



Change in household fuels dominates the decrease in PM_{2.5} exposure and premature mortality in China in 2005–2015

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To tackle the severe fine particle (PM_{2.5}) pollution in China, the government has implemented stringent control policies mainly on power plants, industry, and transportation since 2005, but estimates of the effectiveness of the policy and the temporal trends in health impacts are subject to large uncertainties. By adopting an integrated approach that combines chemical transport simulation, ambient/household exposure evaluation, and health-impact assessment, we find that the integrated population-weighted exposure to PM_{2.5} (IPWE) decreased by 47% (95% confidence interval, 37–55%) from 2005 [180 (146–219) μg/m³] to 2015 [96 (83–111) μg/m³]. Unexpectedly, 90% (86–93%) of such reduction is attributed to reduced household solid-fuel use, primarily resulting from rapid urbanization and improved incomes rather than specific control policies. The IPWE due to household fuels for both cooking and heating decreased, but the impact of cooking is significantly larger. The reduced household-related IPWE is estimated to avoid 0.40 (0.25–0.57) million premature deaths annually, accounting for 33% of the PM_{2.5}-induced mortality in 2015. The IPWE would be further reduced by 63% (57–68%) if the remaining household solid fuels were replaced by clean fuels, which would avoid an additional 0.51 (0.40–0.64) million premature deaths. Such a transition to clean fuels, especially for heating, requires technology innovation and policy support to overcome the barriers of high cost of distribution systems, as is recently being attempted in the Beijing–Tianjin–Hebei area. We suggest that household-fuel use be more highly prioritized in national control policies, considering its effects on PM_{2.5} exposures.

health impact | household air pollution | ambient air pollution | integrated exposure assessment | cooking

Ambient fine particle (PM_{2.5}) concentrations in China are among the highest in the world (1, 2). Since 2005, the Chinese government has implemented emission-control policies that have been continuously tightened. The overarching goal was to cut down the total emissions of air pollutants such as SO₂, particulate matter (PM), and NO_x (3, 4). The focus of the control policies was initially placed on power plants and transportation and extended to industrial sources after 2010. In 2013, China issued the “Air Pollution Prevention and Control Action Plan,” which marked the most stringent policy in the nation’s history (5). In this plan, power, industrial, and transportation sources remained the three foci. As a result, both bottom-up emission estimates and satellite observations indicate that China’s total emissions of SO₂ and primary PM have decreased substantially since about 2006, as well as those of NO_x since 2011 (4, 6–10).

The goal of most air pollution control, however, is to reduce exposure to protect the human health. The exposure and health impacts are induced not only by ambient air pollution (AAP), but also by household air pollution (HAP) from indoor solid-fuel use. The Global Burden of Disease (GBD) study estimated that HAP from

burning of solid cooking fuels resulted in 0.82 million premature deaths in China in 2010, without considering fuel use for space heating. This is comparable to 1.07 million due to AAP (11). Besides contributing to HAP, household fuels also act as one of the most important sources of AAP in China because of their large emission rates per unit fuel and low emission height (12–14). Household fuels have been largely neglected in China’s environmental policies until 2017, when a work plan for clean fuel substitution was launched in Beijing–Tianjin–Hebei and their surrounding areas (15, 16). Nevertheless, the transition from solid fuels to cleaner fuels happened spontaneously, due largely to urbanization-induced population migration and increased income (17–19), which leads to potentially important environmental and health benefits.

Considering the complex driving factors, there are large uncertainties in trends of the integrated population-weighted exposure to PM_{2.5} (IPWE, defined as the total population-weighted exposure to both AAP and HAP) and the associated premature mortality. A systematic assessment of these trends and the key emission sources governing the trends could help to develop optimized control policies based on their health benefits, which would be more effective

Significance

The Chinese government has taken efforts to tackle the nation’s severe ambient fine particle (PM_{2.5}) pollution. Our results suggest that reduced household solid-fuel consumption was the leading contributor to the rapid decrease in the integrated exposure to ambient and household PM_{2.5} pollution during 2005–2015, even though there was no explicit household control policy. In contrast, the emission reductions from power plants, industry, and transportation contributed much less to the decrease of integrated exposure. Clean household heating fuels have become part of recent control policies in northern China, but such policy would be strengthened if extended to heating and cooking countrywide since shift of the remaining household solid fuels to clean fuels could additionally avoid an estimated half-million premature deaths annually.

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than the current emission-oriented policies. The GBD project has conducted a decadal-scale health-impact evaluation based on global modeling and databases (20–22), but not yet created a synthesized assessment of the health impacts from AAP and HAP. An integrated approach and locally derived datasets (e.g., emission inventory and ambient and household measurements) are needed, however, for a comprehensive assessment of the trends in IPWE and associated premature mortality. In addition, it helps in identifying the key emission sources contributing to trends.

This study adopts an integrated model framework that synthesizes AAP and HAP through linking local emission inventory, chemical transport simulation, ambient/household exposure evaluation, and health-impact assessment. The objective is to investigate the trends in IPWE and associated premature mortality, and particularly the contribution of household fuels to the trends, in China during 2005–2015. The AAP exposure from primary and secondary PM_{2.5} is estimated by using the Community Multiscale Air Quality model with the 2D Volatility Basis Set (CMAQ/2D-VBS) (23) with emission inputs derived from a Chinese emission inventory developed in our previous studies (24–27) and updated to 2015 in the present study. The additional HAP exposure due to solid-fuel use is evaluated by combining local statistics, census, and household exposure measurements. Our results indicate that the reduced household solid-fuel use has been a leading contributor to the rapid decline in IPWE and the associated estimated reduction in premature mortality from air pollution in China during 2005–2015. Important air-quality and health benefits would be further achieved if the remaining household solid fuels were replaced by clean fuels.

Results and Discussion

Significant Reduction in IPWE Between 2005 and 2015. Fig. 1 *A* and *B* illustrates the IPWE in China in 2005 and its variations from 2005 to 2015. The mean IPWE for the entire China mainland, including both ambient and household pollution, was 180 $\mu\text{g}/\text{m}^3$ in 2005, with a 95% confidence interval of 146–219 $\mu\text{g}/\text{m}^3$ (see *SI Appendix, section 3* for the uncertainty analysis method). It decreased to 96 (83–111) $\mu\text{g}/\text{m}^3$ in 2015 [i.e., by ~47% (37–55%)]. The significant IPWE decrease essentially occurred across the entire mainland of China (Fig. 1*B*), with mean decreasing rates of 36% and 41% in urban and rural areas, respectively. The national mean decreasing rate exceeded that in either the urban or rural areas, because many rural residents migrated into cities where people suffer from a much smaller IPWE due to a lower HAP exposure from solid-fuel burning (Fig. 1*E*).

The significant nationwide decrease in IPWE is induced by the decline of both HAP and AAP exposures. The HAP exposure, which was seven times larger in rural areas than in urban areas, was responsible for 69% of the national mean IPWE in 2005. Note that the HAP refers to only the increased indoor PM_{2.5} exposure due to household fuels and does not include the exposure due to penetration of AAP indoors (*Methods*). The mean HAP exposure decreased by 56% (56% in urban and 45% in rural areas) during 2005–2015, as a result of reductions in household solid-fuel consumption and the associated air pollutant emissions (Fig. 2). During the 10-y period, the biomass consumption and urban coal consumption dropped by 58% (Fig. 2*A* and *B*). The coal consumption in rural households fluctuated from year to year, but decreased by 5% overall (Fig. 2*A*). A major driver behind the decrease in solid fuels is the rapid urbanization and the associated population migration, because rural residents generally obtain better access to cleaner fuels as they migrate to cities (17, 18). Another important driver is improved income, which makes cleaner fuels more affordable, as suggested by the significant positive correlation between living standards and clean fuel usage in previous surveys (19, 28, 29). As household solid-fuel consumption shrinks, the emissions of PM_{2.5}, black carbon (BC), and organic carbon (OC) from household fuels decreased by 47–57% over the period (Fig. 2*C–F*).

Compared with HAP, the AAP exposure experienced a smaller decrease of 24%, with similar decreasing rates in urban and rural areas. The decline took place in most of China and peaked in major

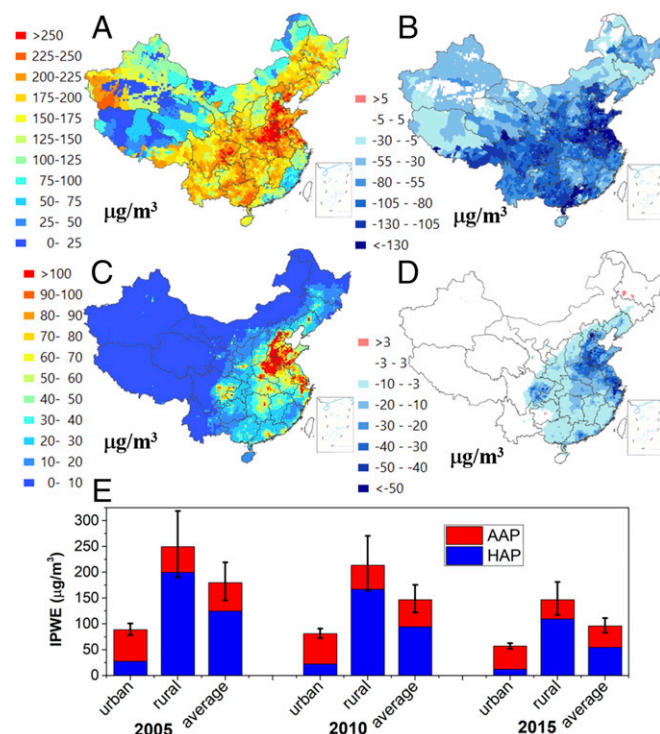


Fig. 1. IPWE in China during 2005–2015. (A–D) Spatial distribution of IPWE (A and B) and AAP exposure (C and D) in 2005 (A and C) and their changes from 2005 to 2015 (B and D). (E) IPWE and the contributions from AAP and HAP in China in 2005, 2010, and 2015. The error bars denote 95% confidence intervals of IPWE estimated by using the Monte Carlo method, as detailed in *SI Appendix, section 3*.

metropolitan regions, particularly the North China Plain (Fig. 1*D*). There is a good agreement between the simulated and observed trends in PM₁₀/PM_{2.5} concentrations and aerosol optical depth (AOD) during 2005–2015 (*SI Appendix, section 2*). The change in AAP exposure is a net result of the changes in pollutant emissions from both household and nonhousehold sources (Fig. 2 and *SI Appendix, Table S1*). In 2005, household fuels accounted for 28%, 42%, 64%, and 23% of the total emissions of PM_{2.5}, BC, OC, and nonmethane volatile organic compounds (NMVOCs), respectively, and <5% of the emissions of SO₂, NO_x, and NH₃. Along with the decrease in emissions from household fuels, the emissions of PM_{2.5}, BC, OC, and SO₂ from power plants, industry, and transportation all experienced considerable reductions due to stringent control measures, leading to an overall reduction of ~40% in total emissions of these four pollutants (Fig. 2 and *SI Appendix, Table S1*). In contrast, NO_x and NMVOC emissions increased by 10% and 36%, respectively, during the 10 y, since the decrease in emissions from household fuels and power plants was not sufficient to offset the growth in other sectors (Fig. 2 and *SI Appendix, Table S1*). All aforementioned emission trends of primary PM and gaseous precursors together result in the moderate decrease in AAP exposure.

Source Contribution to IPWE Reduction. We attribute the changes in IPWE to individual factors, including AAP and HAP from household coal, household biomass, other household fuels (primarily gaseous fuels), and nonhousehold sources, as well as meteorological conditions (*Methods*), as illustrated in Fig. 3*A*. In 2005, among all individual sources, HAP due to household biomass made the largest contribution (110 $\mu\text{g}/\text{m}^3$, 61%) to the mean IPWE in China, followed by AAP from nonhousehold sources (43 $\mu\text{g}/\text{m}^3$, 24%) and HAP from household coal burning (14 $\mu\text{g}/\text{m}^3$, 8%). The large contribution from household biomass results from a combination of large fractions of residents using biomass as their main cooking fuels (10% and 70%, respectively,

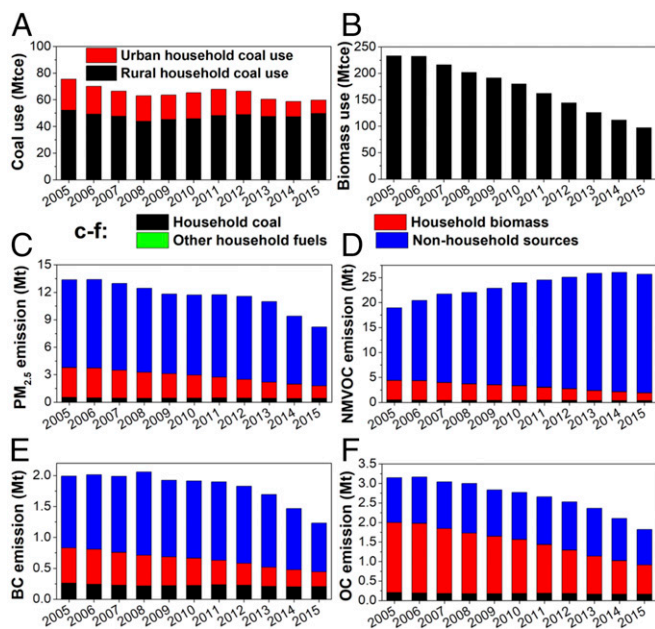


Fig. 2. Household energy consumption and sectoral emissions of major air pollutants in China during 2005–2015: household coal consumption (A); household biomass consumption (B); and emissions of PM_{2.5} (C), NMVOC (D), BC (E), and OC (F) from household and nonhousehold sources. Mt, million tons; Mtce, million tons coal equivalent.

in urban and rural areas in 2005; *SI Appendix, Fig. S3*) and high PM_{2.5} exposures in biomass-using households.

From 2005 to 2015, HAP from household biomass decreased by 67 (41–100) $\mu\text{g}/\text{m}^3$ as a result of a remarkable decrease in biomass consumption by >50% (based on three independent nationwide statistics/surveys; *SI Appendix, section 1 and Fig. S1*). This represents the largest contributor to the reduction of IPWE during the 10-y period, far exceeding the contributions from any other source (Fig. 3A). The AAP from nonhousehold sources and AAP from household biomass, which decreased by 10 and 6 $\mu\text{g}/\text{m}^3$, respectively, are the second and third largest contributors to IPWE reduction. The features of the source contributions differed in urban and rural areas. In the urban area, AAP from nonhousehold sources played the most important role in reducing IPWE, followed by HAP due to household biomass. In the rural area, however, HAP due to household biomass stood out as the dominant factor due to widespread biomass uses. HAP due to household coal contributed 4 $\mu\text{g}/\text{m}^3$ to the decrease in urban IPWE as a result of the >50% decline in urban coal consumption, while its contribution to rural IPWE changes was quite small because of the insignificant change in rural coal use. With the effects of individual fuels combined, the IPWE attributed to all household fuels decreased dramatically by 76 (48–109) $\mu\text{g}/\text{m}^3$ during 2005–2015, representing as high as 90% (86–93%) of the total IPWE reduction. In contrast, the contribution from power plants to the total IPWE reduction was only 4 $\mu\text{g}/\text{m}^3$, or 5% (Fig. 3A), although the power sector as the focus of China’s control policies constituted ~90% of the SO₂ emission reductions since 2005 and 70% of the NO_x reductions since 2011. In most provinces, household-fuel use is the largest contributor to IPWE and its decrease from 2005 to 2015. In some developed provinces (such as Beijing, Tianjin, Shanghai, and Guangdong), however, AAP due to nonhousehold sources plays the most important role (*SI Appendix, Fig. S4*).

We further separately estimate the IPWE from household fuels used for cooking and space heating, as they have quite different policy implications (see *SI Appendix, section 5* for methods). The results are shown in Fig. 3B. In 2005, the IPWE attributed to cooking is ~2.5 times as much as that due to space heating, since heating is only needed in the winter of northern and central

China. From 2005 to 2015, the IPWE attributed to both cooking and heating decreased, but the decreasing rate of solid-fuel cooking (62% nationwide and 53% in rural areas) is significantly larger than that of heating (39% nationwide and 24% in rural areas). This is because the heating activities using natural gas or electricity require a more expensive rural energy distribution system (e.g., the natural gas pipeline network or terminal power grid with sufficient capacity) (30). Other clean energy sources for heating, such as solar energy, geothermal energy, and industrial waste heat, are limited by resource availability. In contrast, various clean cooking energy technologies, particularly those using liquefied petroleum gas, biogas, and electricity, have been increasingly affordable and accessible for many rural residents.

The results presented above are different from traditional source apportionment analysis which only focused on AAP. In 2005, household fuels account for 21% of the AAP exposure in China (compare 76% of IPWE). From 2005 to 2015, household-fuel use contributes 42% to the decrease of AAP exposure, whereas its contribution to the decrease of IPWE is 90%.

Health Impacts and the Role of Household Fuels. Using the IPWE as input, we apply the integrated exposure–response (IER) functions to estimate the PM_{2.5}-related premature mortality (*Methods*), as shown in Fig. 4. In 2005, the PM_{2.5}-related premature deaths amounted to 1.72 (1.47–1.99) million. The marginal contribution of household fuels was estimated at 0.91 (0.72–1.13) million, 53% (46–60%) of the total (see *Methods* for the quantification approach). Considering the curvilinear shape of the IER functions, the marginal contribution would have been even larger if the emissions from nonhousehold sources had been lower. Approximately 80% of

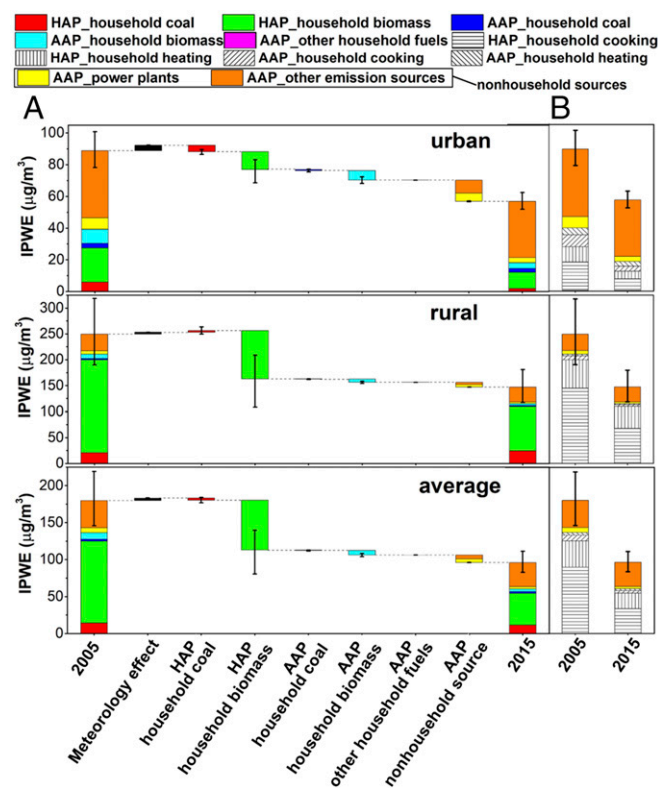


Fig. 3. Contribution of individual sources to IPWE and its changes in China. (A) Contribution of each household-fuel type and nonhousehold sources to IPWE in 2005/2015 (leftmost and rightmost stacked columns) and the changes between the 2 y (in-between columns) in urban (Top), rural (Middle), and all (Bottom) areas. (B) Contribution of household fuels used for cooking/heating and nonhousehold sources to IPWE in 2005/2015. The error bars denote 95% confidence intervals estimated by using the Monte Carlo method.

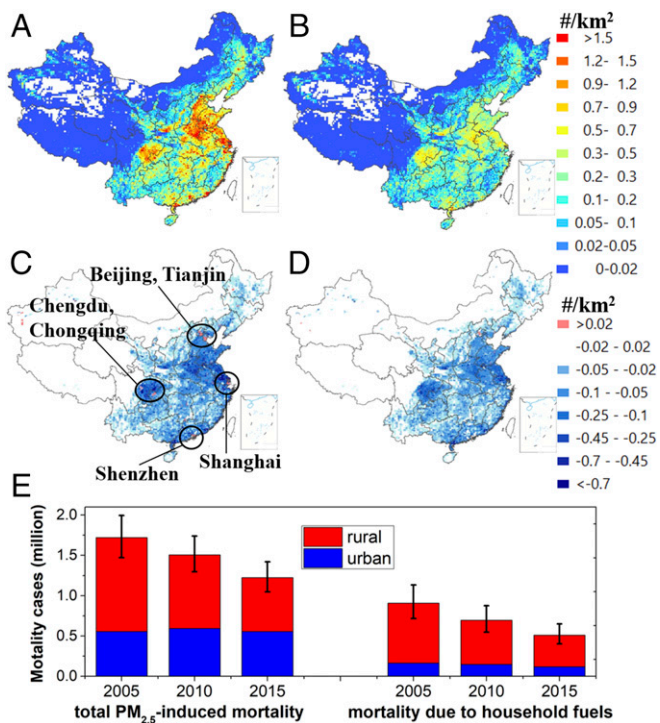


Fig. 4. PM_{2.5}-related premature mortality and the contribution from household fuels in China during 2005–2015. (A–D) Spatial distribution of total PM_{2.5}-related mortality (A and C) and the mortality attributed to household fuels (B and D) in 2005 (A and B) and their changes from 2005 to 2015 (C and D). (E) Total PM_{2.5}-related premature mortality and the mortality due to household fuels in China in 2005, 2010, and 2015. The error bars denote 95% confidence intervals estimated using the Monto Carlo method, as detailed in *SI Appendix, section 3*.

the household-related premature deaths occurred among rural residents (Fig. 4E).

Following the substantial reduction in IPWE, the PM_{2.5}-related premature deaths in 2015 were 29% (22–36%) lower than the 2005 level. The decrease occurred over most of China, except for some urban centers such as Beijing, Shanghai, Shenzhen, and Chengdu (Fig. 4C), where the mortality has increased because the effects of IPWE reduction were counteracted by a rapid increase in the migrant population. Nationwide, the total urban premature mortality remained stable, while the rural mortality decreased dramatically by ~43% (Fig. 4E). During the same period, the mortality due to household fuels decreased by 0.40 (0.25–0.57) million in China, accounting for 80% (69–88%) of the total reduction in PM_{2.5}-related mortality. Almost everywhere in China has witnessed a reduction in household fuel-induced mortality (by 29% and 49% in urban and rural areas, respectively), except for very few spots which have dramatic population growth, such as parts of Beijing (Fig. 4D). Note that the preceding trends in premature mortality are the combined effect of multiple factors, including changes in IPWE, population, age distribution, and background mortality rate. The IPWE, however, is proved to be the predominant contributor to the changes in premature mortality (*SI Appendix, section 6*).

Additional Benefits from Replacing Remaining Household Solid Fuels with Clean Fuels. In 2015, household fuels still contribute 64% of the IPWE and at least 43% of PM_{2.5}-related mortality. In 2017, an action plan for clean heating (15, 16) was launched in northern China (14 provinces), with a focus on Beijing–Tianjin–Hebei and the surrounding areas. The overarching goal is to increase the fraction of clean heating in northern China to 70% by 2021, which means that ~55% of the existing household solid fuels for heating in these provinces shall be replaced with clean energy. We assume

that half of the solid fuels are replaced by natural gas and the other half by electricity (see *SI Appendix, section 7* for detailed methods). A successful implementation of this policy would reduce the emissions of PM_{2.5}, BC, and OC from household fuels by 15–17%, which could subsequently reduce the IPWE by 9.7% (8.8–10.4%) in China and by 21% (19–23%) in northern China (Fig. 5A); this accounts for associated increased emissions from power generation. This is estimated to avoid 0.055 (0.045–0.075) million premature deaths annually (Fig. 5B). Furthermore, if all solid fuels used for cooking and heating in 2015 were thoroughly substituted by electricity and natural gas (50% each), the IPWE in China would be lowered by 60 (47–75) $\mu\text{g}/\text{m}^3$, or 63% (57–68%) of the total (Fig. 5A). The reductions in HAP and AAP exposures would be 54 and 6 $\mu\text{g}/\text{m}^3$, respectively. This implies that ~0.51 (0.40–0.64) million premature deaths could be avoided annually (Fig. 5B).

The estimated health benefit is expected to be even larger if nonhousehold sources were jointly controlled, considering the curvilinear IER functions. The environmental and health benefits of substitution by either electricity or natural gas are similar because the exposure increase due to additional electricity or natural gas consumption are much smaller than the exposure decrease due to reduced solid fuels (*SI Appendix, section 7*). Perhaps surprisingly, the environmental and health benefits are largely insensitive to the assumed energy mix of power systems to supply the needed electricity due to the large difference in intake fractions between household sources and power plants (*SI Appendix, section 7*). All of the preceding control options would bring more dividend to rural people who have been exposed to the highest levels of IPWE—specifically, approximately three-quarters of the avoided premature deaths would be rural residents.

Policy Implications

As stated previously, the decrease in solid-fuel consumption in China during 2005–2015 was primarily driven by rapid urbanization and improved income rather than specific control policies. Given the ongoing urbanization and economic development in China, it is fair to expect that the transition toward clean fuels for cooking will continue, even if no control policy is implemented. The spontaneous transition, however, is expected to slow down due to the slower economic growth and urbanization rate (31). The transition in heating fuels presents a bigger challenge because of the foreseeable barriers of infrastructure development, such as the construction of a natural-gas pipeline network or an upgrade of terminal power grid in the rural areas and “urban villages” in China (30). In addition, the high cost (30) and limited supply of natural gas (32, 33) and electricity may also hinder the transition toward cleaner heating fuels. Indeed, these factors may have prevented many residents

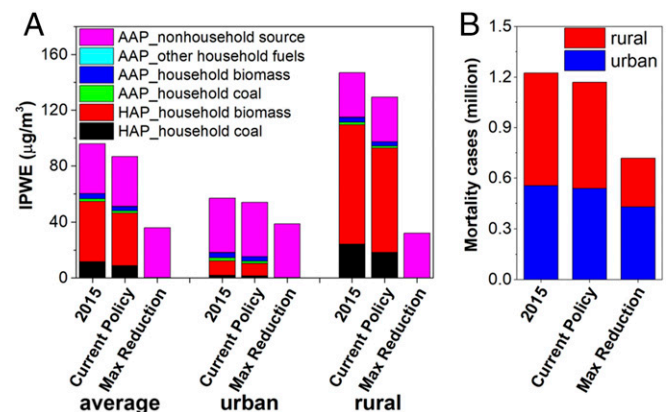


Fig. 5. The impact of replacing household solid fuels with clean energy on IPWE (A) and premature mortality (B) in China. “Current Policy” denotes a scenario in which the official work plan released in 2017 was realized, and “Max Reduction” is a scenario in which all household solid fuels were substituted by electricity and natural gas.

from changing from biomass or coal to clean energy in the last decade. This can be inferred because total rural coal consumption remained relatively stable during 2005–2015 (Fig. 2A), even when the rural population decreased significantly. Promoting and expediting the transition from solid fuels to clean energy (electricity or natural gas), particularly for heating, involves affordable technology innovation, infrastructure construction, clean fuel supply, and financial subsidies. Besides, improving the thermal performance of rural housing through better wall insulation and fenestration could reduce over half of the space heating demand (34), thus lowering the barriers to clean energy transition.

Until recently, China's control policies have primarily targeted large point sources, particularly power plants, with the overarching goal of reducing total emissions of SO₂ and NO_x (and ambient PM_{2.5} concentrations since 2013). Nevertheless, the IPWE reduction due to the emission controls in power plants has been only 5% of that due to the decreased household-fuel use during 2005–2015. In addition, ~90% of the household-related IPWE results from HAP exposure (rather than AAP exposure), but HAP has not been on the agenda of the policymakers in China in recent decades.

We suggest that IPWE should be used as a key metric for the effectiveness evaluation of air-pollution control policies and that the current control policies should be reevaluated and revised based on their benefits on reducing IPWE. As we have defined it, it includes exposures from AAP and from household fuels, two large sources, but could be expanded in the future to accommodate the higher relative exposures to other near-field sources, such as neighborhood industries and vehicles, as has been suggested in India (35). Importantly, household fuels would then be prioritized in health-oriented control policies, given their dominant role in IPWE and associated health impacts. As described above, an action plan for clean heating (15, 16) was launched in northern China in 2017 and was expected to lead to significant health benefits, although this policy was motivated by the need to tackle ambient PM_{2.5} pollution rather than IPWE (15, 16). This plan could also lead to a faster transition in cooking fuels since the rural residents will have easier access to clean fuels after the energy distribution system is constructed or improved. Such efforts are much needed and shall be gradually strengthened and extended to cover solid fuels for both heating and cooking across the whole country, since shift of the remaining household solid fuels to clean fuels could additionally avoid nearly half a million premature deaths. Finally, the present study may also provide guidance to other developing countries, such as India (21, 36), which suffer from similarly severe air pollution due to solid-fuel burning.

Methods

Evaluation of IPWE. IPWE was used to measure the total population-weighted exposure to PM_{2.5} through both AAP and HAP. It is defined as the weighted sum of PM_{2.5} concentrations in all microenvironments where people spend time, including the kitchen, living room, bedroom, outdoor environment, etc. (37). The GBD study as well as most other environmental health studies (20, 21, 38) treated AAP and HAP as separate risks; there are overlaps between the two since the HAP includes contributions from the AAP. In this study, the concept of AAP is consistent with GBD and most other studies (12, 22, 39) which assume that AAP generally penetrates into the household and constitutes a basic exposure level for all people. The HAP refers to only the additional PM_{2.5} exposure due to household-fuel use (37). Thus, the population-weighted exposures from AAP and HAP add up to the total IPWE. This assumption only affected the partitioning between AAP and HAP and did not affect the total IPWE or the conclusion of the present study (SI Appendix, section 4). Another difference from GBD is that we also included noncooking fuels (particularly heating fuels), whose contribution to AAP is fully considered, and the contribution to HAP was indirectly accounted for. IPWE is expressed as:

$$\text{IPWE} = \text{PWE}_{\text{AAP}} + \text{PWE}_{\text{HAP}}, \quad [1]$$

where PWE_{AAP} is the population-weighted PM_{2.5} exposure due to AAP and PWE_{HAP} is the extra population-weighted exposure due to HAP.

PWE_{AAP} was calculated by using the average of ambient PM_{2.5} concentrations in each geographic unit, weighted by the population in that geographic unit. The ambient primary and secondary PM_{2.5} concentrations were simulated by the CMAQ/2D-VBS model (23) at 36- × 36-km resolution (see SI Appendix,

section 2 for details). To provide input to the CMAQ/2D-VBS model, we updated the Chinese emission inventory developed in our previous studies (24–27) to 2015 (see SI Appendix, section 1 for details). The inventory included both primary PMs (PM₁₀, PM_{2.5}, BC, and OC) and gaseous pollutants (SO₂, NO_x, NMVOC, and NH₃) which contribute to secondary PM_{2.5} formation. The county-level populations were acquired from Chinese statistics, and the subcounty distribution of population was based on the LandScan dataset at ~1-km resolution (40). The geographic unit used in calculation was the intersection of counties and 36- × 36-km model grids, so that the data sources with the highest resolution were utilized. Since regional chemical transport models usually underestimate PM_{2.5} concentrations in the urban centers (by ~17% in this study; SI Appendix, section 2) while representing rural areas better, we adjusted PM_{2.5} concentrations in urbanized counties (defined as those with population density >500 per km²) based on monitoring data in 2015 from the Ministry of Environmental Protection's nationwide network covering 1,497 sites in 367 cities, following Brauer et al. (41) and Aunan et al. (37). The same adjustment factors were also applied to 2005 and 2010, considering that the model captures the temporal trends in PM_{2.5}/PM₁₀ concentrations very well (SI Appendix, section 2). This treatment minimized the bias in the relative contributions from AAP and HAP to IPWE.

PWE_{HAP} is estimated as:

$$\text{PWE}_{\text{HAP}} = \frac{1}{P} \sum_{i,j,k} (P_{i,j,k} \cdot \text{HAP}_{j,k}), \quad [2]$$

where P is population, HAP is the extra PM_{2.5} exposure levels of solid-fuel users, i refers to geographic unit, j refers to setting (urban or rural), and k refers to main household cooking fuel type (i.e., coal and biomass). $\text{HAP}_{j,k}$ was estimated by Mestl et al. (42) and subsequently updated in our previous study (37). It was calculated as the proportion of time spent in the different microenvironments (kitchen, living room, bedroom, indoors away from home, and outdoors) multiplied by the PM_{2.5} concentration in the given microenvironment. The PM_{2.5} concentrations in various microenvironments were obtained by summarizing a wide range of measurements in China, and the age, sex and season specific time-activity patterns for urban and rural populations were gathered from literature and surveys (37, 42). We classified a number of "exposure regimes" based on urban/rural setting and main cooking fuels, which were demonstrated to be key determinant factors of HAP exposure levels (37). The annual mean $\text{HAP}_{j,k}$ for urban and rural biomass users was estimated to be 223 (95% confidence interval, 125–321) and 250 (180–320) μg/m³, respectively, and the corresponding values for urban and rural coal users were 38 (28–48) and 117 (98–136) μg/m³, respectively. No extra HAP exposure was considered for clean fuel users. It should be noted that many households use more than one type of fuel, and in some settings, solid fuels are used both for cooking and heating. These impacts were indirectly taken into account in the HAP exposure estimates (37) through the fact that HAP measurements were carried out in settings where heating existed if needed and fuel mixtures often occurred. There were insufficient data to separately estimate the HAP exposure levels for cooking and heating or for multiple fuel mixtures. A nationwide survey (28) revealed that the fraction of solid-fuel users for cooking correlates well with that for heating, supporting our classification according to main cooking fuel. We also calculated IPWE using the HAP exposure levels from the GBD study (21), which are based on in-situ measurements in India, and compared them with the estimate in the present study (SI Appendix, section 8).

Regarding populations using coal and biomass as their main cooking fuels ($P_{i,j,k}$ in Eq. 2), the National Population Census (43, 44) provides county-level data in 2010, which were subsequently combined with provincial-level statistics of household coal and biomass consumption during 2005–2015 (described in SI Appendix, section 1) to derive county-level solid fuel-using populations during 2005–2015, as illustrated in SI Appendix, Fig. 53. The rationale behind this is that the total exposure amount ($P_{i,j,k} \cdot \text{HAP}_{j,k}$ in Eq. 2) for a specific geographic unit, setting (urban or rural), and solid-fuel type is proportional to the solid-fuel consumption, under the assumption that the stove technology remains unchanged over time (see SI Appendix, section 4 for more discussions). A large-scale survey conducted in 2012 reported that 12% and 48% of the urban and rural residents used biomass as their main cooking fuels (28), which is comparable to our estimates (7% and 49%, respectively).

Health-Impact Assessment. Here, we used premature deaths as a health indicator. We estimated the premature deaths attributable to PM_{2.5} pollution based on relative risks of mortality, baseline mortality rate, and population (22, 45). We calculated the relative risks of mortality as a function of PM_{2.5} exposure (IPWE in this study), employing the age- and sex-specific IER functions developed by Cohen et al. (22), which is an updated version of Burnett et al. (45). IER functions were constructed by combining risk estimates from studies of AAP, HAP, and active and second-hand smoking that

cover a full PM_{2.5} exposure range from very small to ~30,000 µg/m³ (22, 45). Therefore, they are suitable for this study which involves large PM_{2.5} exposures from both AAP and HAP over a highly polluted region. The health endpoints considered include ischemic heart disease, stroke, bronchus and lung cancer, and chronic obstructive pulmonary disease for adults and lower respiratory infections for children and adults. We obtained the disease-specific baseline mortality rates by age and gender from the Institute of Health Metrics and Evaluation (46).

Quantification of the Contribution from Individual Sources. We quantified the marginal contribution of a specific emission source (e.g., household coal) to both IPWE and premature deaths by designing a hypothetical scenario in which the air pollutant emissions and HAP exposure from this source are eliminated and comparing it with the baseline scenario where all sources are included. Because of the nonlinearity in emission–concentration relationships, the sum of contributions from household coal, household biomass,

and other household fuels to IPWE is not exactly equal to the contribution from all household fuels. Their difference, however, is within 3% according to our simulation results. Besides, we quantified the effect of meteorological changes using the difference between the baseline simulations in 2005 and a sensitivity scenario where the emissions in 2005 and meteorological fields in 2015 were employed.

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- van Donkelaar A, Martin RV, Brauer M, Boys BL (2015) Use of satellite observations for long-term exposure assessment of global concentrations of fine particulate matter. *Environ Health Perspect* 123:135–143.
- Zhang R, et al. (2015) Formation of urban fine particulate matter. *Chem Rev* 115: 3803–3855.
- Wang S, Hao J (2012) Air quality management in China: Issues, challenges, and options. *J Environ Sci (China)* 24:2–13.
- Wang J, et al. (2017) Particulate matter pollution over China and the effects of control policies. *Sci Total Environ* 584–585:426–447.
- Cai S, et al. (2017) The impact of the “Air Pollution Prevention and Control Action Plan” on PM_{2.5} concentrations in Jing-Jin-Ji region during 2012–2020. *Sci Total Environ* 580:197–209.
- Krotkov NA, et al. (2016) Aura OMI observations of regional SO₂ and NO₂ pollution changes from 2005 to 2015. *Atmos Chem Phys* 16:4605–4629.
- Zhao B, et al. (2017) Decadal-scale trends in regional aerosol particle properties and their linkage to emission changes. *Environ Res Lett* 12:054021.
- Xia YM, Zhao Y, Nielsen CP (2016) Benefits of China’s efforts in gaseous pollutant control indicated by the bottom-up emissions and satellite observations 2000–2014. *Atmos Environ* 136:43–53.
- Liu F, et al. (2016) Recent reduction in NO_x emissions over China: Synthesis of satellite observations and emission inventories. *Environ Res Lett* 11:114002.
- de Foy B, Lu ZF, Streets DG (2016) Satellite NO₂ retrievals suggest China has exceeded its NO_x reduction goals from the twelfth Five-Year Plan. *Sci Rep* 6:35912.
- Global Burden of Disease Collaborative Network (2017) IHME Global Burden of Disease Study 2016 (GBD 2016) results (Institute for Health Metrics and Evaluation, Seattle).
- Lelieveld J, Evans JS, Fnais M, Giannadaki D, Pozzer A (2015) The contribution of outdoor air pollution sources to premature mortality on a global scale. *Nature* 525: 367–371.
- Liu J, et al. (2016) Air pollutant emissions from Chinese households: A major and underappreciated ambient pollution source. *Proc Natl Acad Sci USA* 113:7756–7761.
- Chafe ZA, et al. (2014) Household cooking with solid fuels contributes to ambient PM_{2.5} air pollution and the burden of disease. *Environ Health Perspect* 122: 1314–1320.
- Ministry of Environmental Protection of China (2017) Work plan for air pollution control in Beijing-Tianjin-Hebei and its surrounding areas in 2017 (Ministry of Environmental Protection of China, Beijing). Available at dqhj.mee.gov.cn/dtxx/201703/t20170323_408663.shtml. Accessed October 30, 2018.
- National Development and Reform Commission of China (2017) Work plan for clean heating in winter in northern China (2017–2021) (National Development and Reform Commission of China, Beijing). Available at www.gov.cn/xinwen/2017-12/20/content_5248855.htm. Accessed July 1, 2018.
- Shen H, et al. (2017) Urbanization-induced population migration has reduced ambient PM_{2.5} concentrations in China. *Sci Adv* 3:e1700300.
- Aunan K, Wang S (2014) Internal migration and urbanization in China: Impacts on population exposure to household air pollution (2000–2010). *Sci Total Environ* 481: 186–195.
- Tao S, et al. (2018) Quantifying the rural residential energy transition in China from 1992 to 2012 through a representative national survey. *Nat Energy* 3:567–573.
- Forouzanfar MH, et al.; GBD 2015 Risk Factors Collaborators (2016) Global, regional, and national comparative risk assessment of 79 behavioural, environmental and occupational, and metabolic risks or clusters of risks, 1990–2015: A systematic analysis for the Global Burden of Disease Study 2015. *Lancet* 388:1659–1724.
- Smith KR, et al.; HAP CRA Risk Expert Group (2014) Millions dead: How do we know and what does it mean? Methods used in the comparative risk assessment of household air pollution. *Annu Rev Public Health* 35:185–206.
- Cohen AJ, et al. (2017) Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: An analysis of data from the Global Burden of Diseases Study 2015. *Lancet* 389:1907–1918.
- Zhao B, et al. (2016) Quantifying the effect of organic aerosol aging and intermediate-volatility emissions on regional-scale aerosol pollution in China. *Sci Rep* 6:28815.
- Zhao B, et al. (2013) NO_x emissions in China: Historical trends and future perspectives. *Atmos Chem Phys* 13:9869–9897.
- Wang SX, et al. (2014) Emission trends and mitigation options for air pollutants in East Asia. *Atmos Chem Phys* 14:6571–6603.
- Zhao B, et al. (2013) Impact of national NO_x and SO₂ control policies on particulate matter pollution in China. *Atmos Environ* 77:453–463.
- Fu X, et al. (2017) Increasing ammonia concentrations reduce the effectiveness of particulate pollution control achieved via SO₂ and NO_x emissions reduction in East China. *Environ Sci Technol Lett* 4:221–227.
- Duan XL, et al. (2014) Household fuel use for cooking and heating in China: Results from the first Chinese Environmental Exposure-Related Human Activity Patterns Survey (CEERHAPS). *Appl Energy* 136:692–703.
- Cai S, et al. (2018) Pollutant emissions from residential combustion and reduction strategies estimated via a village-based emission inventory in Beijing. *Environ Pollut* 238:230–237.
- Lei Y (2017) Residential coal control policies should take all factors into consideration. *China Energy News*. Available at paper.people.com.cn/zgnyb/html/2017-12/04/content_1821708.htm. Accessed July 1, 2018.
- Jiang LW, O’Neill BC (2017) Global urbanization projections for the shared socio-economic pathways. *Glob Environ Change* 42:193–199.
- Hornby L, Zhang A (2017) China hit by gas shortages as it moves away from coal. *Financial Times*. Available at https://www.ft.com/content/21cb4ed2-d7f9-11e7-a039-c64b1c09b482. Accessed July 1, 2018.
- Kang Z (2014) Natural gas supply-demand situation and prospect in China. *Nat Gas Ind B* 1:103–112.
- Shan M, Wang P, Li J, Yue G, Yang X (2015) Energy and environment in Chinese rural buildings: Situations, challenges, and intervention strategies. *Build Environ* 91: 271–282.
- Sagar A, Balakrishnan K, Guttikunda S, Roychowdhury A, Smith KR (2016) India leads the way: A health-centered strategy for air pollution. *Environ Health Perspect* 124: A116–A117.
- Smith KR (2000) National burden of disease in India from indoor air pollution. *Proc Natl Acad Sci USA* 97:13286–13293.
- Aunan K, Ma Q, Lund MT, Wang S (2018) Population-weighted exposure to PM_{2.5} pollution in China: An integrated approach. *Environ Int* 120:111–120.
- World Health Organization (2016) World health statistics 2016: Monitoring health for the SDGs, sustainable development goals (World Health Organization, Geneva). Available at www.who.int/gho/publications/world_health_statistics/2016/en/. Accessed July 1, 2018.
- GBD MAPS Working Group (2018) *Tabulation of the 2010 Population Census of the People’s Republic of China* (China Statistics Press, Beijing).
- All China Marketing Research Co. Ltd. (2015) China 2010 county population census data with GIS maps (All China Marketing Research Co. Ltd., Beijing). Available at https://chinadatacenter.net/Data/Services.aspx. Accessed October 30, 2018.
- Burnett RT, et al. (2014) An integrated risk function for estimating the global burden of disease attributable to ambient fine particulate matter exposure. *Environ Health Perspect* 122:397–403.
- Global Burden of Disease Collaborative Network (2017) Global Burden of Disease Study 2016 (GBD 2016) results tool (Institute for Health Metrics and Evaluation, Seattle). Available at ghdx.healthdata.org/gbd-results-tool. Accessed July 1, 2018.