1  Supplementary Information for

2  "Change in household fuels dominates the decrease in PM$_{2.5}$

3  exposure and premature mortality in China in 2005-2015"

4

5  **1. Development of emission inventory**

6     We develop an up-to-date emission inventory of SO$_2$, NOx, PM$_{10}$, PM$_{2.5}$, BC, OC, NMVOC,

7  and NH$_3$ in China for 2005-2015 using the "emission factor method" based on the activity data

8  (energy consumption, industrial product yields, etc.), technology-based uncontrolled emission

9  factors, and penetrations of control technologies. The methodology and data sources to develop

10  the emission inventory were detailed in our previous papers (1-5), in which the emissions as of

11  2012 were calculated. Here we update the emission inventory till 2015, drawing upon the latest

12  information on energy consumption, technology distribution, and control measures, from a variety

13  of sources including statistical data, government bulletins, industrial associations, technological

14  reports, and research surveys. As a focus of this study, we obtain the consumption of all fuels

15  except biomass in the residential and commercial sectors in both urban and rural areas from the

16  Energy Statistical Yearbook of China (6). All fuel consumption in the residential sector in rural

17  areas is assumed to occur in the households, while in urban areas, the fraction of household-fuel

18  use in total residential and commercial energy consumption is achieved from a detailed bottom-up

19  model developed by the Tsinghua University Building Energy Research Center (Tsinghua-BERC)

20  (7-11). SI Appendix Fig. S1a compares the rural coal consumption used in this study (i.e., official

21  statistics) with the estimates from a representative national survey reported by Tao et al. (12),

22  which collected the fuel use data for 2012 as well as recalled historical data for 2002 and 2007.

23  The considerable discrepancy between these two data sources indicates a relatively large

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24  uncertainty in rural coal consumption, which is considered in the Monto Carlo uncertainty analysis

25  described in SI Appendix Section 3. In addition, we conducted a sensitivity analysis in which the

26  IPWE in 2005-2015 and the contribution from household fuels were calculated using the rural coal

27  consumption in Tao et al. (2018). In this case, the IPWE in 2005 is 3.5% larger than our current

28  estimate. Also, household fuels are estimated to lead to a reduction of 81 μg/m$^3$ in IPWE from

29  2005 to 2015, representing 91% of the total IPWE reduction, which is also similar to the estimates

30  in the current study (76 μg/m$^3$ and 90%). Therefore, the discrepancy in rural coal consumption

31  does not change the conclusion of this study. For biomass consumption, there were no published

32  official statistics after 2007. We obtain the statistics compiled by the Ministry of Agriculture of

33  China (the data as of 2007 were published in *China Energy Statistical Yearbook* (13) and those

34  after 2007 are unpublished), nationwide survey results from the Tsinghua-BERC (7-11), and the

35  data from International Energy Agency (14), as summarized in SI Appendix Fig. S1. All these

36  three datasets reveal a consistent trend, namely the biomass consumption in China decreased by

37  over 50% during 2005-2015. In this study, we take the average of the former two datasets (which

38  are bottom-up statistical or survey data by local institutes) as our "best estimate", and account for

39  the discrepancies among different datasets in the uncertainty analysis (SI Appendix Section 3). SI

40  Appendix Fig. S1b shows that our "best estimate" agrees well with the survey data by Tao et al.

41  (12) for years of 2002, 2007, and 2012. The household fuel use and sectoral emissions of major

42  air pollutants are summarized in Fig. 2 and SI Appendix Table S1.

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

44  We simulate the ambient concentrations of primary and secondary PM$_{2.5}$ in China using the

45  CMAQ/2D-VBS model, which was developed by incorporating the 2D-VBS model framework

46  into the default CMAQ model (15). The CMAQ/2D-VBS model explicitly simulates the aging of

47  primary organic aerosol (POA), photo-oxidation of intermediate-volatility organic compounds

48  (IVOCs) and multi-generational aging of secondary organic aerosol (SOA), thus improving the

49  simulation results of SOA. Here we use the "High-Yield VBS" configuration documented in Zhao

50  et al. (15), which agrees best with surface organic aerosol (OA) and SOA observational data. We

51  apply the CMAQ/2D-VBS model to a domain covering China and its surrounding areas (see SI

52  Appendix Fig. S2) with a grid resolution of 36 km×36 km. The Weather Research and Forecasting

53  Model (WRF, version 3.7) is used to generate the meteorological fields. The physical and chemical

54  options, vertical resolution, initial and boundary conditions of WRF and CMAQ/2D-VBS, as well

55  as the spatial allocation of anthropogenic emissions and calculation of biogenic emissions are the

56  same as Zhao et al. (16). The simulation periods cover 2005, 2010, and 2015, as well as 2013 for

57  evaluation purpose.

58  We compare the meteorological parameters simulated by WRFv3.7 with observational data

59  obtained from the National Climatic Data Center (NCDC), where hourly or 3-h observations are

60  available for about 380 sites in the domain. The variables of interest include the temperature at

61  2 m, wind speed at 10 m, and humidity at 2 m. The statistical indices used include the mean

62  observation (Mean OBS), mean simulation (Mean SIM), mean bias (MB), gross error (GE), root

63  mean square error (RMSE), and index of agreement (IOA), the definitions of which are provided

64  in Emery et al. (17). SI Appendix Table S2 summarizes the model performance statistics as well

65  as the benchmarks suggested by Emery et al. (17). Obviously almost all indices fall within the

66  benchmark range except that the MB of temperature slightly exceeds the range in a couple of cases.

67  The occasional slight exceedance is considered to be reasonable since the benchmarks were

68  derived based on performance statistics of meteorological simulations mostly at resolutions of

69  12 km or 4 km, which generally give more accurate predictions than those at 36 km resolution.

70  Therefore, the statistics indicate an overall decent performance of meteorological predictions.

71  In addition, we compare the simulated aerosol optical depth (AOD) and $NO_2$ vertical column

72  density with retrievals from the Moderate Resolution Imaging Spectroradiometer (MODIS)

73  onboard the Aqua satellite (http://ladsweb.nascom.nasa.gov/data/search.html) and Ozone

74  Monitoring Instrument (OMI) onboard the Aura satellite

75  (http://www.temis.nl/airpollution/no2.html), respectively. Only model outputs that are closest to

76  the satellite overpassing time (13:00-14:00 for both MODIS/Aqua and OMI/Aura, Beijing time)

77  are used in comparison to avoid sampling errors due to diurnal cycles. The conversion of vertically

78  resolved aerosol mass concentrations to AOD follows Heald (18) and Martin and Heald (19). SI

79  Appendix Table S3 summarizes the model performance statistics over the Eastern and Central

80  China (ECC, see SI Appendix Fig. S2). The ECC region covers most of the populous areas in

81  China characterized by substantial anthropogenic emissions and high ambient $PM_{2.5}$

82  concentrations. In contrast, the remaining parts of China are significantly affected by natural

83  sources (e.g., wind-blown dust) and foreign emissions, which are not the focus of this study. The

84  statistical indices we apply are slightly different from those applied to the evaluation of

85  meteorological predictions, so as to facilitate inter-study comparison with previous studies (e.g.,

86  Zhang et al. (20) and Zhao et al. (21)). These indices include Mean OBS, Mean SIM, normalized

87  mean bias (NMB), normalized mean error (NME), mean fractional bias (MFB), mean fractional

88  error (MFE), RMSE, and correlation coefficient (R), as documented in Yu et al. (22) and Boylan

89  and Russell (23). The simulated $NO_2$ column agrees fairly well with OMI retrievals in all seasons;

90  the NMBs and correlation coefficients are all $\leq \pm 29\%$ and $\geq 0.69$, and are $\leq \pm 20\%$ and $\geq 0.80$ in

91  most cases. There is also a good agreement between simulated and MODIS-retrieved annual mean

92    AOD, with NMBs within ±5%, whilst the model tends to overestimate AOD in winter and

93    underestimate AOD in summer. This is a common issue in previous CMAQ simulation studies in

94    China (3, 24, 25) and is yet to be addressed in future investigations. With respect to the inter-

95    annual trend which is the focus of the current study, the retrieved and simulated AOD show 17%

96    and 18% decrease from 2005 to 2015, respectively, indicating a good agreement. Both OMI

97    retrievals and CMAQ/2D-VBS simulations reveal that $NO_2$ column increased dramatically from

98    2005 to 2010, followed by a significant decline from 2010 to 2015. The overall percentage changes

99    from 2005 to 2015 are 10% and 5%, respectively.

100       For surface $PM_{2.5}$ concentrations, there were no nationwide continuous observations during the

101   simulation period since the observational network changed substantially between 2012 and 2013.

102   Since   January   2013,   the   Ministry   of   Environmental   Protection   of   China   (MEP,

103   http://datacenter.mep.gov.cn/) provides hourly $PM_{2.5}$ concentrations at 496 sites located in 74

104   major cities in China, including capital cities of all provinces and prefecture-level cities in three

105   metropolitan regions (the North China Plain, Yangtze River Delta, and Pearl River Delta). The

106   network had been expanded to cover 1497 sites in 367 cities by 2015. Before January 2013,

107   however, MEP reported daily primary pollutant and its air pollution index (API) for 84 major cities,

108   which differ significantly from the 74 major cities mentioned above. Using each city's API and

109   primary pollutant, it is possible to back-calculate the daily average concentration for the primary

110   pollutant ($PM_{10}$ for most days) (26). In view of this situation, we make use of the $PM_{2.5}$

111   concentrations in 2013 and 2015, as well as the API-derived $PM_{10}$ concentrations in 2005 and

112   2010. Regarding the former, we only use the sites that are in operation in both 2013 and 2015 (473

113   sites in total) in order to evaluate the temporal trend. It should be noted that, while the target years

114   are 2005, 2010, and 2015 in this study, we also perform simulations for 2013 to facilitate the

115    comparison with surface $PM_{2.5}$ measurements. SI Appendix Table S4 shows the performance

116    statistics for $PM_{10}$ concentrations in 2005 and 2010 over the ECC region, while SI Appendix Table

117    S5 presents the corresponding statistics for $PM_{2.5}$ concentrations in 2013 and 2015. While the

118    model slightly underestimates the $PM_{10}$/$PM_{2.5}$ concentrations partly due to the exclusion of fugitive

119    dust emissions, the performance statistics all meet the model performance criteria proposed by

120    Boylan and Russell (23) except for the MFB in the summer of 2005. With regard to the temporal

121    trend, the modeled and observed $PM_{10}$ concentrations over ECC decreased by 6% and 7%,

122    respectively, from 2005 to 2010. From 2013 to 2015, the decreasing rates of modeled and observed

123    $PM_{2.5}$ concentrations are 24% and 23%, respectively. In summary, the simulated $PM_{10}$/$PM_{2.5}$

124    concentrations generally agree well with nationwide surface observations.

125    **3. Uncertainty analysis**

126        We assess uncertainties in IPWE and the health effects using 50000 Monte Carlo runs based

127    on uncertainties of the input data, including household-fuel consumption, main cooking fuel

128    distributions, HAP exposure levels, and the integrated exposure-response (IER) functions. The

129    household fuel consumption is assumed to be uniformly distributed, with the variation intervals

130    being ±20% for biomass and ±10% for other fuel types, following previous studies (27, 28).

131    Among the three nationwide statistics/surveys of biomass consumption (shown in SI Appendix

132    Fig. S1), the largest deviation from the "best estimate" is about 21%, supporting the uncertainty

133    range for biomass assumed above. For coal consumption, considering the relatively large

134    discrepancy between the official statistics and the survey data (SI Appendix Fig. S1), we increase

135    the variation intervals from ±10% to ±30%. The consequent uncertainty in the AAP exposure

136    simulated by the CMAQ/2D-VBS model is estimated by performing a number of sensitivity

137    simulations with perturbed household fuel consumption. The uncertainties in activity data of non-

138    household sources, the emission factors, as well as the model schemes are not considered because

139    they are not supposed to be major factors affecting the contribution of household fuel use to

140    changes in IPWE and health effects in the last decade, which is the focus of this study.

141        The populations using coal and biomass as main cooking fuels are also assumed to have a

142    uniform distribution and the variation intervals are assumed to be 25% larger than those of the

143    corresponding fuel consumption, since the cooking fuel-using populations in 2005/2015 are

144    derived by integrating census data and fuel consumption statistics. The uncertainty in HAP

145    exposure levels (i.e., $HAP_{j,k}$ in Eq. 2 of Methods) was estimated by Aunan et al. (29).

146        Regarding the health effects, Cohen et al. (30) provided 1000 sets of IER parameters for each

147    health endpoint. In each of the 50000 Monte Carlo runs, a set of IER parameter is randomly chosen

148    together with other randomly sampled inputs from the assumed probability distributions. The

149    Monte Carlo simulation results constitute the probability distributions of IPWE and premature

150    deaths, from which the 95% confidence intervals are derived.

151        We also discuss the influence of a couple of additional factors, including the relative toxicity

152    of different $PM_{2.5}$ chemical components and the changes in stove technology over time, and show

153    that they do not affect the major conclusion that household fuels are the leading contributor to the

154    reduction of IPWE in China in the last decade (SI Appendix Section 4).

155    **4. Supplementary information for the IPWE and health impact calculation**

156    **methods**

157        In the IPWE calculation method, we assume that ambient $PM_{2.5}$ pollution generally

158    penetrates into the household (see Methods). This assumption ensures that the definition of AAP

159    in this study is consistent with previous studies on AAP exposure (30-32), i.e., everyone within a

160    given geographical unit is exposed to the prevailing AAP level. This assumption only affects the

161    partitioning between AAP and HAP, and does not affect the total IPWE. The assumption is mostly

162    true for rural populations whose homes are usually not well sealed (33). For urban households,

163    however, previous measurements (34-38) indicate that the indoor-outdoor ratio of $PM_{2.5}$

164    concentrations for households without indoor sources ranges between 68-93%. Here we conduct a

165    sensitivity analysis in which 80% and 95% of the ambient $PM_{2.5}$ concentrations penetrate into

166    urban and rural households, respectively. Following this hypothesis, the fractional contribution of

167    AAP to total IPWE in 2005 decreases to 27% from the original value of 31%. The contribution of

168    household fuels to IPWE reduction during 2005-2015, i.e., 79 (51 to 113) $\mu g/m^3$ and 91% (87-

169    94%), is very close to the estimates in this study (76 $\mu g/m^3$ and 90%).

170       For the calculation of HAP exposure, there has been no continuous nationwide survey of the

171    populations using coal and biomass as their main cooking fuels ($P_{i,j,k}$ in Eq. 2 of Methods) during

172    2005-2015, but there are several continuous nationwide statistics/surveys of the consumption of

173    household coal and biomass (see SI Appendix Section 1). The total exposure amount ($P_{i,j,k} \cdot$

174    $HAP_{j,k}$ in Eq. 2) for a specific geographic unit, setting (urban or rural), and solid-fuel type is

175    proportional to the solid-fuel consumption, under the assumption that the stove technology remains

176    unchanged over time (influence of this assumption is discussed in the next paragraph). In this study

177    we apply the same HAP exposure levels ($HAP_{j,k}$ in Eq. 2) to all years because of insufficient

178    measurements to derive $HAP_{j,k}$ as a function of time. As such, $P_{i,j,k}$ is proportional to the

179    consumption of solid fuel in the given geographic unit and setting. For this reason, we combine

180    the county-level solid fuel-using populations ($P_{i,j,k}$) in 2010 obtained from the National Population

181    Census (39, 40) with provincial-level statistics of household coal and biomass consumption during

182    2005-2015 to derive county-level solid fuel-using populations during 2005-2015, as illustrated in

183    SI Appendix Fig. S3. Note that changes in $HAP_{j,k}$ are affected by not only improvement of stove

8

184  technology, but also the changes in fuel mixture and fraction of heating fuels. However, the latter

185  factors do not affect the calculation results of PM$_{2.5}$ exposure from HAP since the resulting changes

186  in $HAP_{j,k}$ and $P_{i,j,k}$ offset each other.

187      As stated above, we assume that the stove technology remained unchanged during 2005-

188  2015. This may not be the case since the fraction of improved cookstoves could have increased

189  during 2005-2015 driven by improved living standards and possibly a continued influence of the

190  "Improved Stove Program" launched in the 1980s and 1990s (41). There were no such programs

191  for heating stoves, however, until recently. The improvement of stove technology may result in a

192  decrease in $HAP_{j,k}$. In this case, the fractional contribution of household fuels to the reduction of

193  IPWE in the last decade could be even larger than our current estimate of 90% (86-93%). The

194  possible changes in $HAP_{j,k}$ as a result of the stove technology improvement has been reflected in

195  its error bar, which was subsequently used in the Monto Carlo uncertainty analysis.

196      An assumption that may affect the contribution of household fuels to human health is the

197  relative toxicity of different PM$_{2.5}$ chemical components. In this study, we hypothesize that the

198  health effects depend only on the inhaled amount of PM$_{2.5}$ and are independent of the chemical

199  composition, which appears reasonable in view of the available quantitative epidemiological

200  studies. However, some studies have reported that the carbonaceous aerosols could be significantly

201  more toxic than other aerosol species (31, 42). Since carbonaceous aerosols (BC and OC) account

202  for about 75% of the PM$_{2.5}$ emissions from household-fuel use while contributing only about 27%

203  of PM$_{2.5}$ emissions from non-household sources, assuming carbonaceous aerosols being more toxic

204  could result in an even larger relative contribution of household fuels to the magnitude and changes

205  of premature deaths, as compared to the results in the present study. Nevertheless, this will not

206　alter our key conclusion that household fuels were the leading contributor to the decrease in IPWE

207　and associated mortality in 2005-2015.

208　**5. Estimation of IPWE attributed to cooking and heating**

209　　　As described in Methods, it is not appropriate to separately estimate the HAP exposures due

210　to cooking and space heating based on measured exposure levels, because there were insufficient

211　household measurements supporting this. Therefore, we split the HAP exposure due to cooking

212　and heating based on the assumption that the HAP exposure is proportional to the solid-fuel

213　consumption. The AAP exposure attributed to cooking/heating is quantified by designing a

214　hypothetical scenario in which the household fuels used for cooking/heating are eliminated, and

215　comparing it with the baseline scenario where all sources are included.

216　　　In the rural area, the fractions of solid fuels used for cooking and heating in each province

217　are obtained from Tao et al. (12), which quantified the solid-fuel use in rural China from 1992 to

218　2012 through a national survey. The solid-fuel consumption after 2012 is linearly extrapolated

219　from previous trend. In the urban area, the provincial fractions of solid fuels for cooking and

220　heating are derived from the bottom-up model developed by the Tsinghua University Building

221　Energy Research Center (Tsinghua-BERC) (7-11).

222　　　Chen et al. (43) estimated the household $PM_{2.5}$ exposure from space heating in rural China,

223　using the difference between the indoor $PM_{2.5}$ concentrations in heating and non-heating seasons.

224　They showed that about 24% of the household $PM_{2.5}$ exposure in rural China was attributed to

225　space heating, and the fractional contribution remained stable (within ±1%) from 2005 to 2012.

226　Based on our estimates, however, the fractional contributions of space heating increased from 23%

227　in 2005 to 31% in 2015, because we accounted for the smaller decreasing rate in heating fuels than

228　in cooking fuels during this period, as described in the main text. Note that Chen et al. (43) only

229    considered indoor concentrations and not the general ambient air pollution due to household

230    heating. We have adjusted our calculation to be directly comparable to the results of Chen et al.

231    (43).

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

233        In the main text, we show the trends in $PM_{2.5}$-related premature deaths during 2005-2015,

234    which are the combined effect of changes in IPWE, population, age distribution, and background

235    mortality rate. We take into account all these changes in order to achieve the best possible estimates

236    of the total $PM_{2.5}$-related mortality and the mortality due to household fuels. Here we attempt to

237    isolate the impact of IPWE reduction through a sensitivity analysis in which the population, age

238    distribution, and background mortality rate are kept constant as the 2005 levels. In this case, the

239    $PM_{2.5}$-related mortality is estimated to decrease by 24% (18-30%) from 2005 to 2015, and

240    household fuels contribute 81% (70-89%) of the total mortality reduction, which are similar to the

241    calculation results when changes in all factors are considered (29% and 80%). Therefore, IPWE is

242    the predominant contributor to mortality changes during 2005-2015 at the national level.

243    Nevertheless, the change in population may play an important role in certain regions, such as some

244    large urban centers with rapid increase in migrant population, as illustrated in Fig. 4 and described

245    in the main text.

**7. Benefits of replacing household solid fuels with clean fuels**

247        In 2017, an action plan for clean heating (44, 45) was launched in northern China. The target

248    provinces include Beijing, Tianjin, Hebei, Shanxi, Inner Mongolia, Liaoning, Jilin, Heilongjiang,

249    Shandong, Shaanxi, Gansu, Ningxia, Xinjiang, Qinghai, and part of Henan, with a focus on

250    Beijing-Tianjin-Hebei and the surrounding area. The overarching goal is to increase the fraction

251    of clean heating in northern China to 70% by 2021, which means that about 55% of the existing

252  household solid fuels for heating in these provinces shall be replaced with clean energy, such as

253  natural gas, electricity, combined heat and power with extreme low emissions, solar and

254  geothermal energy, and clean biofuels. While a thorough assessment and comparison of different

255  options is beyond our scope, we aim to illustrate the potential environmental and health benefits

256  that could be achieved through a state-of-the-art control option. For this purpose, we assume that

257  half of the solid fuels are replaced by natural gas and the other half by electricity (50% electric

258  resistance heating and 50% heat pump). As described in the main text, a successful implementation

259  of this policy would reduce the IPWE by 9.7% (8.8-10.4%) in China. The reduction ratio of IPWE

260  is relatively small because the current policy only focuses on household heating fuels (not cooking

261  fuels or non-household sources) in northern China (not middle or southern China). It should be

262  noted that this policy could have co-benefit on the transition in cooking fuels since the rural

263  residents will have easier access to clean fuels after the energy distribution system is constructed

264  or improved. Such co-benefit, however, is difficult to quantify and hence not considered in the

265  calculation. In addition, to evaluate the potential benefits of more stringent controls, we design

266  another scenario in which all solid fuels used for cooking and heating in 2015 were thoroughly

267  replaced with electricity and natural gas (50% each). Following the substitution, the consequent

268  increases in consumption of clean fuels (electricity or natural gas) are calculated according to the

269  assumption that the same amount of useful energy (fuel consumption multiplied by energy

270  efficiency) for both cooking and heating are generated before and after substitution. For cooking,

271  the energy efficiencies of coal stove, biomass stove, natural gas stove, and electric cooking are

272  assumed to be 35%, 17%, 50%, and 90%, respectively (7, 10). With respect to heating, coal stove,

273  biomass stove, natural gas heating, electric resistance heating, and heat pump are assumed to have

274  energy efficiencies of 50%, 24%, 90%, 98%, and 250%, respectively (7, 11, 46). For the electricity

275  substitution, we assume that the additional household electricity consumption in each province

276  was generated within the same province, considering a transmission loss of 6.6%, the average rate

277  in 2015 (47).

278  To compare the benefits of substitution by electricity or natural gas, we design two additional

279  scenarios in which all solid fuels used for cooking and heating in 2015 were thoroughly substituted

280  by electricity and natural gas, respectively. In these two scenarios, the fractional reductions in

281  IPWE compared to 2015 would be 63% (57-68%) and 62% (57%-67%), respectively. The

282  corresponding avoided premature deaths are 0.51 (0.40-0.64) million and 0.50 (0.39-0.64) million,

283  respectively. The benefits are very similar because the exposure increase due to additional

284  electricity or natural gas consumption ($< 0.5$ $\mu g/m^3$) are much smaller than the exposure decrease

285  due to reduced solid fuels (60 $\mu g/m^3$). Therefore, the choice of natural gas or electricity should not

286  be determined based on the health benefit, but rather based on economic cost, availability of

287  resources, and feasibility of infrastructure construction, which merits further in-depth studies.

288  In the scenarios, the energy mix and shares of end-of-pipe control technologies of the added

289  power plants are assumed to be same as the existing units in 2015. To evaluate the impact of the

290  energy mix of the added power plants, we design a sensitivity scenario which assumes that all

291  added power plants were coal-fired units. In this case, the electricity substitution was estimated to

292  reduce 62% (57-67%) of the IPWE in 2015 and avoid 0.50 (0.39-0.64) million premature deaths,

293  very similar to our current estimations, i.e., 63% (57-68%) and 0.51 (0.40-0.64) million. The small

294  sensitivity of the environmental and health benefits to the energy mix is also explained by the fact

295  the $PM_{2.5}$ exposure increase due to additional power plants is much smaller than the $PM_{2.5}$ exposure

296  decrease due to the eliminated household solid fuels. Note that the environmental and health

297  benefits could diminish if the end-of-pipe control measures of the added power plants were not as

298  stringent as they were in 2015. We suggest this is very unlikely to happen considering the stringent

299  emission standard for power plants in China.

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

302      The HAP exposure levels of solid-fuel users (i.e., $HAP_{j,k}$ in Eq. 2 in Methods) are subject to

303  relatively large uncertainties. In the GBD study (48), the parameters derived from in-situ

304  measurements and a prediction model over India were applied to the whole world. The Indian

305  average $PM_{2.5}$ exposures among all solid fuel-using households (which include the contribution

306  from AAP) were estimated at 285 (95% confidence interval, 201-405) $\mu g/m^3$ for children, 337

307  (238-479) $\mu g/m^3$ for women and 204 (144-290) $\mu g/m^3$ for men (48). Applying these estimates (the

308  contribution from AAP was subtracted to be consistent with this study), we estimate an IPWE in

309  2005 of 191 (155-248) $\mu g/m^3$, quite similar to the results of the current study (180 (146-219)

310  $\mu g/m^3$). Also, applying the estimates of GBD (48), household fuels are calculated to lead to a

311  reduction of 77 (50-119) $\mu g/m^3$ in IPWE from 2005 to 2015, representing 91% (87-94%) of the

312  total IPWE reduction, which is also similar to the estimates in the current study (76 $\mu g/m^3$ and

313  90%).

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440

**Tables and Figures**
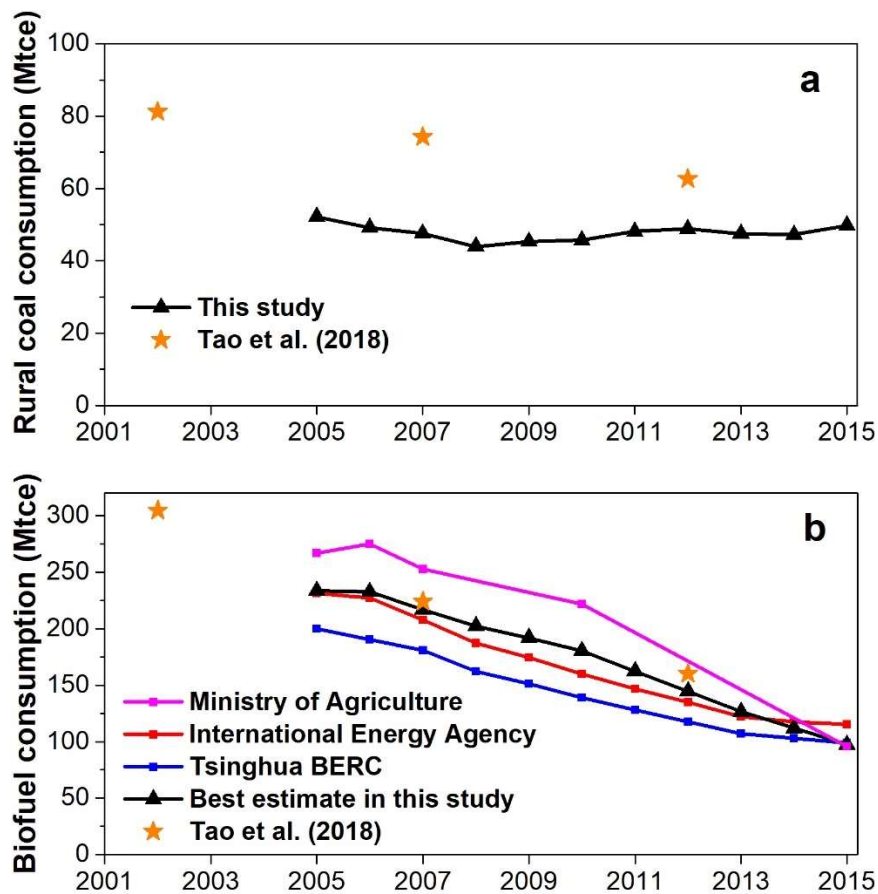
**Figure S1.** Household coal and biomass consumption from different nationwide statistics and surveys. The data sources are described in SI Appendix Section 1. The values of "best estimate in this study" in (b) are derived by taking the average of the data from the Ministry of Agriculture and Tsinghua-BERC, which are bottom-up statistical or survey data compiled by local institutes.
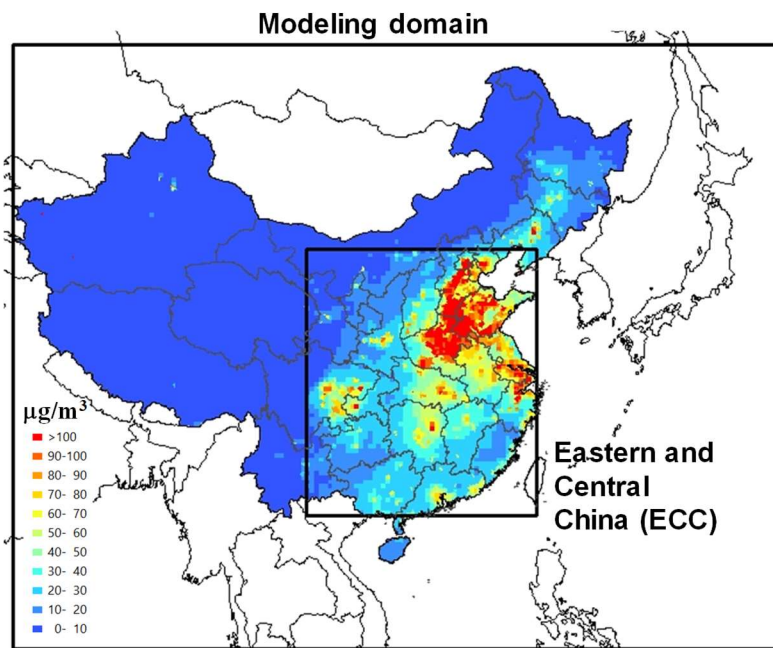
**Figure S2.** Modeling domain used in this study. The inner black box denotes the Eastern and Central China (ECC), which covers most of the populous areas in China characterized by substantial anthropogenic emissions and high ambient PM$_{2.5}$ concentrations. The colors represent ambient PM$_{2.5}$ concentrations in 2005, which are the same as Fig. 1c.

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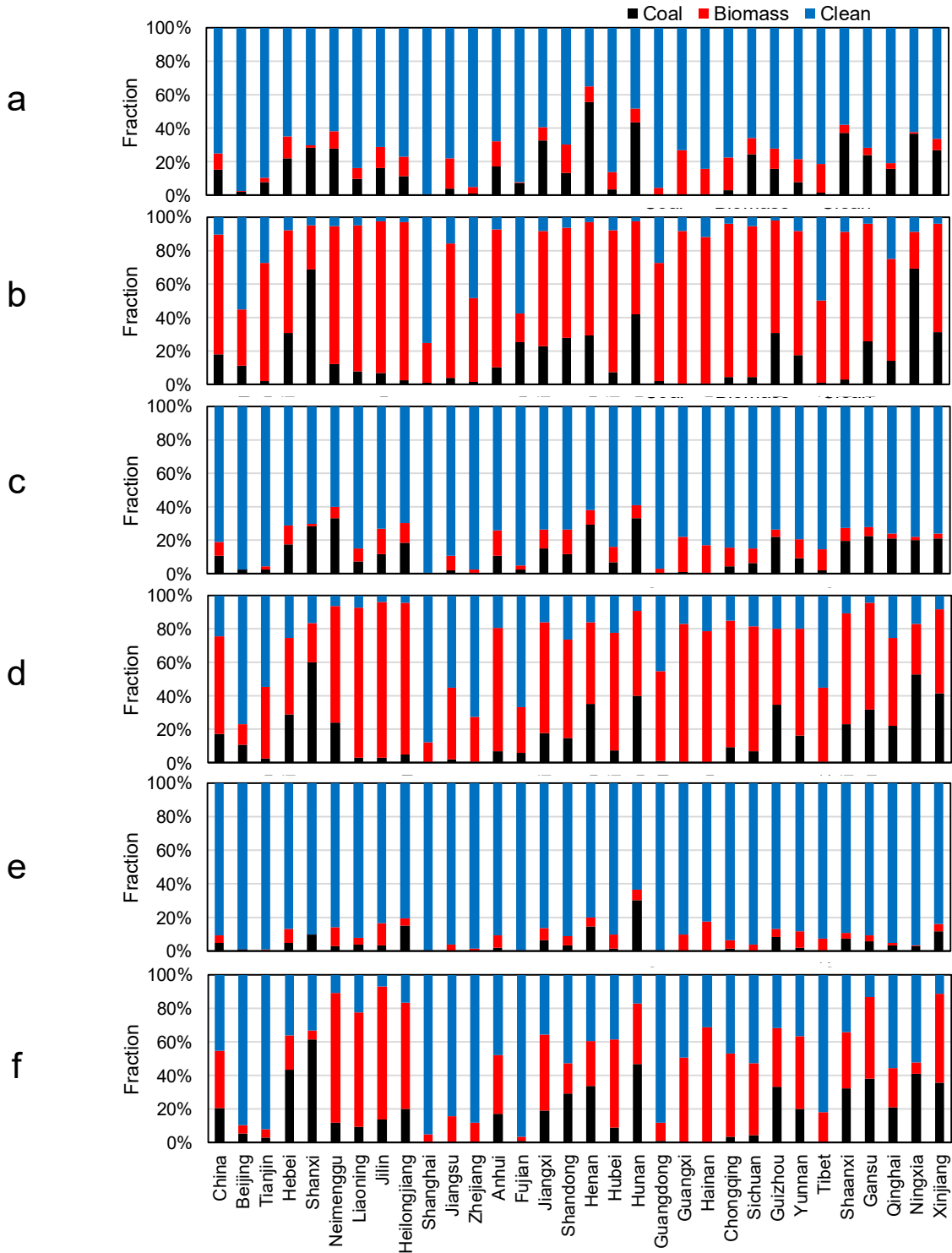
**Figure S3.** Share of major cooking fuels among urban and rural populations by province in 2005 and 2015: **a**, urban, 2005; **b**, rural, 2005; **c**, urban, 2010; **d**, rural, 2010; **e**, urban, 2015; **f**, rural, 2015. "Clean" refers to electricity and gaseous fuels (natural gas, liquified petroleum gas, coal gas, and biogas).
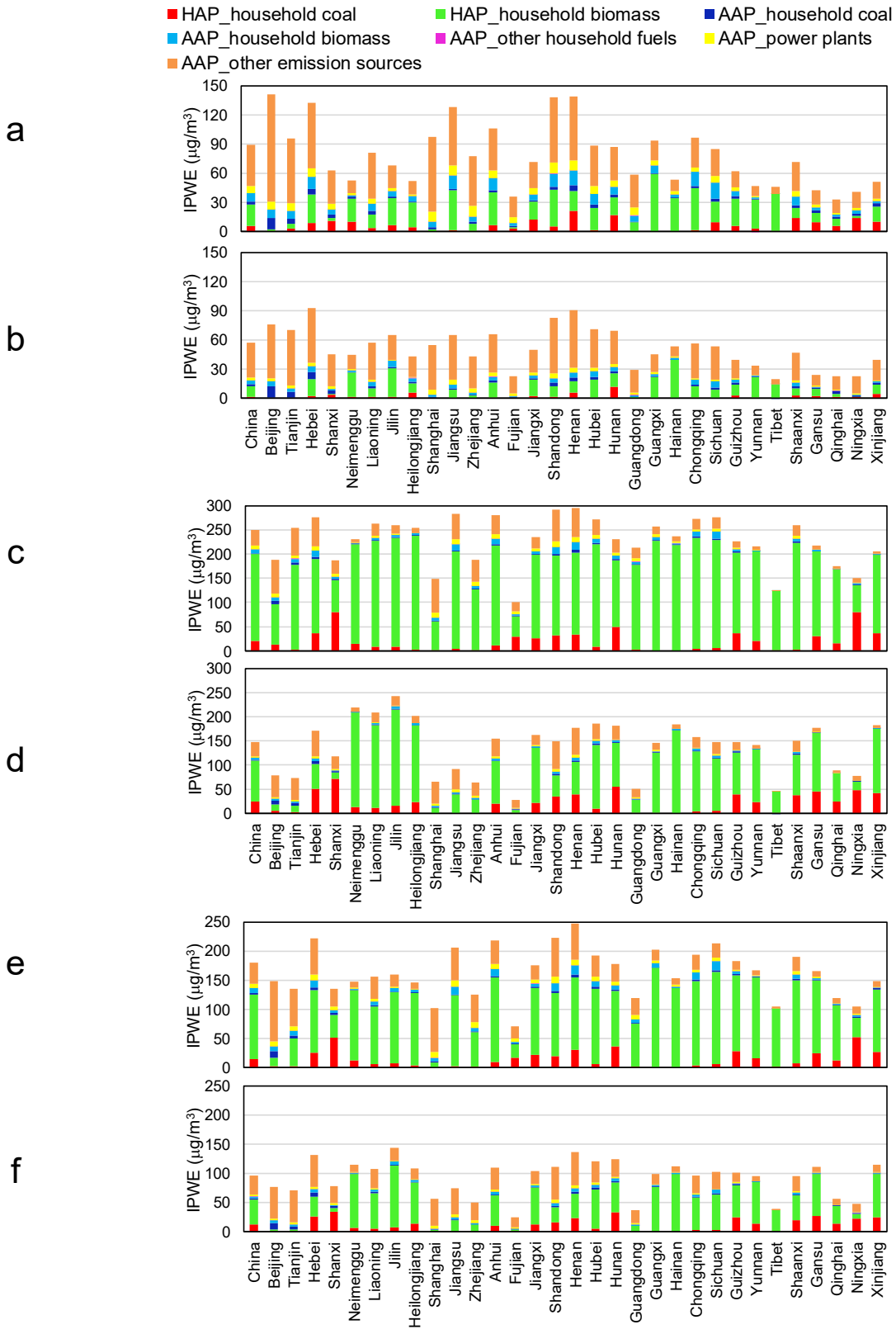
**Figure S4.** Contribution of individual sources to IPWE among urban (**a, b**), rural (**c, d**), and all (**e, f**) populations in 2005 (**a, c, e**) and 2015 (**b, d, f**).

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

459 (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 |

460

461 (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 |

462

463 (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 |

464

465 (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 |

466

467 (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 |

468

469 (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 |

470

471 (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 |

472

473 (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 |

474

475 **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 | | | $\leqslant \pm 0.5$ | $\leqslant 2$ | $\leqslant 2$ | $\geqslant 0.6$ | | | $\leqslant \pm 0.5$ | $\leqslant 2$ | | $\geqslant 0.8$ | | | $\leqslant \pm 1$ | $\leqslant 2$ | | $\geqslant 0.6$ |
| 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 |

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

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

478    **Table S3.** Performance statistics for the comparison between simulated AOD/$NO_2$ column and satellite retrievals over the ECC region

479    (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 |

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

481

482 **Table S4.** Performance statistics for the comparison between simulated and observed PM$_{10}$
483 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% |

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

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

486

487 **Table S5.** Performance statistics for the comparison between simulated and observed PM$_{2.5}$
488 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% |

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

490