Supplementary Information for "Change in household fuels dominates the decrease in $PM_{2.5}$ exposure and premature mortality in China in 2005-2015"

1. Development of emission inventory

We develop an up-to-date emission inventory of SO_2, NOx, PM_{10}, $PM_{2.5}$, BC, OC, NMVOC, and NH_3 in China for 2005-2015 using the "emission factor method" based on the activity data (energy consumption, industrial product yields, etc.), technology-based uncontrolled emission factors, and penetrations of control technologies. The methodology and data sources to develop the emission inventory were detailed in our previous papers (1-5), in which the emissions as of 2012 were calculated. Here we update the emission inventory till 2015, drawing upon the latest information on energy consumption, technology distribution, and control measures, from a variety of sources including statistical data, government bulletins, industrial associations, technological reports, and research surveys. As a focus of this study, we obtain the consumption of all fuels except biomass in the residential and commercial sectors in both urban and rural areas from the Energy Statistical Yearbook of China (6). All fuel consumption in the residential sector in rural areas is assumed to occur in the households, while in urban areas, the fraction of household-fuel use in total residential and commercial energy consumption is achieved from a detailed bottom-up model developed by the Tsinghua University Building Energy Research Center (Tsinghua-BERC) (7-11). SI Appendix Fig. S1a compares the rural coal consumption used in this study (i.e., official statistics) with the estimates from a representative national survey reported by Tao et al. (12), which collected the fuel use data for 2012 as well as recalled historical data for 2002 and 2007. The considerable discrepancy between these two data sources indicates a relatively large

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uncertainty in rural coal consumption, which is considered in the Monto Carlo uncertainty analysis described in SI Appendix Section 3. In addition, we conducted a sensitivity analysis in which the IPWE in 2005-2015 and the contribution from household fuels were calculated using the rural coal consumption in Tao et al. (2018). In this case, the IPWE in 2005 is 3.5% larger than our current estimate. Also, household fuels are estimated to lead to a reduction of 81 $\mu g/m^3$ in IPWE from 2005 to 2015, representing 91% of the total IPWE reduction, which is also similar to the estimates in the current study (76 $\mu g/m^3$ and 90%). Therefore, the discrepancy in rural coal consumption does not change the conclusion of this study. For biomass consumption, there were no published official statistics after 2007. We obtain the statistics compiled by the Ministry of Agriculture of China (the data as of 2007 were published in *China Energy Statistical Yearbook* (13) and those after 2007 are unpublished), nationwide survey results from the Tsinghua-BERC (7-11), and the data from International Energy Agency (14), as summarized in SI Appendix Fig. S1. All these three datasets reveal a consistent trend, namely the biomass consumption in China decreased by over 50% during 2005-2015. In this study, we take the average of the former two datasets (which are bottom-up statistical or survey data by local institutes) as our "best estimate", and account for the discrepancies among different datasets in the uncertainty analysis (SI Appendix Section 3). SI Appendix Fig. S1b shows that our "best estimate" agrees well with the survey data by Tao et al. (12) for years of 2002, 2007, and 2012. The household fuel use and sectoral emissions of major air pollutants are summarized in Fig. 2 and SI Appendix Table S1.

2. Configuration and evaluation of the CMAQ/2D-VBS simulations

We simulate the ambient concentrations of primary and secondary $PM_{2.5}$ in China using the CMAQ/2D-VBS model, which was developed by incorporating the 2D-VBS model framework into the default CMAQ model (15). The CMAQ/2D-VBS model explicitly simulates the aging of primary organic aerosol (POA), photo-oxidation of intermediate-volatility organic compounds (IVOCs) and multi-generational aging of secondary organic aerosol (SOA), thus improving the simulation results of SOA. Here we use the "High-Yield VBS" configuration documented in Zhao et al. (15), which agrees best with surface organic aerosol (OA) and SOA observational data. We apply the CMAQ/2D-VBS model to a domain covering China and its surrounding areas (see SI Appendix Fig. S2) with a grid resolution of 36 km×36 km. The Weather Research and Forecasting Model (WRF, version 3.7) is used to generate the meteorological fields. The physical and chemical options, vertical resolution, initial and boundary conditions of WRF and CMAQ/2D-VBS, as well as the spatial allocation of anthropogenic emissions and calculation of biogenic emissions are the same as Zhao et al. (16). The simulation periods cover 2005, 2010, and 2015, as well as 2013 for evaluation purpose.

We compare the meteorological parameters simulated by WRFv3.7 with observational data obtained from the National Climatic Data Center (NCDC), where hourly or 3-h observations are available for about 380 sites in the domain. The variables of interest include the temperature at 2 m, wind speed at 10 m, and humidity at 2 m. The statistical indices used include the mean observation (Mean OBS), mean simulation (Mean SIM), mean bias (MB), gross error (GE), root mean square error (RMSE), and index of agreement (IOA), the definitions of which are provided in Emery et al. (17). SI Appendix Table S2 summarizes the model performance statistics as well as the benchmarks suggested by Emery et al. (17). Obviously almost all indices fall within the benchmark range except that the MB of temperature slightly exceeds the range in a couple of cases. The occasional slight exceedance is considered to be reasonable since the benchmarks were derived based on performance statistics of meteorological simulations mostly at resolutions of

12 km or 4 km, which generally give more accurate predictions than those at 36 km resolution. Therefore, the statistics indicate an overall decent performance of meteorological predictions.

In addition, we compare the simulated aerosol optical depth (AOD) and NO_2 vertical column density with retrievals from the Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the Aqua satellite (http://ladsweb.nascom.nasa.gov/data/search.html) and Ozone Monitoring Instrument (OMI) onboard the Aura satellite (http://www.temis.nl/airpollution/no2.html), respectively. Only model outputs that are closest to the satellite overpassing time (13:00-14:00 for both MODIS/Aqua and OMI/Aura, Beijing time) are used in comparison to avoid sampling errors due to diurnal cycles. The conversion of vertically resolved aerosol mass concentrations to AOD follows Heald (18) and Martin and Heald (19). SI Appendix Table S3 summarizes the model performance statistics over the Eastern and Central China (ECC, see SI Appendix Fig. S2). The ECC region covers most of the populous areas in China characterized by substantial anthropogenic emissions and high ambient $PM_{2.5}$ concentrations. In contrast, the remaining parts of China are significantly affected by natural sources (e.g., wind-blown dust) and foreign emissions, which are not the focus of this study. The statistical indices we apply are slightly different from those applied to the evaluation of meteorological predictions, so as to facilitate inter-study comparison with previous studies (e.g., Zhang et al. (20) and Zhao et al. (21)). These indices include Mean OBS, Mean SIM, normalized mean bias (NMB), normalized mean error (NME), mean fractional bias (MFB), mean fractional error (MFE), RMSE, and correlation coefficient (R), as documented in Yu et al. (22) and Boylan and Russell (23). The simulated NO_2 column agrees fairly well with OMI retrievals in all seasons; the NMBs and correlation coefficients are all $\leq \pm 29\%$ and ≥ 0.69, and are $\leq \pm 20\%$ and ≥ 0.80 in most cases. There is also a good agreement between simulated and MODIS-retrieved annual mean

AOD, with NMBs within ±5%, whilst the model tends to overestimate AOD in winter and underestimate AOD in summer. This is a common issue in previous CMAQ simulation studies in China (3, 24, 25) and is yet to be addressed in future investigations. With respect to the inter-annual trend which is the focus of the current study, the retrieved and simulated AOD show 17% and 18% decrease from 2005 to 2015, respectively, indicating a good agreement. Both OMI retrievals and CMAQ/2D-VBS simulations reveal that NO_2 column increased dramatically from 2005 to 2010, followed by a significant decline from 2010 to 2015. The overall percentage changes from 2005 to 2015 are 10% and 5%, respectively.

For surface $PM_{2.5}$ concentrations, there were no nationwide continuous observations during the simulation period since the observational network changed substantially between 2012 and 2013. Since January 2013, the Ministry of Environmental Protection of China (MEP, http://datacenter.mep.gov.cn/) provides hourly $PM_{2.5}$ concentrations at 496 sites located in 74 major cities in China, including capital cities of all provinces and prefecture-level cities in three metropolitan regions (the North China Plain, Yangtze River Delta, and Pearl River Delta). The network had been expanded to cover 1497 sites in 367 cities by 2015. Before January 2013, however, MEP reported daily primary pollutant and its air pollution index (API) for 84 major cities, which differ significantly from the 74 major cities mentioned above. Using each city's API and primary pollutant, it is possible to back-calculate the daily average concentration for the primary pollutant (PM_{10} for most days) (26). In view of this situation, we make use of the $PM_{2.5}$ concentrations in 2013 and 2015, as well as the API-derived PM_{10} concentrations in 2005 and 2010. Regarding the former, we only use the sites that are in operation in both 2013 and 2015 (473 sites in total) in order to evaluate the temporal trend. It should be noted that, while the target years are 2005, 2010, and 2015 in this study, we also perform simulations for 2013 to facilitate the comparison with surface $PM_{2.5}$ measurements. SI Appendix Table S4 shows the performance statistics for PM_{10} concentrations in 2005 and 2010 over the ECC region, while SI Appendix Table S5 presents the corresponding statistics for $PM_{2.5}$ concentrations in 2013 and 2015. While the model slightly underestimates the PM_{10}/$PM_{2.5}$ concentrations partly due to the exclusion of fugitive dust emissions, the performance statistics all meet the model performance criteria proposed by Boylan and Russell (23) except for the MFB in the summer of 2005. With regard to the temporal trend, the modeled and observed PM_{10} concentrations over ECC decreased by 6% and 7%, respectively, from 2005 to 2010. From 2013 to 2015, the decreasing rates of modeled and observed $PM_{2.5}$ concentrations are 24% and 23%, respectively. In summary, the simulated PM_{10}/$PM_{2.5}$ concentrations generally agree well with nationwide surface observations.

3. Uncertainty analysis

We assess uncertainties in IPWE and the health effects using 50000 Monte Carlo runs based on uncertainties of the input data, including household-fuel consumption, main cooking fuel distributions, HAP exposure levels, and the integrated exposure-response (IER) functions. The household fuel consumption is assumed to be uniformly distributed, with the variation intervals being ±20% for biomass and ±10% for other fuel types, following previous studies (27, 28). Among the three nationwide statistics/surveys of biomass consumption (shown in SI Appendix Fig. S1), the largest deviation from the “best estimate” is about 21%, supporting the uncertainty range for biomass assumed above. For coal consumption, considering the relatively large discrepancy between the official statistics and the survey data (SI Appendix Fig. S1), we increase the variation intervals from ±10% to ±30%. The consequent uncertainty in the AAP exposure simulated by the CMAQ/2D-VBS model is estimated by performing a number of sensitivity simulations with perturbed household fuel consumption. The uncertainties in activity data of non-household sources, the emission factors, as well as the model schemes are not considered because they are not supposed to be major factors affecting the contribution of household fuel use to changes in IPWE and health effects in the last decade, which is the focus of this study.

The populations using coal and biomass as main cooking fuels are also assumed to have a uniform distribution and the variation intervals are assumed to be 25% larger than those of the corresponding fuel consumption, since the cooking fuel-using populations in 2005/2015 are derived by integrating census data and fuel consumption statistics. The uncertainty in HAP exposure levels (i.e., $HAP_{j,k}$ in Eq. 2 of Methods) was estimated by Aunan et al. (29).

Regarding the health effects, Cohen et al. (30) provided 1000 sets of IER parameters for each health endpoint. In each of the 50000 Monte Carlo runs, a set of IER parameter is randomly chosen together with other randomly sampled inputs from the assumed probability distributions. The Monte Carlo simulation results constitute the probability distributions of IPWE and premature deaths, from which the 95% confidence intervals are derived.

We also discuss the influence of a couple of additional factors, including the relative toxicity of different $PM_{2.5}$ chemical components and the changes in stove technology over time, and show that they do not affect the major conclusion that household fuels are the leading contributor to the reduction of IPWE in China in the last decade (SI Appendix Section 4).

4. Supplementary information for the IPWE and health impact calculation methods

In the IPWE calculation method, we assume that ambient $PM_{2.5}$ pollution generally penetrates into the household (see Methods). This assumption ensures that the definition of AAP in this study is consistent with previous studies on AAP exposure (30-32), i.e., everyone within a given geographical unit is exposed to the prevailing AAP level. This assumption only affects the partitioning between AAP and HAP, and does not affect the total IPWE. The assumption is mostly true for rural populations whose homes are usually not well sealed (33). For urban households, however, previous measurements (34-38) indicate that the indoor-outdoor ratio of $PM_{2.5}$ concentrations for households without indoor sources ranges between 68-93%. Here we conduct a sensitivity analysis in which 80% and 95% of the ambient $PM_{2.5}$ concentrations penetrate into urban and rural households, respectively. Following this hypothesis, the fractional contribution of AAP to total IPWE in 2005 decreases to 27% from the original value of 31%. The contribution of household fuels to IPWE reduction during 2005-2015, i.e., 79 (51 to 113) μg/m^3 and 91% (87-94%), is very close to the estimates in this study (76 μg/m^3 and 90%).

For the calculation of HAP exposure, there has been no continuous nationwide survey of the populations using coal and biomass as their main cooking fuels ($P_{i,j,k}$ in Eq. 2 of Methods) during 2005-2015, but there are several continuous nationwide statistics/surveys of the consumption of household coal and biomass (see SI Appendix Section 1). The total exposure amount ($P_{i,j,k} \cdot HAP_{j,k}$ 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 (influence of this assumption is discussed in the next paragraph). In this study we apply the same HAP exposure levels ($HAP_{j,k}$ in Eq. 2) to all years because of insufficient measurements to derive $HAP_{j,k}$ as a function of time. As such, $P_{i,j,k}$ is proportional to the consumption of solid fuel in the given geographic unit and setting. For this reason, we combine the county-level solid fuel-using populations ($P_{i,j,k}$) in 2010 obtained from the National Population Census (39, 40) with provincial-level statistics of household coal and biomass consumption during 2005-2015 to derive county-level solid fuel-using populations during 2005-2015, as illustrated in SI Appendix Fig. S3. Note that changes in $HAP_{j,k}$ are affected by not only improvement of stove technology, but also the changes in fuel mixture and fraction of heating fuels. However, the latter factors do not affect the calculation results of $PM_{2.5}$ exposure from HAP since the resulting changes in $HAP_{j,k}$ and $P_{i,j,k}$ offset each other.

As stated above, we assume that the stove technology remained unchanged during 2005-2015. This may not be the case since the fraction of improved cookstoves could have increased during 2005-2015 driven by improved living standards and possibly a continued influence of the "Improved Stove Program" launched in the 1980s and 1990s (41). There were no such programs for heating stoves, however, until recently. The improvement of stove technology may result in a decrease in $HAP_{j,k}$. In this case, the fractional contribution of household fuels to the reduction of IPWE in the last decade could be even larger than our current estimate of 90% (86-93%). The possible changes in $HAP_{j,k}$ as a result of the stove technology improvement has been reflected in its error bar, which was subsequently used in the Monto Carlo uncertainty analysis.

An assumption that may affect the contribution of household fuels to human health is the relative toxicity of different $PM_{2.5}$ chemical components. In this study, we hypothesize that the health effects depend only on the inhaled amount of $PM_{2.5}$ and are independent of the chemical composition, which appears reasonable in view of the available quantitative epidemiological studies. However, some studies have reported that the carbonaceous aerosols could be significantly more toxic than other aerosol species (31, 42). Since carbonaceous aerosols (BC and OC) account for about 75% of the $PM_{2.5}$ emissions from household-fuel use while contributing only about 27% of $PM_{2.5}$ emissions from non-household sources, assuming carbonaceous aerosols being more toxic could result in an even larger relative contribution of household fuels to the magnitude and changes of premature deaths, as compared to the results in the present study. Nevertheless, this will not alter our key conclusion that household fuels were the leading contributor to the decrease in IPWE and associated mortality in 2005-2015.

5. Estimation of IPWE attributed to cooking and heating

As described in Methods, it is not appropriate to separately estimate the HAP exposures due to cooking and space heating based on measured exposure levels, because there were insufficient household measurements supporting this. Therefore, we split the HAP exposure due to cooking and heating based on the assumption that the HAP exposure is proportional to the solid-fuel consumption. The AAP exposure attributed to cooking/heating is quantified by designing a hypothetical scenario in which the household fuels used for cooking/heating are eliminated, and comparing it with the baseline scenario where all sources are included.

In the rural area, the fractions of solid fuels used for cooking and heating in each province are obtained from Tao et al. (12), which quantified the solid-fuel use in rural China from 1992 to 2012 through a national survey. The solid-fuel consumption after 2012 is linearly extrapolated from previous trend. In the urban area, the provincial fractions of solid fuels for cooking and heating are derived from the bottom-up model developed by the Tsinghua University Building Energy Research Center (Tsinghua-BERC) (7-11).

Chen et al. (43) estimated the household $PM_{2.5}$ exposure from space heating in rural China, using the difference between the indoor $PM_{2.5}$ concentrations in heating and non-heating seasons. They showed that about 24% of the household $PM_{2.5}$ exposure in rural China was attributed to space heating, and the fractional contribution remained stable (within ±1%) from 2005 to 2012. Based on our estimates, however, the fractional contributions of space heating increased from 23% in 2005 to 31% in 2015, because we accounted for the smaller decreasing rate in heating fuels than in cooking fuels during this period, as described in the main text. Note that Chen et al. (43) only considered indoor concentrations and not the general ambient air pollution due to household heating. We have adjusted our calculation to be directly comparable to the results of Chen et al. (43).

6. Contributors to the changes in premature mortality from 2005 to 2015

In the main text, we show the trends in $PM_{2.5}$-related premature deaths during 2005-2015, which are the combined effect of changes in IPWE, population, age distribution, and background mortality rate. We take into account all these changes in order to achieve the best possible estimates of the total $PM_{2.5}$-related mortality and the mortality due to household fuels. Here we attempt to isolate the impact of IPWE reduction through a sensitivity analysis in which the population, age distribution, and background mortality rate are kept constant as the 2005 levels. In this case, the $PM_{2.5}$-related mortality is estimated to decrease by 24% (18-30%) from 2005 to 2015, and household fuels contribute 81% (70-89%) of the total mortality reduction, which are similar to the calculation results when changes in all factors are considered (29% and 80%). Therefore, IPWE is the predominant contributor to mortality changes during 2005-2015 at the national level. Nevertheless, the change in population may play an important role in certain regions, such as some large urban centers with rapid increase in migrant population, as illustrated in Fig. 4 and described in the main text.

7. Benefits of replacing household solid fuels with clean fuels

In 2017, an action plan for clean heating (44, 45) was launched in northern China. The target provinces include Beijing, Tianjin, Hebei, Shanxi, Inner Mongolia, Liaoning, Jilin, Heilongjiang, Shandong, Shaanxi, Gansu, Ningxia, Xinjiang, Qinghai, and part of Henan, with a focus on Beijing-Tianjin-Hebei and the surrounding area. The overarching goal is to increase the fraction of clean heating in northern China to 70% by 2021, which means that about 55% of the existing household solid fuels for heating in these provinces shall be replaced with clean energy, such as natural gas, electricity, combined heat and power with extreme low emissions, solar and geothermal energy, and clean biofuels. While a thorough assessment and comparison of different options is beyond our scope, we aim to illustrate the potential environmental and health benefits that could be achieved through a state-of-the-art control option. For this purpose, we assume that half of the solid fuels are replaced by natural gas and the other half by electricity (50% electric resistance heating and 50% heat pump). As described in the main text, a successful implementation of this policy would reduce the IPWE by 9.7% (8.8-10.4%) in China. The reduction ratio of IPWE is relatively small because the current policy only focuses on household heating fuels (not cooking fuels or non-household sources) in northern China (not middle or southern China). It should be noted that this policy could have co-benefit on the 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 co-benefit, however, is difficult to quantify and hence not considered in the calculation. In addition, to evaluate the potential benefits of more stringent controls, we design another scenario in which all solid fuels used for cooking and heating in 2015 were thoroughly replaced with electricity and natural gas (50% each). Following the substitution, the consequent increases in consumption of clean fuels (electricity or natural gas) are calculated according to the assumption that the same amount of useful energy (fuel consumption multiplied by energy efficiency) for both cooking and heating are generated before and after substitution. For cooking, the energy efficiencies of coal stove, biomass stove, natural gas stove, and electric cooking are assumed to be 35%, 17%, 50%, and 90%, respectively (7, 10). With respect to heating, coal stove, biomass stove, natural gas heating, electric resistance heating, and heat pump are assumed to have energy efficiencies of 50%, 24%, 90%, 98%, and 250%, respectively (7, 11, 46). For the electricity substitution, we assume that the additional household electricity consumption in each province was generated within the same province, considering a transmission loss of 6.6%, the average rate in 2015 (47).

To compare the benefits of substitution by electricity or natural gas, we design two additional scenarios in which all solid fuels used for cooking and heating in 2015 were thoroughly substituted by electricity and natural gas, respectively. In these two scenarios, the fractional reductions in IPWE compared to 2015 would be 63% (57-68%) and 62% (57%-67%), respectively. The corresponding avoided premature deaths are 0.51 (0.40-0.64) million and 0.50 (0.39-0.64) million, respectively. The benefits are very similar because the exposure increase due to additional electricity or natural gas consumption (< 0.5 $\mu g/m^3$) are much smaller than the exposure decrease due to reduced solid fuels (60 $\mu g/m^3$). Therefore, the choice of natural gas or electricity should not be determined based on the health benefit, but rather based on economic cost, availability of resources, and feasibility of infrastructure construction, which merits further in-depth studies.

In the scenarios, the energy mix and shares of end-of-pipe control technologies of the added power plants are assumed to be same as the existing units in 2015. To evaluate the impact of the energy mix of the added power plants, we design a sensitivity scenario which assumes that all added power plants were coal-fired units. In this case, the electricity substitution was estimated to reduce 62% (57-67%) of the IPWE in 2015 and avoid 0.50 (0.39-0.64) million premature deaths, very similar to our current estimations, i.e., 63% (57-68%) and 0.51 (0.40-0.64) million. The small sensitivity of the environmental and health benefits to the energy mix is also explained by the fact the $PM_{2.5}$ exposure increase due to additional power plants is much smaller than the $PM_{2.5}$ exposure decrease due to the eliminated household solid fuels. Note that the environmental and health benefits could diminish if the end-of-pipe control measures of the added power plants were not as stringent as they were in 2015. We suggest this is very unlikely to happen considering the stringent emission standard for power plants in China.

8. Comparison with IPWE estimates using the HAP exposure levels from the GBD study

The HAP exposure levels of solid-fuel users (i.e., $HAP_{j,k}$ in Eq. 2 in Methods) are subject to relatively large uncertainties. In the GBD study (48), the parameters derived from in-situ measurements and a prediction model over India were applied to the whole world. The Indian average $PM_{2.5}$ exposures among all solid fuel-using households (which include the contribution from AAP) were estimated at 285 (95% confidence interval, 201-405) μg/m^3 for children, 337 (238-479) μg/m^3 for women and 204 (144-290) μg/m^3 for men (48). Applying these estimates (the contribution from AAP was subtracted to be consistent with this study), we estimate an IPWE in 2005 of 191 (155-248) μg/m^3, quite similar to the results of the current study (180 (146-219) μg/m^3). Also, applying the estimates of GBD (48), household fuels are calculated to lead to a reduction of 77 (50-119) μg/m^3 in IPWE from 2005 to 2015, representing 91% (87-94%) of the total IPWE reduction, which is also similar to the estimates in the current study (76 μg/m^3 and 90%).

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Tables and Figures

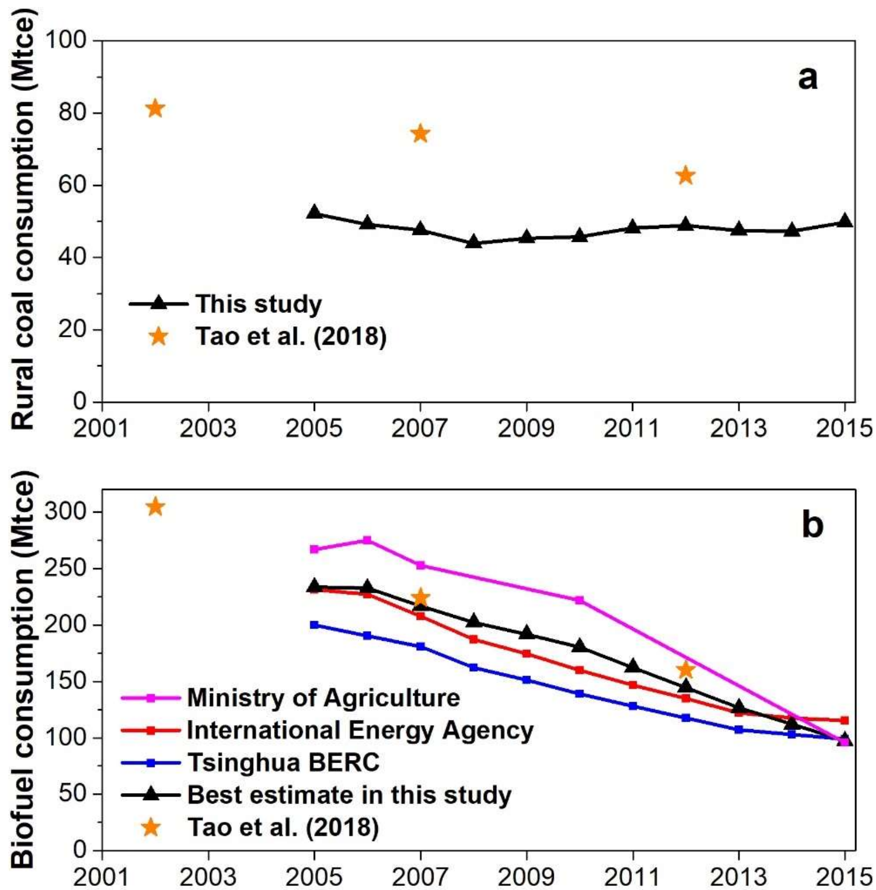


Figure S1. Household coal and biomass consumption from different nationwide statistics and surveys. The data sources are described in SI Appendix Section 1. The values of "best estimate in this study" in (b) are derived by taking the average of the data from the Ministry of Agriculture and Tsinghua-BERC, which are bottom-up statistical or survey data compiled by local institutes.

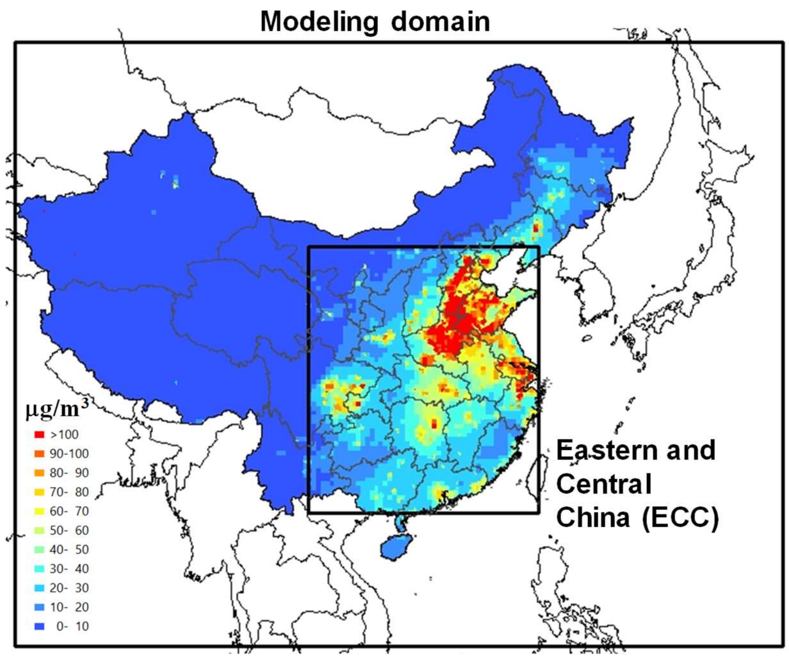


Figure S2. Modeling domain used in this study. The inner black box denotes the Eastern and Central China (ECC), which covers most of the populous areas in China characterized by substantial anthropogenic emissions and high ambient $PM_{2.5}$ concentrations. The colors represent ambient $PM_{2.5}$ concentrations in 2005, which are the same as Fig. 1c.

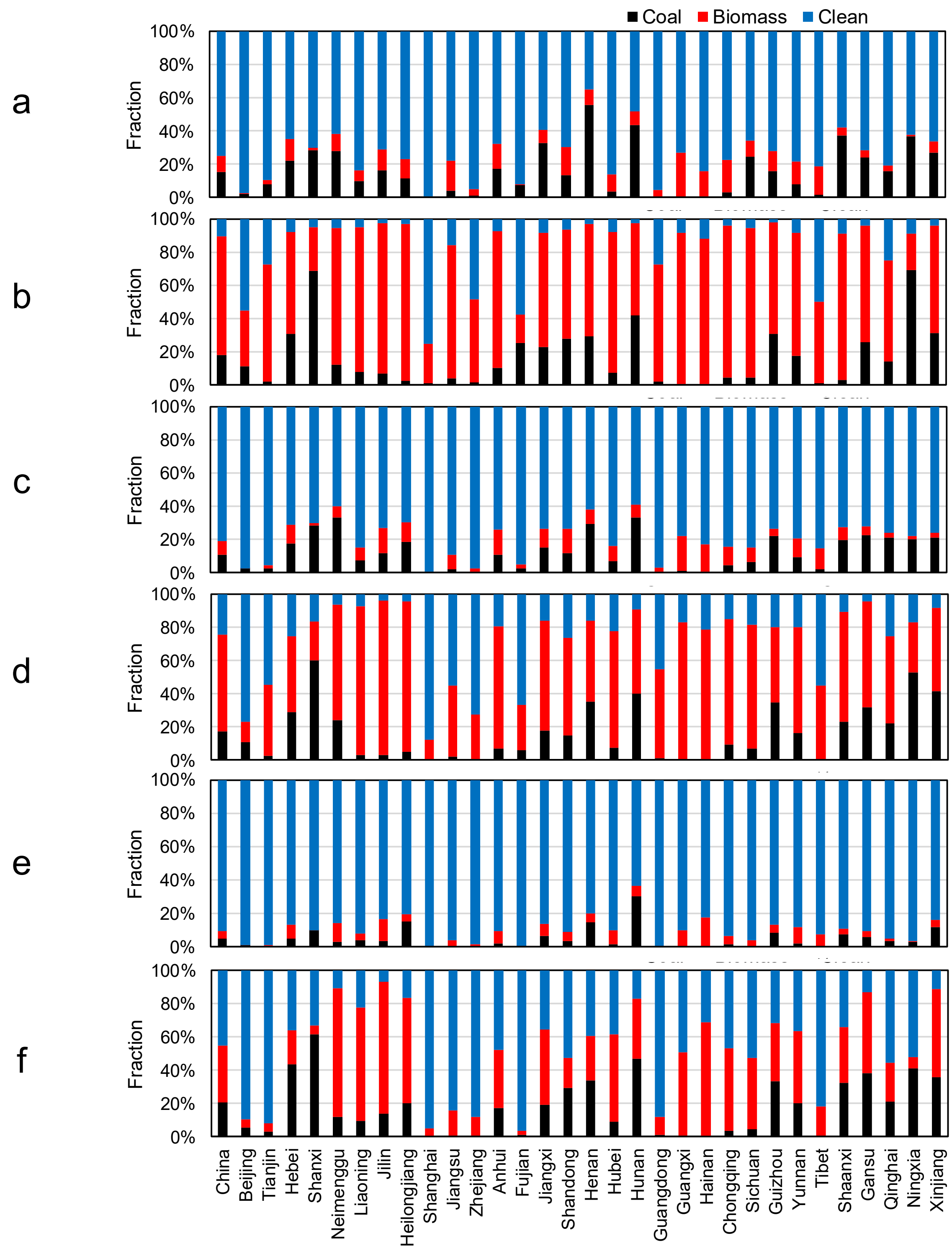
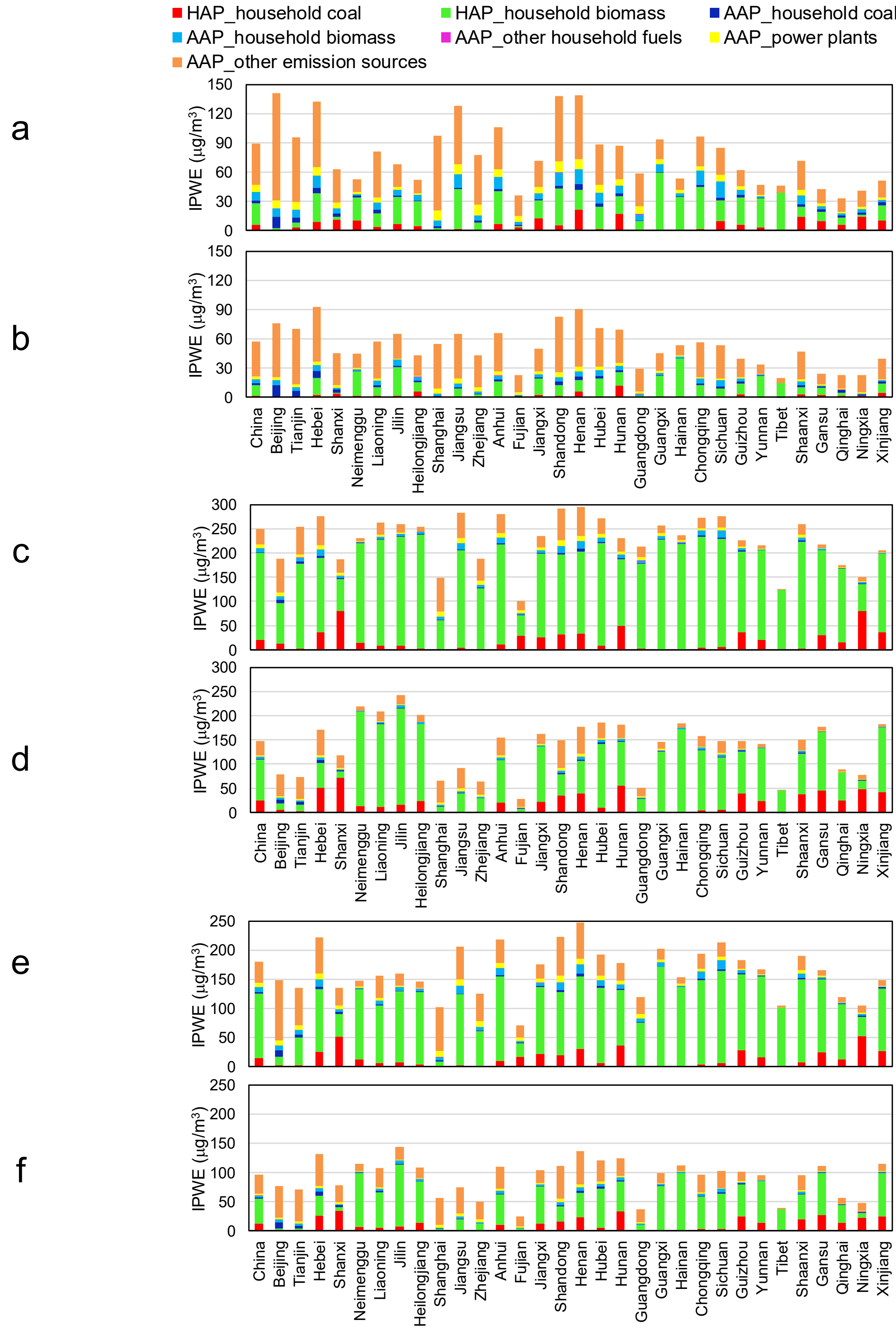


Figure S3. Share of major cooking fuels among urban and rural populations by province in 2005 and 2015: **a**, urban, 2005; **b**, rural, 2005; **c**, urban, 2010; **d**, rural, 2010; **e**, urban, 2015; **f**, rural, 2015. “Clean” refers to electricity and gaseous fuels (natural gas, liquified petroleum gas, coal gas, and biogas).



456 **Figure S4.** Contribution of individual sources to IPWE among urban (**a, b**), rural (**c, d**), and all (**e,**
457 **f**) populations in 2005 (**a, c, e**) and 2015 (**b, d, f**).

Table S1. Emissions of major air pollutants by sector in China during 2005-2015 (kt).

(a) SO_2

	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Power plants	15414	13703	11562	8446	6546	6272	6919	6914	6223	4886	4578
Industrial combustion	6580	7099	7508	8819	9266	9482	9728	9683	6997	4595	5439
Industrial process	4599	5090	5529	5184	5359	6216	6174	5993	6565	5625	3735
Cement	1246	1357	1404	1218	1120	1326	1433	1540	1704	1131	927
Steel	1222	1431	1744	1830	2002	2515	2278	2062	2286	1742	768
Residential & commercial	2403	2352	2289	2763	2977	3123	3390	3569	3379	3438	3598
Household coal	1269	1205	1173	1143	1159	1199	1245	1232	1099	1096	1128
Household biomass	82	82	76	71	67	63	57	51	44	39	34
Other household fuels	15	15	18	19	20	22	23	26	23	24	25
Transportation	499	484	469	460	443	434	381	321	268	224	167
On-road	272	243	215	186	158	130	108	86	64	42	20
Off-road	228	241	254	274	285	304	273	236	204	181	146
Biomass open burning	84	86	84	87	85	86	88	91	90	91	86
Total	29579	28814	27441	25759	24677	25613	26679	26571	23522	18859	17603

(b) NO_x

	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Power plants	7180	7829	8405	8238	8036	8156	8330	8026	7960	5563	4020
Industrial combustion	3133	3445	3673	4073	4195	4550	4484	4401	3372	2791	2312
Industrial process	2752	3102	3507	3857	4444	5043	5247	5446	5960	5038	4702
Cement	1088	1282	1506	1673	2001	2358	2519	2676	2922	2163	1934
Steel	274	324	399	423	466	592	563	535	546	603	555
Residential & commercial	966	948	892	933	945	943	965	959	882	851	852
Household coal	199	184	175	166	167	172	179	175	159	154	157
Household biomass	539	537	500	467	443	416	375	334	292	258	225
Other household fuels	5	5	6	6	7	7	8	9	8	8	9
Transportation	5940	6170	6546	6907	7763	8780	9255	9404	10162	10180	10204
On-road	4052	4177	4441	4666	5468	6382	6950	7268	7747	7793	7735
Off-road	1888	1992	2105	2241	2295	2398	2305	2136	2416	2387	2469
Biomass open burning	493	502	489	506	499	503	516	529	527	530	502
Total	20463	21996	23512	24514	25883	27974	28797	28765	28863	24953	22591

(c) PM_{10}

	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Power plants	1951	1842	1709	1424	1253	1261	1275	1123	1114	1055	1084
Industrial combustion	1518	1585	1586	1713	1692	1696	1791	1847	1292	1214	1337
Industrial process	8454	8484	8217	7606	6921	6856	6977	7068	7344	5421	3940
Cement	4944	4718	4200	3358	2999	2796	2897	2980	2985	1990	1264
Steel	958	1077	1222	1190	1262	1350	1336	1320	1397	1277	1206
Residential & commercial	4615	4555	4282	4302	4243	4121	4067	3857	3577	3361	3253
Household coal	666	618	586	556	560	576	599	586	532	518	527
Household biomass	3362	3348	3120	2911	2760	2596	2337	2080	1822	1611	1402
Other household fuels	6	7	8	8	9	10	10	11	10	11	11
Transportation	756	738	756	754	802	829	790	720	697	642	578
On-road	443	410	411	405	458	502	492	462	454	416	375
Off-road	313	328	344	349	344	327	298	257	244	225	203
Biomass open burning	1634	1664	1621	1677	1653	1666	1711	1756	1748	1758	1666

Total	18927	18869	18171	17476	16563	16428	16611	16370	15773	13451	11859

(d) $PM_{2.5}$

	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Power plants	1177	1114	1040	872	767	767	769	693	664	628	648
Industrial combustion	963	1018	1033	1131	1135	1160	1217	1274	870	807	888
Industrial process	5185	5252	5146	4791	4335	4326	4444	4545	4712	3495	2506
Cement	3152	3008	2679	2143	1911	1777	1835	1881	1867	1168	698
Steel	724	811	918	892	941	999	985	969	1029	926	872
Residential & commercial	3992	3942	3690	3557	3447	3312	3141	2911	2624	2413	2252
Household coal	518	481	456	432	435	448	466	456	414	403	410
Household biomass	3257	3244	3022	2820	2674	2514	2264	2015	1765	1561	1358
Other household fuels	6	7	8	8	9	10	10	11	10	11	11
Transportation	695	700	716	714	760	761	748	682	660	608	548
On-road	399	389	390	383	434	451	466	438	430	394	355
Off-road	296	311	326	331	326	310	282	244	231	214	193
Biomass open burning	1348	1372	1337	1383	1363	1374	1411	1448	1442	1450	1374
Total	13359	13398	12963	12449	11806	11701	11731	11553	10973	9401	8217

(e) BC

	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Power plants	46	41	37	28	19	14	15	10	8	8	9
Industrial combustion	157	164	165	179	176	176	181	186	123	108	118
Industrial process	477	515	537	629	502	512	520	527	536	393	207
Cement	11	11	11	10	10	10	11	11	11	6	5
Steel	20	23	27	26	30	32	33	35	38	38	37
Residential & commercial	871	851	798	770	752	732	710	667	603	563	537
Household coal	259	240	228	216	218	224	233	228	207	201	205
Household biomass	570	568	529	494	468	440	396	353	309	273	238
Other household fuels	1	1	1	1	1	1	1	1	1	1	1
Transportation	385	388	397	396	421	423	415	378	365	337	303
On-road	216	211	211	208	235	247	255	239	234	215	193
Off-road	169	177	186	188	186	177	161	139	132	122	110
Biomass open burning	54	55	54	56	55	56	57	59	58	59	56
Total	1991	2014	1989	2058	1924	1913	1899	1827	1694	1467	1231

(f) OC

	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Power plants	13	12	12	10	8	8	9	12	13	13	17
Industrial combustion	38	40	41	43	43	47	47	48	34	31	33
Industrial process	416	446	462	524	437	445	448	450	459	327	184
Cement	34	34	33	29	30	31	32	34	34	19	14
Steel	37	42	46	45	48	49	46	44	48	44	40
Residential & commercial	2009	1987	1856	1739	1662	1580	1452	1313	1158	1042	936
Household coal	207	192	182	173	174	179	186	182	166	161	164
Household biomass	1791	1784	1662	1551	1471	1383	1245	1108	971	858	747
Other household fuels	2	2	2	3	3	3	3	3	3	3	3
Transportation	130	131	134	134	142	142	140	128	124	114	103
On-road	77	75	75	74	84	86	89	84	83	75	68

Off-road	53	56	59	60	59	56	51	44	42	38	35
Biomass open burning	539	549	535	553	545	550	564	579	577	580	550
Total	3145	3165	3039	3003	2838	2771	2661	2528	2365	2106	1823

(g) NMVOC

	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Industrial combustion	69	76	83	87	93	107	113	119	119	125	129
Industrial process	4020	4629	5193	5434	5522	5912	6000	6054	7048	6858	6037
Residential & commercial	4628	4559	4204	4003	3845	3664	3421	3142	2798	2544	2344
Household coal	476	441	419	397	400	411	428	419	380	369	375
Household biomass	3913	3875	3540	3282	3091	2887	2578	2274	1974	1728	1489
Other household fuels	40	41	46	46	47	48	54	55	48	47	52
Transportation	5199	5454	5914	5888	5758	5403	5696	6027	6155	6403	6677
On-road	4359	4568	4987	4940	4807	4451	4764	5142	5286	5555	5843
Off-road	841	885	928	948	951	952	932	885	869	848	834
Biomass open burning	1135	1156	1165	1148	1148	1157	1188	1219	1214	1147	1030
Solvent use	3697	4349	4935	5219	6249	7480	7818	8155	8155	8599	9043
Other sources	193	207	228	255	255	256	307	358	387	394	432
Total	18941	20428	21722	22034	22870	23979	24543	25075	25877	26070	25692

(h) NH_3

	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Livestock	5882	5981	4981	5194	5405	5538	5508	5717	5803	5928	5657
fertilizer application	3682	3772	3746	3728	3800	3826	3852	3871	3843	3815	3769
Industrial process	239	247	266	272	282	275	270	301	302	288	308
Other sources	621	618	617	624	635	642	657	666	680	684	698
Total	10425	10618	9610	9819	10122	10281	10287	10555	10627	10714	10432

Table S2. Performance statistics for the comparison between simulated and observed meteorological variables.

	Wind speed						Temperature						Humidity					
	Mean OBS	Mean SIM	MB	GE	RMSE	IOA	Mean OBS	Mean SIM	MB	GE	RMSE	IOA	Mean OBS	Mean SIM	MB	GE	RMSE	IOA
Unit	m/s	m/s	m/s	m/s	m/s		K	K	K	K	K		g/kg	g/kg	g/kg	g/kg	g/kg	
Benchmark			≤± 0.5	≤2	≤2	≥ 0.6			≤ ± 0.5	≤2		≥ 0.8			≤±1	≤2		≥ 0.6
Spr-05[a]	3.43	3.23	-0.20	1.32	1.82	0.77	283.3	283.0	-0.32	1.62	2.25	0.98	4.62	4.61	-0.01	0.78	1.06	0.98
Sum-05	2.74	2.57	-0.17	1.16	1.64	0.71	296.0	295.8	-0.20	1.53	2.07	0.97	11.61	10.77	-0.84	1.63	2.12	0.95
Fal-05	2.70	2.65	-0.05	1.12	1.57	0.76	283.4	283.0	-0.39	1.50	2.00	0.98	5.68	5.42	-0.26	0.85	1.17	0.97
Win-05	2.73	2.77	0.04	1.19	1.65	0.74	264.8	264.3	-0.44	1.91	2.73	0.98	1.89	1.93	0.05	0.32	0.50	0.98
Spr-10	3.08	3.02	-0.06	1.22	1.72	0.78	285.8	285.3	-0.43	1.68	2.51	0.98	6.55	6.58	0.03	0.92	1.28	0.98
Sum-10	2.50	2.38	-0.12	1.06	1.51	0.74	297.2	296.8	-0.42	1.61	2.31	0.97	13.47	12.91	-0.56	1.61	2.14	0.96
Fal-10	2.52	2.53	0.01	1.05	1.48	0.80	287.1	286.6	-0.48	1.60	2.30	0.98	8.07	7.73	-0.34	1.07	1.46	0.97
Win-10	2.78	2.86	0.08	1.18	1.67	0.78	274.4	273.8	**-0.56**[b]	1.86	2.78	0.99	3.57	3.68	0.11	0.59	0.87	0.98
Spr-15	2.97	2.88	-0.09	1.21	1.67	0.78	287.4	286.8	**-0.54**	1.76	2.67	0.97	7.32	7.17	-0.15	1.09	1.55	0.97
Sum-15	2.55	2.42	-0.13	1.10	1.55	0.75	296.6	296.2	-0.41	1.63	2.35	0.97	14.13	13.40	-0.73	1.73	2.38	0.95
Fal-15	2.47	2.46	-0.01	1.06	1.48	0.77	287.1	286.7	-0.40	1.59	2.31	0.98	9.06	8.67	-0.38	1.11	1.59	0.97
Win-15	2.56	2.64	0.07	1.13	1.58	0.77	275.2	274.6	**-0.63**	1.86	2.82	0.98	4.01	4.00	-0.02	0.61	0.90	0.98

[a] Spr – spring, Sum – summer, Fal – fall, Win – winter.

[b] The values exceeding the benchmark range are highlighted in bold.

Table S3. Performance statistics for the comparison between simulated AOD/NO_2 column and satellite retrievals over the ECC region (units are 10^{15} molecules/cm^2 for NO_2 vertical column density and Dobson unit for AOD, except those noted in the table).

	AOD								NO_2 column							
	Mean OBS	Mean SIM	NMB (%)	NME (%)	MFB (%)	MFE (%)	RMSE	R (no unit)	Mean OBS	Mean SIM	NMB (%)	NME (%)	MFB (%)	MFE (%)	RMSE	R (no unit)
Avg-05[a]	0.53	0.54	1	22	-9	27	0.16	0.83	3.92	4.19	7	31	-13	35	2.28	0.91
Spr-05	0.60	0.58	-3	27	-15	37	0.21	0.78	3.47	3.23	-7	34	-33	47	2.03	0.89
Sum-05	0.59	0.40	-32	33	-45	46	0.22	0.86	2.68	2.09	-22	43	-53	62	1.85	0.82
Fal-05	0.48	0.47	-2	20	-7	24	0.12	0.85	3.80	4.27	12	36	-8	36	2.82	0.89
Win-05	0.49	0.78	62	71	43	54	0.42	0.63	5.79	7.19	24	40	21	44	3.72	0.90
Avg-10	0.53	0.51	-5	22	-13	28	0.16	0.79	5.10	6.12	20	35	6	35	3.48	0.89
Spr-10	0.61	0.54	-12	28	-23	36	0.23	0.71	4.10	5.30	29	49	17	53	3.63	0.84
Sum-10	0.62	0.41	-33	37	-44	47	0.28	0.81	2.95	3.03	3	48	-19	47	3.05	0.73
Fal-10	0.44	0.40	-10	24	-12	29	0.14	0.76	5.50	6.15	12	35	1	34	3.99	0.86
Win-10	0.46	0.72	55	66	41	53	0.40	0.63	7.88	9.99	27	38	20	41	4.90	0.91
Avg-15	0.44	0.44	0	31	-8	34	0.19	0.62	4.30	4.38	2	34	-20	39	2.73	0.85
Spr-15	0.54	0.55	0	36	-12	40	0.26	0.63	3.44	3.47	1	42	-22	48	2.62	0.80
Sum-15	0.48	0.30	-37	42	-49	54	0.25	0.67	2.75	2.25	-18	49	-48	62	2.22	0.69
Fal-15	0.36	0.35	-3	32	-3	35	0.16	0.63	4.32	4.84	12	42	-8	42	3.41	0.82
Win-15	0.41	0.62	50	63	38	56	0.36	0.52	6.71	6.94	4	32	-9	39	3.76	0.88

[a] Avg – annual average, Spr – spring, Sum – summer, Fal – fall, Win – winter.

Table S4. Performance statistics for the comparison between simulated and observed PM_{10} concentrations in major cities over the Eastern and Central China (ECC).

Variables[a]	2005					2010					Model performance criteria
	Avg	Spr	Sum	Fal	Win	Avg	Spr	Sum	Fal	Win	
Mean SIM (μg/m^3)	66.2	79.2	41.7	66.4	77.3	62.4	60.5	42.0	66.1	81.1	
Mean OBS (μg/m^3)	96.1	102.5	75.5	98.6	107.9	89.5	91.5	69.1	92.3	104.9	
NMB (%)	-31	-23	-45	-33	-28	-30	-34	-39	-28	-23	
NME (%)	49	50	51	42	49	46	49	52	43	40	
MFB (%)	-48	-44	**-69**[b]	-42	-39	-47	-53	-58	-40	-37	±60%
MFE (%)	63	64	71	54	64	60	64	68	56	52	±75%

[a] Avg – annual average, Spr – spring, Sum – summer, Fal – fall, Win – winter.

[b] The values exceeding the model performance criteria are highlighted in bold.

Table S5. Performance statistics for the comparison between simulated and observed $PM_{2.5}$ concentrations in major cities over the ECC region in 2013 and 2015.

Variables[a]	2013					2015					Model performance criteria
	Avg	Spr	Sum	Fal	Win	Avg	Spr	Sum	Fal	Win	
Mean SIM (μg/m^3)	61.9	58.6	38.8	54.3	95.1	46.7	45.0	30.6	40.4	70.9	
Mean OBS (μg/m^3)	74.1	66.2	46.7	69.8	113.8	56.5	51.4	39.7	49.8	85.2	
NMB (%)	-16	-12	-17	-22	-16	-17	-13	-23	-19	-17	
NME (%)	35	36	38	32	35	34	38	39	34	30	
MFB (%)	-21	-21	-21	-26	-17	-25	-24	-32	-24	-20	±60%
MFE (%)	40	42	41	37	40	41	44	45	39	35	±75%

[a] Avg – annual average, Spr – spring, Sum – summer, Fal – fall, Win – winter.