 19 $\mu g \cdot m^{-3}$ (40 ± 9% of 67 ± 41 $\mu g \cdot m^{-3}$), 44 ± 27 $\mu g \cdot m^{-3}$ (43 ± 10% of 99 ± 54 $\mu g \cdot m^{-3}$), and 25 ± 14 $\mu g \cdot m^{-3}$ (35 ± 8% of 70 ± 35 $\mu g \cdot m^{-3}$) in Beijing, Tianjin, and Hebei provinces, respectively. Annually, elimination of residential sources in the BTH region reduced emissions of primary $PM_{2.5}$ by 32%, compared with 5%, 6%, and 58% achieved by eliminating emissions from the transportation, power, and industry sectors, respectively. We also find air quality in Beijing would benefit substantially from reductions in residential emissions from regional controls in Tianjin and Hebei, indicating the value of policies at the regional level.

$PM_{2.5}$ | secondary aerosols | regional pollution transport | residential emissions | source contribution

Over the past 30 years, China has experienced rapid economic growth, accompanied by accelerating urbanization, which has increased consumption of fossil fuels and worsened air quality. Although considerable efforts have been made to control air pollution, the focus has largely been on the power, transport, and, to a lesser extent, industry sectors, and reduction per unit activity has been offset by economic growth and increasing fossil fuel use (1). An air pollution control approach that prioritizes reductions from sources that create the highest pollutant exposures would be more effective in reducing the health impacts of air pollution. As the largest coal consumer, the power sector receives priority in efforts to reduce air pollutant emissions, and has significantly reduced emissions of sulfur dioxide (SO_2) and particulate matter (PM) in recent years (2). Industry and transportation emissions have also received attention (3), but the contribution of residential emissions to ambient air pollution has been relatively neglected. The residential sector is the largest emitter of carbonaceous aerosols (4, 5), which are formed by the inefficient combustion of fossil fuel and biomass in unregulated cooking and heating devices. Household combustion of coal also emits SO_2, a precursor to secondary $PM_{2.5}$ (particulate matter with aerodynamic diameter equal or smaller than 2.5 micrometer). In 2010, the residential sector accounted for around 18% of total energy consumption in China, but contributed 10%, 50%, and 69% of anthropogenic SO_2, black carbon (BC), and organic carbon (OC) emissions, respectively (5).

Although not the focus of this paper, use of solid fuels (coal and biomass) for heating and cooking in households contributes directly to exposures in and around residences and is a major source of ill health in China. The Global Burden of Disease study found that direct household exposure to air pollution from solid fuels was responsible for ~0.8 million premature deaths in China in 2013, about equal to the number of premature deaths from ambient particle pollution. Together, they make up the second largest risk factor in the country, ranked between high blood pressure and smoking (6–8). In addition to exposure within households, these emissions contribute to ambient air pollution, and thus affect populations over wide areas. To achieve the National Air Pollution Prevention and Control Action Plan (2013–2017) targets (hereafter the "Action Plan") efficiently, regional data are needed to prioritize modifications to the structure of the energy sector to reduce health-damaging emissions from all sectors, including households. There have been estimates of the contribution of household emissions to ambient pollution in China based on global databases and models (9, 10). These analyses use coarse resolution models and have not been

Significance

China suffers from severe outdoor air pollution and associated public health impacts. In response, the government has imposed restrictions on major pollution sources such as vehicles and power plants. We show that due to uncontrolled and inefficient combustion of solid fuels in household devices, emission reductions from the residential sector may have greater air quality benefits in the North China Plain, including Beijing than reductions from other sectors. These benefits would be largest in the winter heating season when severe air pollution occurs. Household emissions, mostly from space heating and cooking with solid fuels, are an important and generally unrecognized source of ambient air pollution in China and other developing countries. Alternative fuels and other ways of reducing emissions would have large benefits.

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informed by local measurements, and are thus inadequate by themselves to guide local actions.

Here, we use the Weather Research and Forecasting model with Chemistry (WRF-Chem) (11) to analyze the benefits of two residential emission mitigation scenarios during the heating season on $PM_{2.5}$ concentrations in Beijing and in the Beijing, Tianjin, and Hebei (BTH) region of northern China. In 2010, the population of this region was ∼104 million people, representing about 15% of the national population living in areas with significant household space-heating needs in winter (mid-November to mid-March). These needs are largely met using coal in simple devices with high emission factors in many households. This study provides a basis for further discussion of alternative emission control strategies across energy demand sectors in China.

Study Region and Scenarios

The study region is the BTH region in the North China Plain (Fig. S1). Beijing is China's capital, and the city of Tianjin is adjacent to Beijing. Hebei is the province surrounding the two megacities. In 2010, the urban population in Beijing, Tianjin, and Hebei was 86%, 80%, and 45% of the total population in each of the three provinces (12), respectively. The BTH region occupies only 2.3% of the total national land area; however, in 2010, it had 8%, 11%, and 12% of the national population, gross domestic product, and energy consumption, respectively (12).

To examine the contribution of residential emissions to regional air pollution, we designed three scenarios. BASE is the baseline scenario in which a WRF-Chem simulation used the Multiresolution Emission Inventory of China (MEIC; www.meicmodel.org) emission inventory for January and February 2010. Residential emissions were removed in the Beijing (BJR) scenario and in the Beijing, Tianjin, and Hebei (BTHR) regional scenario, respectively. The difference between the WRF-Chem BASE and BJR or BTHR scenario simulations provides an estimate of the total contribution of the residential sector from each region to regional outdoor air pollution. In addition, to simulate potential mitigation strategies more realistically, we conducted sensitivity simulations in which residential emissions were reduced by 25%, 50%, and 75% of the BASE emissions in the BTH region.

Contributions of Emissions

Table S1 summarizes coal and biomass combustion by sector in the BTH region. In all three provinces, power plants are the largest coal consumer, followed by industry, whereas the residential sector uses the least amount of solid fuel (including biomass). The picture is different, however, when sectoral contributions to emissions of various air pollutants are compared, with a consistently large proportion of aerosol species found to originate from the residential sector. Fig. 1 shows the relative contributions of the transportation, power, industry, and residential sectors to $PM_{2.5}$, BC, OC, SO_2, and nitrogen oxides ($NO_x = NO + NO_2$) emissions at the provincial scale from the MEIC. In 2010, the primary $PM_{2.5}$, BC, OC, SO_2, and NO_x emissions in the BTH region were 1,100 kilotons (kt), 170 kt, 272 kt, 2,010 kt, and 2,830 kt, respectively. The residential sector accounted for 32%, 44%, 71%, 15%, and 4%, respectively, of the total emissions of each pollutant. During January and February, more fuel is burned for heating, and the contribution of the residential sector to total emissions is greater. Fig. S2 presents the spatial distribution of residential emissions and their share of the total anthropogenic emissions in January and February of 2010 in eastern China. The highest emissions are distributed in the east, covering the southeast of the BTH area, Shandong Province, and the north of Henan Province. Emission "hot spots" are located over most cities. The residential sector contributes more than 50% of the emissions of $PM_{2.5}$, BC, and OC in northern China. For OC, the residential sector contribution can exceed 95%.

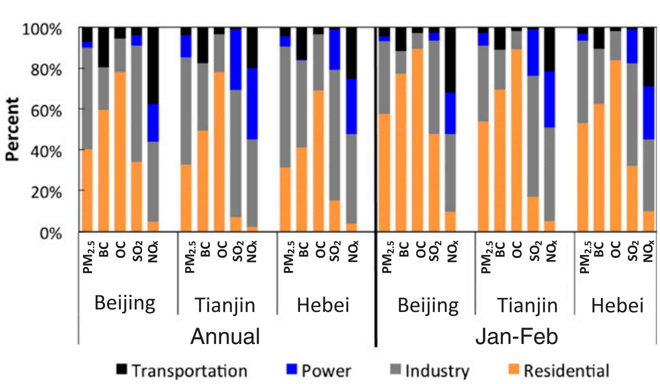

Fig. 1. Relative contributions of the transport, power, industry, and residential sectors to $PM_{2.5}$, BC, OC, SO_2, and NO_x emissions in Beijing, Tianjin, and Hebei in 2010 and in January and February of 2010 alone.

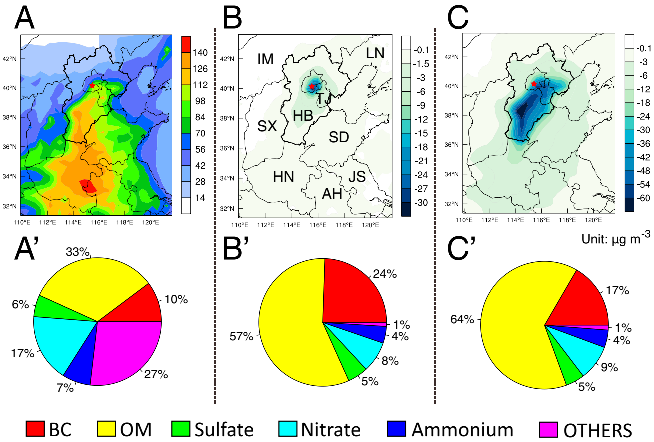

Fig. 2. Scenario outcomes for January 1–February 28, 2010. The red star indicates the Beijing city center. (*A*) BASE scenario distribution of mean $PM_{2.5}$ concentrations. (*A′*) Chemical composition of $PM_{2.5}$ in the BTH region in the BASE scenario. (*B*) Mean $PM_{2.5}$ concentration decrease in the BJR scenario (BJR minus BASE). (*B′*) Chemical composition of the eliminated $PM_{2.5}$ over the BTH region in the BJR scenario (BJR minus BASE). (*C*) Mean $PM_{2.5}$ concentration decreases in the BTHR scenario (BTHR minus BASE). (*C′*) Chemical composition of the eliminated $PM_{2.5}$ over the BTH region in the BTHR scenario (BTHR minus BASE). The region surrounded by the thicker line in *A*–*C* is the BTH region. The provinces in eastern China are marked in *B*. AH, Anhui; HB, Hebei; HN, Henan; IM, Inner Mongolia; JS, Jiangsu; LN, Liaoning; SD, Shandong; SX, Shanxi; TJ, Tianjin.

Results

Air Quality Improvements in the BTH Area. Before the scenario analysis, meteorological fields (including hourly surface air temperature, relative humidity, wind speed, wind direction, and daily precipitation), $PM_{2.5}$ mass concentration, and the $PM_{2.5}$ chemical composition in the model were evaluated (Tables S2–S4). Details are provided in *SI Materials and Methods*. Fig. 2*A* shows the distribution of mean $PM_{2.5}$ surface concentrations over eastern China from January 1 to February 28, 2010 in the BASE simulation. The top of the surface layer is ∼26 m. In the BTH region, high $PM_{2.5}$ concentrations were distributed in the southeast, along the Yan and Taihang Mountains, from the north to south. This spatial distribution has a similar pattern to the pattern of primary emissions (Fig. S2), except along the coast, where the sea-land breeze transports and dilutes the pollutants. The corresponding chemical composition of $PM_{2.5}$ in the BTH region is presented in Fig. 2*A′*. On average, BC, organic matter (OM; OM/OC = 1.5), sulfate, nitrate, and ammonium account for 10%, 33%, 6%, 17%, and 7% of the $PM_{2.5}$ mass concentration in the region, respectively. The OM fraction is dominant, and sulfate, nitrate, and ammonium are also important, accounting for 30% of the

$PM_{2.5}$ mass concentration. As shown in Fig. 1, during January and February 2010, the residential sector contributed 65% and 85% of the BC and OC emissions in the BTH region, and consequently was responsible for most of the BC and OM in the $PM_{2.5}$ in the BTH region.

Fig. 2*B* shows the decreases in the two-month $PM_{2.5}$ average mass concentration over eastern China with no emissions from the residential sector in Beijing. Reductions in $PM_{2.5}$ concentrations are centered over southeast Beijing, with the largest reduction of over 30 $\mu g{\cdot}m^{-3}$ found in the south. In addition to reductions in Beijing, $PM_{2.5}$ in the surrounding areas of Hebei and Tianjin decrease by 3–6 $\mu g{\cdot}m^{-3}$ due to regional transport of air pollutants. The chemical composition of the eliminated $PM_{2.5}$ concentrations over the BTH region is presented in Fig. 2*B*′; the contributions of BC, OM, sulfate, nitrate, and ammonium are 24%, 57%, 5%, 8%, and 4%, respectively. Carbonaceous particles are the major component of the reduced $PM_{2.5}$ mass concentration (81%), whereas the remaining 17% reduced is composed of secondary inorganic particles.

Fig. 2*C* depicts the changes in average $PM_{2.5}$ concentration between the BTHR and BASE scenarios over eastern China. Setting residential emissions to zero simultaneously in the Beijing, Tianjin, and Hebei provinces greatly expanded $PM_{2.5}$ reductions over the entire BTH region with reductions extending over the surrounding provinces with substantially larger reductions than in the BJR scenario. The changes in concentration located along the Yan and Taihang Mountains, divide the BTH region into roughly two parts, northwest and southeast. The largest decrease in $PM_{2.5}$ occurs over southern Hebei, along a line following a series of large cities in the province [i.e., Langfang, Baoding, Shijiazhuang, Xingtai, Handan (from north to south)] where residential emissions are the largest. In Shijiazhuang, the capital of Hebei Province, the average $PM_{2.5}$ concentration fell by over 60 $\mu g{\cdot}m^{-3}$. The BTH region is 0.21 million km^2, and the mean decrease across the regions over the January–February period varied from 7 to 52% depending on location and by 36% on average. Although no emissions restrictions were implemented in other neighboring provinces, air quality also improved to the south and east of the BTH region, and the $PM_{2.5}$ reductions in Shandong Province and Henan Province ranged from 2–29 $\mu g{\cdot}m^{-3}$ and 2–35 $\mu g{\cdot}m^{-3}$ depending on location and by 7 $\mu g{\cdot}m^{-3}$ and 8 $\mu g{\cdot}m^{-3}$

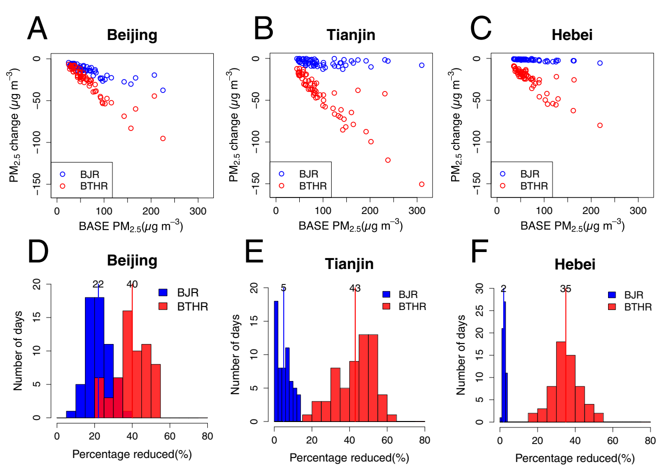

Fig. 3. Decrease in daily mean surface $PM_{2.5}$ concentration in each province as a result of elimination of residential emissions. Scatter plots of decreases in daily $PM_{2.5}$ concentrations in the BJR (blue) and BTHR (red) scenarios relative to daily $PM_{2.5}$ concentrations in the BASE scenario in Beijing (*A*), Tianjin (*B*), and Hebei (*C*). Frequency histograms of percent $PM_{2.5}$ reduction in Beijing (*D*), Tianjin (*E*), and Hebei (*F*) in BJR (blue) and BTHR (red) scenarios. The darker red color in *D* indicates the overlapping region of color bars in the BJR and BTHR scenarios. Vertical blue (red) lines and associated values indicate the mean percent reduction for the BJR (BTHR) scenario.

Table 1. Average $PM_{2.5}$ concentrations in the BASE, BJR, and BTHR scenario simulations, and concentration decreases and percent reductions in the BJR and BTHR scenarios from elimination of residential emissions

	Concentration,* $\mu g{\cdot}m^{-3}$			Concentration decrease,* $\mu g{\cdot}m^{-3}$		Percent reduction,* %	
Region	BASE	BJR	BTHR	BJR	BTHR	BJR	BTHR
Beijing	67 ± 41	53 ± 36	40 ± 26	14 ± 7	28 ± 19	22 ± 6	40 ± 9
Tianjin	99 ± 54	94 ± 53	55 ± 32	5 ± 3	44 ± 27	5 ± 4	43 ± 10
Hebei	70 ± 35	69 ± 34	45 ± 24	2 ± 1	25 ± 14	2 ± 1	35 ± 8
BTH	72 ± 36	69 ± 35	46 ± 24	3 ± 1	26 ± 15	4 ± 1	36 ± 7

Results show mean and SD of daily average values, and changes in those values relative to the BASE case, for each region for January–February 2010.
*Results are presented using area-based concentrations; to estimate health effects, population-weighted concentrations are needed to estimate population exposures.

on average, respectively. Similar to the BJR scenario, the dominant components of the eliminated $PM_{2.5}$ were OM and BC, accounting for 64% and 17% of the total mass, respectively (Fig. 2*C*′). However, in the BTHR scenario, the contribution of OM to the eliminated $PM_{2.5}$ was larger and the contribution of BC to the eliminated $PM_{2.5}$ was smaller than in the BJR scenario. The higher contribution of OM in the BTHR scenario is because biomass constituted a larger share of the residential energy structure in Hebei and Tianjin than in Beijing (compare Table S1) and OC is mainly from residential biomass use (4).

Air Quality Improvements at the Provincial Level. To evaluate daily air quality improvements at the provincial level, we derive the area-weighted daily average surface $PM_{2.5}$ concentrations over the BTH region. Fig. 3 shows the decrease in daily area-based average $PM_{2.5}$ concentrations for each of the three provinces. For each province, the mean and SD of $PM_{2.5}$ concentration, concentration decrease from the BASE simulation, and percentage of $PM_{2.5}$ reduced are summarized in Table 1. Details of the provincial calculation in Fig. 3 and Table 1 are provided in *SI Materials and Methods*.

The absolute reduction in $PM_{2.5}$ concentrations was positively and strongly correlated with the BASE $PM_{2.5}$ concentration in both the BJR and BTHR scenarios. Fig. 3 shows that on more polluted days, the decrease in $PM_{2.5}$ concentration is generally larger, indicating that the emission control measures in the residential sector decreased $PM_{2.5}$ concentrations more when air pollution concentrations were higher. During the heavy polluted periods in winter, the North China Plain is often dominated by a weak high-pressure system with low surface winds (13), which leads to weak mixing and diffusion; hence, emission reductions during those periods are particularly beneficial to local regions. In the BJR scenario, the average $PM_{2.5}$ concentration in Beijing decreased by 14 ± 7 $\mu g{\cdot}m^{-3}$ (mean and SD of 59-d daily average values) and the BASE $PM_{2.5}$ concentration decreased by 22 ± 6%. Details on how these values are obtained are included in *SI Materials and Methods*. At the same time, the average $PM_{2.5}$ concentration in Tianjin and Hebei also decreased, but the decreases were less than 5%. Because the emission reductions in Beijing are relatively small compared with the emissions from Tianjin and Hebei, emission reductions in Beijing do not help much to improve the air quality in Beijing's surrounding provinces. In the BTHR scenario, on the other hand, the residential emission elimination strategies in the BTH region resulted in decreases in $PM_{2.5}$ concentration of 28 ± 19 $\mu g{\cdot}m^{-3}$, 44 ± 27 $\mu g{\cdot}m^{-3}$, and 25 ± 14 $\mu g{\cdot}m^{-3}$ on average for Beijing, Tianjin, and Hebei, respectively, which were 40 ± 9%, 43 ± 10%, and 35 ± 8%, respectively, of the BASE $PM_{2.5}$ concentration. Thus, reductions of residential emissions in regions surrounding Beijing would also substantially improve Beijing's air quality as well as reduce pollutant contributions in downwind regions.

Previous studies have shown that regional transport is an important source of air pollution in Beijing (14, 15). When the prevailing wind is southerly, air pollutants from Hebei, Shandong, and Henan are transported to Beijing (15, 16), and the contribution of emissions from surrounding regions to $PM_{2.5}$ in Beijing has been found to be 34–39% (14, 15). Our study also finds that regional air quality management is critical. Although air pollution in Beijing receives considerable attention, we found $PM_{2.5}$ concentrations to be substantially higher south of the BTH region (Fig. 2*A*). In our study, the elimination of residential emissions in Beijing alone decreased $PM_{2.5}$ concentrations in the city by 14 ± 7 $\mu g{\cdot}m^{-3}$ ($22 \pm 6\%$), whereas the elimination of residential emissions in the BTH region decreased $PM_{2.5}$ concentrations in Beijing nearly twice as much, by 28 ± 19 $\mu g{\cdot}m^{-3}$ ($40 \pm 9\%$) (Table 1 and Fig. 3*A*), as well as reducing $PM_{2.5}$ concentrations in Tianjin and Hebei by $43 \pm 10\%$ and $35 \pm 8\%$, respectively.

Fig. 3 *D–F* presents the frequency histograms of the percent decrease in daily $PM_{2.5}$ concentrations in the BJR and BTHR scenarios for the 59 simulation days in January–February 2010. The average daily $PM_{2.5}$ decrease was 22%, 5%, and 2% in the BJR scenario and 40%, 43%, and 35% in the BTHR scenario in the Beijing, Tianjin, and Hebei Provinces, respectively. Although emission control measures were implemented in the model on all days, the $PM_{2.5}$ percent decreases varied significantly (e.g., in Beijing, the $PM_{2.5}$ percent decrease varied from 21 to 55% in the BTHR scenario) due to meteorological conditions. Compared with the relatively clean periods, the polluted periods were generally associated with lower boundary layer height and wind speed and higher temperature and relative humidity (Fig. S3). These conditions lead to high $PM_{2.5}$ concentrations due to weak mixing and diffusion. As a result, percent decreases in $PM_{2.5}$ concentration due to emission reductions from local sources are generally larger during polluted periods than during clean periods.

To inform policy initiatives, we also conducted simulations in which we reduced residential emissions by 25%, 50%, and 75% in the BTH region, The results show that the air quality benefits of reducing residential emissions in the BTH region during the heating season are approximately linear, indicating that policies to reduce emissions from the residential sector will likely lead approximately linearly to reductions in ambient $PM_{2.5}$ concentrations.

Discussion

During January and February 2010, in the BTH region, the residential sector contributed 53%, 65%, 85%, 32%, and 9% of primary $PM_{2.5}$, BC, OC, SO_2, and NO_x emissions, respectively. The WRF-Chem simulations indicate that during the residential heating season, the elimination of residential emissions in Beijing alone would decrease surface $PM_{2.5}$ concentrations by $22 \pm 6\%$ in Beijing and the elimination of residential emissions in the BTH region would decrease surface $PM_{2.5}$ concentrations by $36 \pm 7\%$ in the BTH region. Compared with the power and industrial sectors, although the residential sector consumed less solid fuel (Table S1), it made larger contributions to emissions of primary particles in winter (Fig. 1), owing primarily to the low combustion and thermal efficiencies of cooking and heating stoves and absence of any end-of-pipe controls.

In China, residential solid fuel combustion results in large emissions of PM, although the emission factor varies with fuel type, fuel properties, and burning conditions. Zhang et al. (17) reported mean total suspended particulate emission factors of 8.05, 3.82, and 1.30 $g{\cdot}kg^{-1}$ for crop residues, wood, and coal, respectively, burned in various stoves. Studies in China found that burning bituminous coal briquettes led to a higher PM emission factor than burning of anthracite briquettes, and burning bituminous coal chunks has an even higher emission factor (18–20). Of these emissions, more than 94% of the PM is below 0.95 μm in diameter, whereas only about 1% is above 7.2 μm in diameter (18), indicating the dominance of fine particulates ($PM_{2.5}$) in residential emissions. Zhi et al. (20) reported that emission factors (EFs) of PM ($PM_{2.5}$ dominant), OC, and elemental carbon (EC) are 7.33, 4.16, and 0.08 $g{\cdot}kg^{-1}$ and 14.8, 5.93, and 3.81 $g{\cdot}kg^{-1}$ for bituminous coal briquettes and chunks, respectively, and that they are 1.21, 0.06, and 0.004 $g{\cdot}kg^{-1}$ and 1.08, 0.10, and 0.007 $g{\cdot}kg^{-1}$ for anthracite briquettes and chunks, respectively. Anthracite burns more cleanly and emits less PM and volatile organic compounds (VOCs) than bituminous coal, but is more expensive and harder to light and poses hazards from carbon monoxide poisoning. In comparison, the national average $PM_{2.5}$ emission factor in coal-fired power plants was estimated to be 0.53 $g{\cdot}kg^{-1}$ in 2010 (2). This emission factor is substantially less than 10% of the $PM_{2.5}$ emission factor for the residential bituminous coal combustion process (18–20).

Field studies have observed comparable annual mean PM_{10} (particulate matter with aerodynamic diameter equal or smaller than 10 micrometer) concentrations in urban areas (180 ± 171 $\mu g{\cdot}m^{-3}$) and rural villages (182 ± 154 $\mu g{\cdot}m^{-3}$) at 18 sites across northern China, suggesting that the severe outdoor air pollution in rural areas is partially derived from household solid fuel combustion (21). In 2013, the State Council issued the Action Plan, under which the BTH region is required to achieve a 25% reduction in annual mean $PM_{2.5}$ concentrations from the 2012 level by 2017. Strategies focusing on emission reductions and changes in energy systems in the power, industry, and transportation sectors have been given considerable attention (3), but air quality would benefit from greater attention on the residential sector.

There are clear opportunities to reduce ambient $PM_{2.5}$ concentrations and potentially achieve climate co-benefits via mitigation efforts in households. With significant pollutant emissions, residential sources are close to dwellings and have near-ground emissions that have a greater impact on surface air pollution levels and result in higher human exposure than is typical for power or industrial sources (22) [i.e., the intake fraction is much higher (23)]. Solid fuel (including biomass and coal) used for household heating and cooking emits air pollutants, short-lived greenhouse pollutants like BC, and a range of greenhouse gases. Cleaner stoves, such as advanced fan-stoves using pelletized biomass, and intrinsically clean energies at end use, such as natural gas, liquefied petroleum gas (LPG), and electricity, are potential mitigation strategies in the residential sector. Truly clean-burning coal stoves could have direct indoor and outdoor air quality and human health benefits, but not help significantly in climate mitigation. On the other hand, clean energies with lower climate footprints can be used as interim steps (e.g., LPG) while moving to long-term solutions (natural gas, biogas, electricity, and wind and solar energy), which can completely replace solid fuel. In the urban and suburban areas around Beijing, replacing household coal with natural gas has already been implemented, and with increasing import from Russia and development of shale gas reserves in China, there is potential to expand the use of natural gas to many cities and even to large preurban areas around the country. Care will need to be taken to avoid leakage of methane, the primary component of natural gas and a strong greenhouse gas. For households in remote regions, LPG, biogas, and electricity generated with wind or solar power are longer term low-emission options. For meeting space-heating needs, to be efficient, these clean fuels need to be accompanied by improved heat retention in households: better insulation and reduced leakage.

A number of epidemiological studies have addressed the health effects of household solid fuel use for heating and cooking due to exposures in the household environment (6, 24, 25). In addition to helping address the problem of household air pollution, substitution of solid fuels with low-emission energy sources in the residential sector can improve widespread outdoor air quality. The climate benefit of using natural gas and electricity, however, depends on the source of power production and what they are displacing, whereas the use of biogas (with care to prevent leakage) and wind and solar energy can be expected to bring significant climate co-benefits in nearly all situations. Widespread adoption of these residential mitigation strategies will substantially help meet the ambient $PM_{2.5}$ targets in the Action Plan and provide large human health benefits via reductions in both local household and regional outdoor exposure to $PM_{2.5}$.

The year 2010 was chosen for this study because a detailed emission inventory is available. We compared the average planetary boundary layer (PBL) height of January and February from 2010 to 2014 using meteorological data derived from the National Centers for Environmental Prediction Final Analysis, and found the average boundary layer height over the BTH region in January and February of 2010 to be the highest of the 5-y period. Our study shows that even with meteorological conditions such as these, which are relatively favorable for pollutant dilution via mixing, the elimination of residential emissions in the BTH region was highly effective at reducing surface $PM_{2.5}$ concentrations. In periods of more stable synoptic conditions (e.g., winter 2013) with lower surface wind speed and PBL height (13), residential emissions likely made an even larger contribution to haze formation. Additional analysis is needed to characterize better the role of residential emissions in severe air pollution episodes during these periods.

Our analysis examines the heating season, when larger quantities of coal and biomass are burned and the contributions of the residential sector to total emissions are larger than at other times of year. In contrast, the relative contribution of secondary inorganic aerosols is larger in summer, when high temperatures and humidity and strong atmospheric oxidation favor secondary aerosol formation (26). More research is needed to evaluate the contribution of emissions from the residential sector on air pollution during each season.

The WRF-Chem simulation introduced uncertainties into the results. As discussed in *SI Materials and Methods* (Fig. S4 and Tables S3 and S4), although the mass concentration and daily trends were captured well by the WRF-Chem model, the simulated $PM_{2.5}$ species differed from observations. In particular, BC is overestimated and sulfate is underestimated. By comparing our data with field data from recent publications (*SI Materials and Methods*), we found BC was overestimated by 36–149% at various sites, which was also found in a study using Community Multi-Scale Air Quality (CMAQ) Model (27), and the difference may be due to uncertainties in emissions from coal boilers and stoves, as well as diesel trucks in the MEIC emission inventory. During January and February 2010, the residential sector dominated BC emissions, accounting for 77%, 70%, and 62% of the total emissions in Beijing, Tianjin, and Hebei Provinces, respectively. The overestimation of BC would result in overestimation of the contributions of residential emissions to $PM_{2.5}$ concentrations. In contrast, sulfate was underestimated by 39–90% in the present study, and a possible reason might be the missing pathways of sulfate enhancement by mineral aerosols in the WRF-Chem model, including aqueous oxidation, catalyzed oxidation, and SO_2 heterogeneous reactions, which are estimated to contribute 40% of the total sulfate production during winter (28).

WRF-Chem model results vary with horizontal resolution; hence, model resolution is an additional source of uncertainty. In this study, the WRF-Chem domain covers mainland China, with a horizontal resolution of 36 km. This resolution is the same as used in several recent model studies in northern China and the BTH region (27, 29). A number of studies apply nested simulations with a horizontal resolution in the innermost domain of 12 km (30) or 9 km (31). Wang et al. (30) compared domain-wide $PM_{2.5}$ predictions at 36-km and 12-km grid resolutions, and found that the use of a finer grid changed $PM_{2.5}$ performance from a slight underprediction to a moderate overprediction. In our study, the 36-km resolution achieved better model performance than the 12-km resolution.

Conclusions

Due to rapid economic development and high levels of solid fuel combustion, China is facing severe air pollution problems. To achieve targets in the Action Plan efficiently, it is critical to prioritize the reduction and replacement of high-emitting end-use energy combustion processes with clean energy across a variety of sectors. Residential emissions from direct combustion of solid fuel in low-efficiency stoves contribute substantially to regional $PM_{2.5}$ loads. Reduction of emissions from the residential sector via the replacement of solid fuels with other cleaner energy sources could substantially improve air quality in the BTH region of eastern China.

On an annual emissions basis, elimination of residential sources in the BTH region would reduce emissions of primary $PM_{2.5}$ and SO_2 by about 32% and 15%, respectively, compared with 5%, 6%, and 58% of primary $PM_{2.5}$ and 1%, 20%, and 63% of SO_2 by eliminating emissions from the transportation, power, and industry sectors, respectively. Indeed, residential sources contribute far more to primary $PM_{2.5}$ emissions annually in Beijing and the surrounding region than the transportation and power sectors combined and, in winter, more than industry.

In the present study, we estimate the implications of reducing residential emissions for ambient concentrations in the BJR scenario and in the BTHR scenario. Eliminating residential emissions in January to February 2010 in Beijing alone produced a decrease of 14 ± 7 $\mu g \cdot m^{-3}$ ($22 \pm 6\%$) in the $PM_{2.5}$ concentration in Beijing, whereas removing residential emissions in the whole of the BTH region brought a decrease in the $PM_{2.5}$ concentrations of 28 ± 19 $\mu g \cdot m^{-3}$ ($40 \pm 9\%$), 44 ± 27 $\mu g \cdot m^{-3}$ ($43 \pm 10\%$), and 25 ± 14 $\mu g \cdot m^{-3}$ ($35 \pm 8\%$) in the three provinces in Beijing, Tianjin, and Hebei, respectively.

The residential sector has been relatively overlooked in ambient air pollution control strategies. Our analysis indicates that air quality in the Beijing region would substantially benefit from reducing residential sector emissions from both within Beijing and within surrounding provinces. To evaluate potential health benefits, however, will require additional assessment to determine changes in population exposure resulting from specific mitigation strategies. A careful assessment of the contribution of residential emissions to annual average regional ambient pollution levels as well as an analysis of the benefits of specific mitigation options would provide valuable guidance to the formation of future air quality policy designed to meet the Action Plan air quality targets.

We found regional air quality management to be of great importance. Compared with controlling emissions just in Beijing, controlling residential emissions in the BHT region resulted in twice as large a reduction in $PM_{2.5}$ concentrations (28 vs. 14 $\mu g \cdot m^{-3}$) in Beijing itself, indicating the importance of interregional transport. Therefore, to achieve consistent air quality improvements, it may be necessary not only to develop provincial air quality management plans that address household as well as other sources but to build a long-term regional framework for emission controls among all of the provinces in northern China.

Materials and Methods

We use the WRF-Chem (version 3.6) modeling system to simulate outdoor air quality. WRF-Chem is a fully coupled regional meteorology-chemistry model that simulates meteorology and the emission, transport, mixing, and chemical transformation of trace gases and aerosols (11). Our WRF-Chem domain covers China, Japan, North and South Korea, and parts of other countries (Fig. S1), with a horizontal resolution of 36 km. The vertical grid of 31 levels extends from the surface (the surface layer is ~26 m deep) to the model top of 50 hectopascals. The carbon-bond mechanism version Z (CBM-Z) gas phase chemistry (32) and the four-bin Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) aerosol module (33) were used, and the Fast-j radiation scheme (34) was chosen to calculate the photolysis rates. Boundary conditions were obtained from the Model of Ozone and Related Tracers (MOZART-4) (35) for the year 2010, at 6-h resolution. Details on model configuration are provided in *SI Materials and Methods*. Anthropogenic and biogenic emissions were included in the BASE simulation. Anthropogenic

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emissions in China for 2010 were derived from the MEIC model developed by Tsinghua University, which has been used in several other studies (27, 28). The MEIC inventory includes five anthropogenic source sectors: power, industry, transportation, residential, and agriculture (only NH_3). Open biomass burning, which usually occurs in summer and autumn (36), was not included in the study. The emission inventory considers seasonal variations by monthly emission data, and for the residential sector, the $PM_{2.5}$, BC, OC, SO_2, and NO_x emissions are typically highest in winter. Anthropogenic emissions from other Asian countries were generated from the INTEX-B emissions inventory (37). Biogenic emissions were predicted online by WRF-Chem according to the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (38).

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