



# GeoHealth

# **RESEARCH ARTICLE**

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#### **Key Points:**

- Solid biofuel stove emissions contribute significantly to surface ambient air quality over China, India, and sub-Saharan Africa
- Globally 382,000 annual premature deaths are attributable to ambient PM<sub>2.5</sub> and ozone exposure from solid biofuel stove emissions
- Promoting clean household energy programs over low-income and middle-income countries to improve air quality and human health is recommended

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# Impacts of Global Solid Biofuel Stove Emissions on Ambient Air Quality and Human Health

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**Abstract** Global solid biofuel stove emissions strongly impact air quality, climate change, and human health. However, investigations of the impacts of global solid biofuel stove emissions on human health associated with  $PM_{2.5}$  (particulate matter with aerodynamic diameter  $\leq 2.5 \mu m$ ) and ozone (O<sub>3</sub>) are limited. Here, we quantify the impacts of global solid biofuel stove emissions on ambient PM2.5 and O3 air quality and the associated human health effects for the year 2010, using the Community Atmosphere Model coupled with Chemistry version 5.3. Annual mean surface PM2.5 concentrations from global solid biofuel stove emissions averaged over 2006–2010 are up to 23.1  $\mu$ g m<sup>-3</sup>, with large impacts found over China, India, sub-Saharan Africa, and eastern and central Europe. For surface O<sub>3</sub> impacts, we find that global solid biofuel stove emissions lead to increases in surface  $O_3$  concentrations by up to 5.7 ppbv for China, India, and sub-Saharan Africa, and negligible impacts or reductions of up to 0.5 ppbv for the US, Europe, and parts of South America. Global solid biofuel stove emissions for the year 2010 contribute to 382,000 [95% confidence interval (95CI): 349,000-409,000] annual premature deaths associated with PM2.5 and O<sub>3</sub> exposure, with the corresponding years of life lost as 8.10 million years (95CI: 7.38-8.70 million years). Our study highlights air quality and human health benefits of mitigating emissions from the global solid biofuel stove sector, especially over populous regions of low-income and middle-income countries, through promoting clean household energy programs for the residential energy supply.

**Plain Language Summary** Ambient air pollutant emissions from global solid biofuel stove sector, including trace gases and aerosols, play a vital role in our surface air quality associated with particulate matter and ozone, which subsequently adversely impact human health. In this study, we employ a global chemistry-climate model to simulate the effects of global solid biofuel stove emissions on ambient air quality and human health. Our study finds that global solid biofuel stove emissions contribute significantly to surface PM<sub>2.5</sub> and ozone air quality over China, India, and sub-Saharan Africa. In addition, annual total PM<sub>2.5</sub>-induced and ozone-induced premature deaths of ~382,000 are attributable to the global solid biofuel stove emissions. Our study highlights the air quality and human health benefits by promoting clean household energy globally for the residential energy supply over low-income and middle-income countries, especially over those populous regions.

# 1. Introduction

Globally, around 41% of households relied on solid fuels (including coal) for cooking in 2010 (Bonjour et al., 2013). Emissions from the global solid fuel sector negatively influence air quality (Archer-Nicholls et al., 2016; Bonjour et al., 2013; Liu et al., 2016), climate change (Archer-Nicholls et al., 2019; Butt et al., 2016; Huang et al., 2018; Kodros et al., 2015; Lacey et al., 2017), and human health (Anenberg et al., 2017a; Chafe et al., 2014; Conibear et al., 2018a, 2018b; Lacey et al., 2017; Zhao et al., 2018). A subsector of solid fuel is solid biofuel stove, which is defined as wood fuels, agriculture residues and dung used for household cooking, heating, and lighting (Huang et al., 2018). This study focuses on the impacts of global solid biofuel stove emissions on air quality and human health, as a way to provide insights for the clean cookstove intervention programs (Global Alliance for Clean Cookstoves, 2015; Rosenthal et al., 2018).

Air pollutant emissions from solid biofuel stoves include black carbon (BC), primary organic matter (POM), nonmethane volatile organic compounds (NMVOCs), carbon monoxide (CO), nitrogen oxides ( $NO_x$ ), sulfur dioxide ( $SO_2$ ), and ammonia ( $NH_3$ ), which are short-lived climate forcers (SLCFs). Among the SLCFs,



NMVOCs, NO<sub>x</sub>, and CO are important ozone (O<sub>3</sub>) precursors (Huang et al., 2017), and NMVOCs, NO<sub>x</sub>, SO<sub>2</sub>, and NH<sub>3</sub> are precursors of secondary organic and inorganic aerosols (Huang et al., 2020). Solid biofuel stoves also emit long-lived greenhouse gases (GHGs), including carbon dioxide and methane (Anenberg et al., 2017a). On a global scale, several studies have reported the global impacts of solid fuel sector emissions on air quality and human health (Butt et al., 2016; Chafe et al., 2014; Kodros et al., 2015; Lelieveld, 2017; Lelieveld et al., 2015; Silva et al., 2016), in which the emissions from residential solid biofuel stove and coal sectors lumped together in the bottom-up emission inventory. However, the quantitative and qualitative analyses of the impacts of global solid biofuel stove emissions on air quality and human health are relatively limited. Owing to the importance of solid biofuel stove emissions for regional ambient air quality, most of the published literature focuses on the associated PM2.5 (particulate matter with aerodynamic diameter ≤2.5 µm) air quality impacts (Archer-Nicholls et al., 2016; Carter et al., 2016; Chowdhury et al., 2019; Liu et al., 2016; Reddington et al., 2019). A limited number of studies report the impacts of solid fuel emissions on O<sub>3</sub> air quality (Conibear et al., 2018a; Rooney et al., 2019). However, none of these studies investigate the global PM2.5 and O3 air quality impacts associated with the solid biofuel stove sector, which underscores the necessity of a timely assessment of global impacts in order to gauge the benefits of clean cookstove interventions regionally, nationally, and globally (Global Alliance for Clean Cookstoves, 2015), or promote clean household energy programs (Quinn et al., 2018).

Exposure to ambient  $PM_{2.5}$  and  $O_3$  air pollutants is associated with cardiovascular and respiratory diseases in humans (Huang et al., 2020; Lelieveld et al., 2015; Stanaway et al., 2018; Turner et al., 2016). Based on the data from the World Health Organization, household air pollution caused roughly 3.8 million deaths in the year 2016 (WHO, 2018). The published literature on the impacts of residential emissions on human health primarily focuses on  $PM_{2.5}$  (Butt et al., 2016; Chafe et al., 2014; Conibear et al., 2018b; Kodros et al., 2015; Lacey et al., 2017; Zhao et al., 2018) or  $O_3$  (Conibear et al., 2018a). Limited studies have quantified the global impacts of residential solid fuel emissions on human health associated with  $PM_{2.5}$  and  $O_3$  (Lelieveld et al., 2015; Silva et al., 2016). However, global  $PM_{2.5}$ -induced and  $O_3$ -induced premature deaths associated with emissions from the solid biofuel stove sector have not been thoroughly quantified yet.

We follow our previous study (Huang et al., 2018) to additionally quantify the air quality and human health impacts associated with global solid biofuel stove emissions, by using the Community Earth System Model (CESM) CAM5-Chem (Community Atmosphere Model coupled with Chemistry) model version 5.3. Section 2 describes our methods for air quality model simulations and the mathematical approaches used to quantify human health associated with  $PM_{2.5}$  and  $O_3$ . Results are presented in Section 3. We conclude our study with discussions in Section 4.

# 2. Methods

#### 2.1. CAM5-Chem Model Simulations

In this study, we employ the CESM (version 1.2.2.1) CAM5-Chem model (Emmons et al., 2010; Huang et al., 2018; Lamarque et al., 2012; Tilmes et al., 2015) to investigate the impacts of global solid biofuel stove emissions on air quality and human health. CAM5-Chem contains a fully coupled Ozone-NO<sub>x</sub>-VOC-Aerosol chemistry scheme, with horizontal resolution of 0.9° latitude  $\times$  1.25° longitude and 56 vertical levels from the surface to about 40 km. CAM5-Chem is driven by offline Goddard Earth Observing System model version 5 (GEOS-5) meteorological fields, with gas-phase chemistry following the Model for Ozone and Related chemical Tracers, version 4 (Lamarque et al., 2012) and an aerosol microphysical scheme using 3-mode modal aerosol model (Liu et al., 2012).

Global anthropogenic emissions for CAM5-Chem simulations are from the IIASA GAINS (International Institute for Applied Systems Analysis Greenhouse Gas-Air Pollution Interactions and Synergies) ECLIPSE V5a (Evaluating the Climate and Air Quality Impacts of Short-lived Pollutants version 5a) inventory for the year 2010 (Amann et al., 2011, 2013; Klimont et al., 2017; Stohl et al., 2015), with horizontal resolution of 0.5° latitude  $\times$  0.5° longitude. This study expands on our previous study that estimated the impacts of global solid biofuel stove aerosol emissions on global climate (Huang et al., 2018). The model configuration in this study is identical to Huang et al. (2018), except that we now use version 1.2.2.1 of CESM, which is named when porting CESM version 1.2.2 to the NCAR Cheyenne supercomputers. The code of this version



is identical to CESM version 1.2.2, which is used in Huang et al. (2018). We run two model simulations: a control simulation and a sensitivity simulation, which differs from the control simulation only in that the global solid biofuel stove emissions are turned off. Both simulations are run from 2005 to 2010, with the first year discarded as spin-up and the last 5 years used for analysis. The impacts of global solid biofuel stove sector emissions associated with air quality and human health are calculated as the difference between the two simulations (control-sensitivity simulation). Due to the nonlinear exposure-outcome association for  $PM_{2.5}$  exposure, we acknowledge that this subtraction approach differs substantially from the attribution approach that scales the health impacts from total  $PM_{2.5}$  exposure by the fraction of  $PM_{2.5}$  exposure from the solid biofuel stove sector (Kodros et al., 2016). Simulated BC, organic aerosols, and aerosol optical depth in CAM5-Chem using ECLIPSE V5a were previously evaluated against a suite of observations (Huang et al., 2018). We have also conducted additional evaluations of model simulated  $PM_{2.5}$  and  $O_3$  against observations in this study (Section 3.1).

#### 2.2. PM<sub>2.5</sub>-Induced and O<sub>3</sub>-Induced Premature Deaths Calculation

We employed the newly developed Global Exposure Mortality Model (GEMM; Burnett et al., 2018) to quantify the  $PM_{2.5}$ -induced premature deaths for the global solid biofuel stove emissions. The Hazard Ratio (HR) functions associated with GEMM are derived based on cohort studies from 16 countries globally that are attributable to ambient  $PM_{2.5}$  exposure. In GEMM, it predicts nonaccidental deaths due to noncommunicable diseases (NCDs) and lower respiratory infections (LRIs) associated with long-term exposure to ambient  $PM_{2.5}$ . In this study, we estimate the  $PM_{2.5}$ -induced premature deaths due to NCD + LRI using GEMM for the control and sensitivity simulations. The HR function in GEMM at each horizontal model grid box of longitude (*i*) and latitude (*j*) for each health endpoint (*h*) and each 5-years age interval group (*a*) is calculated as

$$HR_{i,j,h,a} = \exp\left\{\frac{\theta \ln^{\left(\frac{z_{i,j}}{\alpha}+1\right)}}{1+\exp^{\left(-\frac{z_{i,j}-\mu}{\nu}\right)}}\right\}, z_{i,j} = \max\left(0, C_{i,j} - C_{0}\right)$$
(1)

where  $C_{i,j}$  is the annual mean surface PM<sub>2.5</sub> concentrations in the grid cell (i, j);  $C_0$  is the counterfactual PM<sub>2.5</sub> concentration as 2.4 µg m<sup>-3</sup>, below which no premature death risk associated with PM<sub>2.5</sub> exposure is assumed.  $\theta$ ,  $\alpha$ ,  $\mu$ , and  $\nu$  are the GEMM model fit parameters, which are obtained directly from Burnett et al. (2018). The standard errors (SE) of  $\theta$  are used to estimate the uncertainty and 95% confidence interval (95CI) of the PM<sub>2.5</sub>-induced premature deaths.

We follow Turner et al. (2016) and Huang et al. (2020) to account for O<sub>3</sub>-induced chronic obstructive pulmonary diseases (COPD), which shows that HR increases by 14% (95CI: 8–21%) per 10 ppbv increase in annual mean daily maximum 8-h (MDA8) O<sub>3</sub> concentrations. For each grid cell (*i*, *j*), HR associated with O<sub>3</sub>-induced COPD exposure is shown in Equation (2).

$$HR_{i,j} = \exp^{(\eta \Delta Y_{i,j})}, \Delta Y_{i,j} = \max(0, Y_{i,j} - 26.7)$$
(2)

where  $\eta$  is the log-linear slope between O<sub>3</sub> concentrations and HR associated with COPD, with mean value of 0.0131 (95CI: 0.0077–0.0191).  $Y_{i,j}$  is the annual mean MDA8 O<sub>3</sub> concentration in grid cell (*i*, *j*). We assume that the threshold value of MDA8 O<sub>3</sub> concentration is 26.7 ppb, below which there will be no mortality risk associated with O<sub>3</sub>-induced COPD.

The global premature deaths associated with  $PM_{2.5}$  and  $O_3$  are calculated as a function of population density (POP), the baseline mortality rate (BMR), and HR for each disease in response to different levels of  $PM_{2.5}$  or  $O_3$  concentrations. Both  $PM_{2.5}$ -induced and  $O_3$ -induced premature deaths are calculated at the horizontal grid resolution of 0.1° latitude  $\times$  0.1° longitude. Gridded  $PM_{2.5}$  and  $O_3$  concentrations from CAM5-Chem model simulations are regridded from 0.9° latitude  $\times$  1.25° longitude to 0.1° latitude  $\times$  0.1° longitude. The



mathematical formula for premature deaths ( $M_{i,j,h,a}$ ) associated with PM<sub>2.5</sub> and O<sub>3</sub> in each grid cell (longitude *i*, latitude *j*) for each health endpoint (*h*) and each age group is shown as

$$M_{i,j,h,a} = POP_{i,j,a} \times BMR_{i,j,h,a} \times \begin{bmatrix} (HR_{i,j,h,a} - 1) \\ HR_{i,j,h,a} \end{bmatrix}$$
(3)

where  $POP_{i,j,a}$  is the gridded population density in each grid cell (*i*, *j*) for each age group (*a*), which is calculated as a product of total population counts in each grid box and the age-specific (5-years age interval) population fraction. The gridded population density is downloaded from the Gridded Population of the World version 4 (GPWv4) for the year 2010 (Center for International Earth Science Information Network-CIES-IN-Columbia University, 2018) at the horizontal resolution of 2.5 min × 2.5 min, which is regridded to 0.1° latitude × 0.1° longitude. We use the age-specific (5-years age interval) death numbers and rates in each region from the 2017 Global Burden of Disease for the year 2010 (GBD 2017 Risk Factors Collaborators, 2018; hereafter referred to GBD2017) to derive the age-specific populations. For each grid box in each region, we assume this fraction is constant. BMR is the baseline mortality rate, which is estimated following the GBD2017 study for the year 2010. For each health endpoint, the BMR values vary for each 5-years age interval and each region. The regional annual total PM<sub>2.5</sub>-induced or O<sub>3</sub>-induced premature deaths are summed up from each 5-years age interval.

Years of life lost (YLL) for each grid cell (i, j), health endpoint (h), and age group (a) are calculated as

$$YLL_{i,j,h,a} = M_{i,j,h,a} \times MYLL_{i,j,h,a}$$
(4)

where  $MYLL_{i,j,h,a}$  is the mean YLL for each health endpoint and each age group attributable to all causes from the GBD2017 study for the year 2010.

Following Huang et al. (2020), we estimate premature deaths and the corresponding YLLs for each disease associated with  $PM_{2.5}$  and  $O_3$  by dividing the continent of the world into 11 regions, which include China, India, rest of Asia (ROA), sub-Saharan Africa (SSA), northern Africa and the Middle East (NAME), eastern and central Europe (ECEurope), western Europe (WEurope), the United States of America (USA), Latin America (LATIN), Canada, and rest of the world (ROW). For each 5-years interval age group, we assume that regional BMR for each disease in each region is constant. We sum the health endpoints associated with  $PM_{2.5}$  and  $O_3$  from each 5-years interval age group to estimate the regional and global total premature deaths and YLLs.

# 3. Results

#### 3.1. Evaluations of Model Simulated Surface PM<sub>2.5</sub> and O<sub>3</sub> Concentrations

We used the annual mean surface  $PM_{2.5}$  measurement networks from China, India, Europe and USA to compare with model simulated surface  $PM_{2.5}$  concentrations averaged over 2006–2010 from CAM5-Chem (Figure 1). For  $PM_{2.5}$  measurements over China, due to the unavailability of measurement data from 2006 to 2010, we used the 2012–2014 measurements from 40 ground stations, as presented in Liu et al. (2018), to compare with model simulations. We compiled the sporadic measurements of surface  $PM_{2.5}$  concentrations during 2006–2010 across India from the existing literatures, which included measurements over Agra (Kulshrestha et al., 2009), Anantapur (Balakrishnaiah et al., 2011), Chennai (Bathmanabhan & Saragur Madanayak, 2010), Dibrugarh (Pathak et al., 2013), Hyderabad (Guttikunda et al., 2013), and New Delhi (Tiwari et al., 2009). Long-term measurements of surface  $PM_{2.5}$  concentrations during the period of 2006–2010 in Europe were from the European Monitoring and Evaluation Programme (EMEP; https://www.emep.int), and in the United States of America were from the US Air Quality System Data Mart (https://aqs.epa.gov/aqsweb/airdata/download\_files.html), with a total of 101 and 216 measurement sites in Europe and US, respectively. Figure 1 shows the scatter plots of surface  $PM_{2.5}$  concentrations between observations and model simulations from China (Figure 1a), India (Figure 1b), Europe (Figure 1c), and US (Figure 1d). We define the normalized mean bias (NMB) between observations (denoted as *O*) and model simulations (denoted as





**Figure 1.** Scatter plots of surface  $PM_{2.5}$  concentrations between observations and model simulations from (a) China, (b) India, (c) Europe, and (d) USA. The total number of observational sites (*n*), measurement-model regression correlation coefficient (*r*), slope (*k*), and normalized mean bias (NMB) are inserted in each panel.

*M*) as *NMB* =  $\begin{pmatrix} \sum_{i} (M_{i} - O_{i}) / \\ \sum_{i} O_{i} \end{pmatrix} \times 100\%$ , where *i* represents each observational site in each region. Glob-

ally, model simulated surface PM<sub>2.5</sub> concentrations agreed with observations within a factor of 2, with NMB value of ~37.5%. Regionally, model simulated surface PM<sub>2.5</sub> concentrations from India (NMB = 3.4%) and Europe (NMB = 7.7%) agreed quite well with observations. However, we found that CAM5-Chem model overestimated surface PM<sub>2.5</sub> concentrations over China (NMB = 48%) and US (NMB = 53%). This is likely due to the combined effects of overestimates of both sulfate (Tilmes et al., 2015) and secondary organic aerosols (Tsigaridis et al., 2014) concentrations in CAM5-Chem, compared with observations.

For comparisons of surface  $O_3$  between model simulations and observations, we employed the gridded monthly mean surface O<sub>3</sub> concentrations from the Tropospheric Ozone Assessment Report (TOAR) at the horizontal resolution of  $2^{\circ}$  latitude  $\times 2^{\circ}$  longitude during the period of 2006–2010 (Schultz et al., 2017) to compare with model simulated surface O<sub>3</sub> concentrations from CAM5-Chem. We regridded the model simulated surface  $O_3$  from  $0.9^\circ$  latitude  $\times 1.25^\circ$  longitude to  $2^\circ$  latitude  $\times 2^\circ$  longitude in order to compare with observations. Figure 2 shows the spatial variability of differences in surface O<sub>3</sub> concentrations between model simulations and observations (model minus observation). The CAM5-Chem model tends to overestimate surface  $O_3$  concentrations, with large positive mean bias of 17.9 ppbv. The overestimates of surface O<sub>3</sub> concentrations from CAM5-Chem have been reported in Tilmes et al. (2015) and Lamarque et al. (2012). The improved representations of reactive nitrogen and organic nitrate compounds in the CESM version 2 have reduced the biases of summertime surface  $O_3$  over the southeast US, compared with observations (Emmons et al., 2020), which may be a factor that leads to the high bias of model simulated  $O_3$  concentrations in CAM5-Chem. In addition, other factors that resulted in high bias of surface  $O_3$  in CAM5-Chem may include the coarse model resolution, misrepresentation of physical processes and offline meteorology (Lamarque et al., 2012; Tilmes et al., 2015). We acknowledge that high biases of surface O<sub>3</sub> concentrations in our control simulation may result in uncertainty in the annual total premature deaths associated with COPD attributable to long-term surface O<sub>3</sub> exposure.





**Figure 2.** Global gridded spatial variability of differences in surface  $O_3$  concentrations between the control model simulation in our study (CTRL) and the observations from TOAR at the horizontal resolution of 2° latitude × 2° longitude during the period of 2006–2010. White spaces are the grid boxes without measurements. The mean biases are calculated as the mean of differences between the control simulations and TOAR in each grid box where both observations and simulations are available during 2006–2010. TOAR, Tropospheric Ozone Assessment Report.

#### 3.2. Impacts of Global Solid Biofuel Stove Emissions on Surface PM<sub>2.5</sub> and O<sub>3</sub> Air Quality

Table 1 shows the annual regional and global budgets of various species for the solid biofuel stove sector emissions for the year 2010. Specifically, annual BC emissions from the global solid biofuel stove sector are 2.31 Tg, accounting for about 23.7% of the global anthropogenic and natural sources of BC (Huang et al., 2018). Large contributions to the sectoral BC burden come from SSA, China, ROA, and India, with fractional contributions relative to the global annual totals of BC from solid biofuel stove sector as 30.4%, 18.4%, 18.3%, and 14.7 %, respectively. A similar scenario occurs for POM associated with the global solid

Regional and Global Annual Budgets of Various Species From Global Solid Biofuel Stove Emissions (Units: kt Species/yr)								
	BC	POM	NMVOCs	СО	NO <sub>x</sub>	SO <sub>2</sub>	NH <sub>3</sub>	
China	423.9	1,706	5,980	44,302	571.4	540.9	65.1	
India	338.8	1,155	4,338	31,413	274.9	135.3	45.1	
ROA	422.5	1,267	4,406	31,529	289.3	344.9	47.4	
SSA	703.2	2,214	9,455	59,819	627.9	261.6	85.8	
NAME	34.6	90.3	404.5	1,839	26.1	10.9	3.59	
ECEurope	58.3	140.4	584.8	1,907	55.0	23.7	5.42	
WEurope	57.8	96.6	452.1	2,138	87.0	26.4	8.49	
LATIN	102.4	326.5	1,206	7,578	83.5	34.8	11.4	
USA	37.2	89.9	466.6	1,152	24.2	10.1	3.31	
Canada	21.6	43.7	174.6	751.8	14.2	5.90	1.93	
ROW	5.19	11.9	62.4	146.2	3.34	1.29	0.46	
Global <sup>a</sup>	2,310	10,370	28,700	190,000	2,500	1,400	280	

<sup>a</sup>Due to the edge effect of the regridding process, the sum of the regional emissions does not exactly match the global total shown in the table.

Table 1





Solid Biofuel Stove Surface PM2.5 Concentrations

**Figure 3.** Spatial distribution of annual mean surface PM<sub>2.5</sub> concentrations from the global solid biofuel stove emissions, averaged over 2006–2010. Domain definitions of eastern China (purple), northern India and Nepal (blue), and northeastern SSA (green) for regional analyses are inserted in the plot as colored rectangular boxes.

biofuel stove emissions, with global annual total POM emissions of 10.37 Tg, accounting for 20.8% of global POM emissions from all anthropogenic (18.9 Tg/yr) and natural (31.0 Tg/yr) sources. The largest source of POM emissions globally is from natural biomass burning (Huang et al., 2013). The fractional contributions of POM emissions from SSA, China, ROA, and India relative to the global annual total solid biofuel stove POM emissions are 21.4%, 16.5%, 12.2%, and 11.1%, respectively.

In terms of O<sub>3</sub> precursors, global annual total NMVOCs (28.7 Tg/yr) and CO (190 Tg/yr) emissions from the solid biofuel stove sector account for ~35% each relative to the corresponding global annual total anthropogenic NMVOC (81.1 Tg/yr) and CO (548 Tg/yr) emissions for the year 2010 (Huang et al., 2018). Globally, annual total NO<sub>x</sub> (2.5 Tg/yr), SO<sub>2</sub> (1.4 Tg/yr), and NH<sub>3</sub> (0.3 Tg/yr) emissions associated with the solid biofuel stove sector relative to the corresponding global annual total anthropogenic emissions are marginal, which only account for about 2%, 1.4%, and 0.5 %, respectively (Table 1).

Annual mean surface  $PM_{2.5}$  concentrations from the global solid biofuel stove sector, averaged over 2006–2010, range up to 23.1 µg m<sup>-3</sup>, with large burdens simulated over China, India, southeastern Asia, SSA, and eastern and central Europe (Figure 3). We define three regions for statistical analysis, which are eastern China, northern India and Nepal, and northeastern SSA, as shown in colored rectangular boxes in Figure 3. Annual mean surface  $PM_{2.5}$  concentrations averaged over eastern China, northern India and Nepal, and northeastern SSA, as shown in colored rectangular boxes in Figure 3. Annual mean surface  $PM_{2.5}$  concentrations averaged over eastern China, northern India and Nepal, and northeastern SSA are 7.7, 11.0, 3.3 µg m<sup>-3</sup>, of which primary carbonaceous aerosols (BC + POM) relative to total  $PM_{2.5}$  concentrations associated with solid biofuel stove over these three regions account for 63.6%, 79.1%, 72.7%, respectively. This suggests that secondary organic and inorganic contributions to total  $PM_{2.5}$  concentrations from solid biofuel stove emissions in these areas are less important than the primary carbonaceous sources. This is primarily driven by the low emissions of NO<sub>x</sub>, SO<sub>2</sub>, and NH<sub>3</sub> associated with the global solid biofuel stove sector (Table 1), resulting in lower levels of oxidants and secondary aerosols, compared with other anthropogenic emission sectors, such as energy, industry, agriculture, and transportation (Huang et al., 2018, 2020).

Major impacts of global solid biofuel stove emissions on surface  $O_3$  are confined to source regions with significant emissions from central-eastern China, South Asia (particularly India), Southeast Asia, and SSA (except South Africa), with solid biofuel stove emissions driving annual mean surface  $O_3$  enhancements of up to 5.7 ppbv over central China (Figure 4). Regional annual mean surface  $O_3$  concentrations due to solid biofuel stove emissions averaged over 2006–2010 in central-eastern China, northern India and Nepal, and





Solid Biofuel Stove Surface Ozone Concentrations

**Figure 4.** Spatial distribution of annual mean surface  $O_3$  concentrations from the global solid biofuel stove emissions, averaged over 2006–2010. Domain definitions of eastern China (purple), northern India and Nepal (blue), and northeastern SSA (green) for regional analyses are inserted in the plot as colored rectangular boxes.

northeastern SSA (as shown in Figure 4) are 2.1, 2.9, and 1.2 ppbv, respectively. We also observe positive impacts for surface  $O_3$  from the solid biofuel stove sector over western SSA. Solid biofuel stove emissions over the US, Europe, and part of South America lead to reductions (up to -0.5 ppbv) or negligible impacts on surface  $O_3$  air quality, likely due to the combined effect of small-scale  $NO_x$  emissions associated with the solid biofuel stove sector and the nonlinearity of  $O_3$  chemical production (Jiang et al., 2019; Lin et al., 2017).

#### 3.3. Impact of Global Solid Biofuel Stove Emissions on Premature Deaths

Global annual total PM<sub>2.5</sub>-induced and O<sub>3</sub>-induced premature deaths of 9.29 million (95CI: 8.31–10.22 million) are attributable to natural and anthropogenic pollution sources for the control simulation for the year 2010 (Table 2), with the fraction of  $PM_{2.5}$ -induced premature deaths accounting for ~89.7%. This is consistent with previous studies using GEMM (Burnett et al., 2018; Lelieveld et al., 2019). The annual total premature deaths of NCD + LRI attributable to long-term  $PM_{2.5}$  exposure for the year 2010 in this study are over a factor of 2 higher than the previous studies using the integrated exposure-response (IER) functions developed in GBD for the estimates of five causes of deaths, which are lower respiratory infection, stroke, ischemic heart disease, lung cancer, and COPD (Anenberg et al., 2017b; Huang et al., 2020; Lelieveld et al., 2015). China and India collectively account for about 50% of the total global annual  $PM_{2.5}$ -induced and O<sub>3</sub>-induced premature deaths. Figure 5 shows the global spatial distribution of annual total PM2,5-induced and O3-induced premature deaths associated with global solid biofuel stove emissions, with global mean annual total premature deaths of ~382,000 (95CI: 349,000-409,000). The corresponding mean YLLs for the global solid biofuel stove sector are ~8.10 million years (95CI: 7.38-8.70 million years). PM<sub>2.5</sub>-induced premature deaths account for 92.1% of global annual total PM<sub>2.5</sub>-induced and O<sub>3</sub>-induced premature deaths, which suggests that global premature deaths for the solid biofuel stove sector are dominated by PM<sub>2.5</sub> pollution.

Figure 6 shows regional annual total  $PM_{2.5}$ -induced and  $O_3$ -induced premature deaths from solid biofuel stove emissions, with the largest regional impacts found in China, followed by India, ROA, and SSA. The percentage of regional annual total premature deaths in response to  $PM_{2.5}$  and  $O_3$  exposure for China, India, ROA, and SSA relative to the global totals associated with solid biofuel stove emissions are 36.9%, 28.8%, 20.2%, and 5.6 %, respectively. Similarly, China ranks the highest in terms of regional annual total YLLs [2.65 million years (95CI: 2.44–2.82 million years)] attributable to  $PM_{2.5}$  and  $O_3$  exposure from solid biofuel



# Table 2

Global and Regional  $PM_{2.5}$ -Induced and  $O_3$ -Induced Annual Premature Deaths (×1,000) for the Control Simulation Associated With Total Natural and Anthropogenic Emissions (Denoted as "CONTROL") and the Sensitivity Simulation Associated With the Global Solid Biofuel Stove Sector for the Year 2010

Region		Control	Solid biofuel stove
China	PM <sub>2.5</sub>	2,386 (2,204, 2,561)	122.4 (114.7, 129.4)
	O <sub>3</sub>	372.9 (242.8, 487.7)	18.49 (13.56, 21.11)
	$PM_{2.5} + O_3$	2,759 (2,447, 3,049)	140.9 (128.3, 150.5)
India	PM <sub>2.5</sub>	1,635 (1,512, 1,753)	93.44 (87.74, 98.65)
	O <sub>3</sub>	302.9 (198.8, 393.3)	16.62 (12.46, 18.54)
	$PM_{2.5} + O_3$	1,938 (1,711, 2,146)	110.1 (100.2, 117.2)
ROA	PM <sub>2.5</sub>	1,322 (1,219, 1,422)	73.57 (68.35, 78.50)
	O <sub>3</sub>	105.7 (67.79, 140.4)	3.701 (2.640, 4.370)
	$PM_{2.5} + O_3$	1,428 (1,287, 1,562)	77.27 (70.99, 82.87)
NAME	PM <sub>2.5</sub>	698.4 (650.5, 743.5)	0.505 (0.469, 0.540)
	O <sub>3</sub>	21.01 (13.46, 27.93)	0.002 (0.002, 0.003)
	$PM_{2.5} + O_3$	719.4 (664.0, 771.4)	0.507 (0.471, 0.543)
SSA	PM <sub>2.5</sub>	581.9 (538.2, 623.9)	20.22 (18.77, 21.59)
	O <sub>3</sub>	13.29 (8.172, 18.45)	1.092 (0.695, 1.458)
	$PM_{2.5} + O_3$	595.2 (546.4, 642.4)	21.31 (19.47, 23.04)
ECEurope	PM <sub>2.5</sub>	609.8 (557.9, 660.6)	10.09 (9.289, 10.87)
	O <sub>3</sub>	21.99 (13.71, 30.09)	-0.114 (-0.075, -0.146)
	$PM_{2.5} + O_3$	631.8 (571.6, 690.7)	9.978 (9.214, 10.72)
USA	PM <sub>2.5</sub>	288.9 (263.7, 313.6)	3.277 (3.010, 3.537)
	O <sub>3</sub>	48.98 (31.35, 65.19)	-0.328 (-0.231, -0.393)
	$PM_{2.5} + O_3$	337.9 (295.1, 378.8)	2.949 (2.779, 3.144)
LATIN	PM <sub>2.5</sub>	302.6 (276.3, 328.4)	9.644 (8.860, 10.41)
	O <sub>3</sub>	22.69 (14.14, 31.08)	-0.020 (-0.014, -0.025)
	$PM_{2.5} + O_3$	325.3 (290.4, 359.5)	9.624 (8.846, 10.38)
WEurope	PM <sub>2.5</sub>	465.2 (424.8, 504.9)	8.634 (7.931, 9.315)
	O <sub>3</sub>	45.86 (28.78, 62.31)	-0.330 (-0.220, -0.418)
	$PM_{2.5} + O_3$	511.1 (453.6, 567.2)	8.304 (7.711, 8.897)
Canada	PM <sub>2.5</sub>	19.83 (18.06, 21.57)	1.302 (0.945, 1.118)
	O <sub>3</sub>	2.992 (1.875, 4.071)	-0.017 (-0.012, -0.021)
	$PM_{2.5} + O_3$	22.82 (19.94, 25.64)	1.015 (0.933, 1.097)
ROW	PM <sub>2.5</sub>	2.128 (1.944, 2.309)	0.086 (0.079, 0.092)
	O <sub>3</sub>	0.823 (0.500, 1.157)	-0.009 (-0.006, -0.012)
	$PM_{2.5} + O_3$	2.951 (2.444, 3.466)	0.077 (0.073, 0.080)
Global	PM <sub>2.5</sub>	8,331 (7,685, 8,956)	342.9 (320.2, 364.0)
	O <sub>3</sub>	959.1 (621.3, 1,262)	39.08 (28.80, 44.46)
	$PM_{2.5} + O_3$	9,290 (8,306, 10,218)	381.9 (349.0, 408.5)





**Figure 5.** Spatial distribution of annual total  $PM_{2.5}$ -induced and  $O_3$ -induced premature deaths from the global solid biofuel stove emissions. The horizontal resolution of the plot is  $0.9^{\circ}$  latitude  $\times 1.25^{\circ}$  longitude.

stove emissions in 2010, as shown in Figure 7, followed by India [2.55 million years (95CI: 2.35–2.71 million years)], ROA [1.73 million years (95CI: 1.59–1.85 million years)], and SSA [0.55 million years (95CI: 0.43–0.66 million years)].









Figure 7. Regional annual total YLLs associated with PM<sub>2.5</sub>-induced and O<sub>3</sub>-induced premature deaths from solid biofuel stove emissions. Error bars represent 95% confidence intervals.

# 3.4. Comparisons With Other Studies

Figure 8 compares our estimates of  $PM_{2.5}$ -induced and  $O_3$ -induced annual total premature deaths from solid biofuel stove sector emissions with those from other publications, globally (Figure 8a), in India (Figure 8b), and in China (Figure 8c). Global annual total  $PM_{2.5}$ -induced and  $O_3$ -induced premature deaths associated with the residential and commercial energy use sector for the year 2005 by Silva et al. (2016) and 2010 by Lelieveld et al. (2015) were up to 728,700 (95CI: 440,300–1,015,000) and 1,022,000 (Figure 8a), respectively, which were a factor of 1.9 and 2.7 higher than the values in our study [382,000 (95CI: 349,000–409,000)]. There are two main factors contributing to these differences. First, the residential and commercial energy use sector in Silva et al. (2016) and Lelieveld et al. (2015)



**Figure 8.** Comparisons of annual total premature deaths associated with solid fuel emissions between our study and previous studies for the globe (a), India (b), and China (c). Our study is presented as annual total  $PM_{2,5}$ -induced and  $O_3$ -induced premature deaths for the solid biofuel stove emissions. Error bars denote the 95% confidence intervals.

includes residential coal combustion, whereas our study solely accounts for solid biofuel stove emissions (wood fuels + agricultural residues + dung). We speculate that inclusion of residential coal in our modeling study will improve the agreement of the annual total  $PM_{2.5}$ -induced and  $O_3$ -induced premature deaths attributable to solid fuel sector with Silva et al. (2016) and Lelieveld et al. (2015), in conjunction with the much higher mortality HRs used in GEMM than previous IER functions. Second, in terms of BMR for each health endpoint in each region, our study used GBD2017 for the year 2010, whereas Silva et al. (2016) used the GBD 2010 Risk Factors Collaborators (2012; hereafter referred to as GBD2010) for the year 2005 and Lelieveld et al. (2015) used GBD2010 for the year 2010. We also acknowledge that there exist large differences between ECLIPSE V5a that used in our study and other regional or global emission inventories in terms of trace gases and aerosols emissions (Saikawa et al., 2017a, 2017b).

In terms of  $PM_{2.5}$ -induced premature deaths associated with the global solid fuel sector, global annual total  $PM_{2.5}$ -induced premature deaths of 342,900 (95CI: 320,200–364,000) from our study are comparable to Chafe et al. (2014), Butt et al. (2016), and Lacey et al. (2017), which have reported that global annual total  $PM_{2.5}$ -induced premature deaths of 370,000, 308,000 (95CI: 113,300–497,000), and 260,000 (95CI: 137,000–268,000), respectively, are attributable to household solid fuel emissions (Figure 8a). However, both Chafe et al. (2014) and Butt et al. (2016) included coal usage as residential emissions in their studies, and the solid fuel emission inventory in Lacey et al. (2017) only accounted for carbonaceous aerosols and SO<sub>2</sub>. More recently, Kodros et al. (2018) reported that global annual mean ambient  $PM_{2.5}$ -induced premature deaths associated with household solid fuel usage for the year 2015 were 660,000 (95CI: 520,000–1,000,000), which was 92% higher than our estimate (Figure 8a). The primary driver of the substantial differences between Kodros et al. (2018) and our study is that the household residential emissions in Kodros et al. (2018) included residential coal combustion over east Asia, which emits about 1.9 Tg S for the year 2000 (Kodros et al., 2015). SO<sub>2</sub> from residential coal emissions contributes significantly to sulfate formation, leading to much higher annual PM<sub>2.5</sub> concentrations and associated premature deaths.

At the national scale, annual total  $PM_{2.5}$  and  $O_3$ -induced premature deaths in India for the year 2010 from solid biofuel stove sector were 110,100 (95CI: 100,200-117,200), which agrees reasonably well with Chowdhury et al. (2019), as shown in Figure 8b. However, Conibear et al. (2018b, 2018a) reported that PM2.5-induced and O3- induced annual premature deaths associated with residential energy use for the year 2015 in India were 256,000 (95CI: 162,000-340,000) and 41,000 (95CI: 15,000-61,000), respectively, which were roughly a factor of 2.74 and 2.47 higher than the corresponding averaged annual total PM2.5-induced and O3- induced premature deaths associated with solid biofuel stove emissions in India from our study (Figure 8b). In China, Zhao et al. (2018) estimated that there were about 400,000 (95CI: 250,000-640,000) annual avoided premature deaths for the year 2015 from the solid fuel use attributable to ambient and household  $PM_{2.5}$  exposure (Figure 8c). These results are over 3 times higher than our study. Again, inclusion of residential coal combustion in the residential energy use sector as shown in Conibear et al. (2018a, 2018b) for India and Zhao et al. (2018) for China is the main reason leading to the lower estimates in our study. Moreover, Zhao et al. (2018) considers household (indoor) air pollution as part of the associated PM<sub>2.5</sub> exposure estimates, whereas our study only focuses on ambient PM<sub>2.5</sub> and O<sub>3</sub> exposure. We acknowledge that substantial disease burdens from household PM<sub>2.5</sub> exposure are attributable to global solid biofuel stove sector emissions (WHO, 2018). Health impact assessments often consider both ambient and household exposures to capture the total disease burdens associated with solid fuel sector emissions (Aunan et al., 2018; GBD 2017 Risk Factor Collaborators, 2018; Zhao et al., 2018).

# 4. Discussions and Conclusions

We employ an advanced chemistry-climate model, NCAR CESM CAM5-Chem, at the horizontal resolution of 0.9° latitude  $\times$  1.25° longitude, to quantify the impacts of global solid biofuel stove emissions on surface PM<sub>2.5</sub> and O<sub>3</sub> air quality and the associated premature deaths for the year 2010. Annual mean surface PM<sub>2.5</sub> concentrations associated with the global solid biofuel stove sector reach up to 23.1 µg m<sup>-3</sup> locally in northern India with high concentrations found over China, India, SSA, and ECEurope. Surface O<sub>3</sub> impacts

from global solid biofuel stove emissions are nonnegligible only over significant source regions, including China, India, and sub-Saharan Africa, with annual mean surface  $O_3$  concentrations reaching up to 5.7 ppby. Negative to negligible impacts for surface  $O_3$  concentrations are found over the US, Europe, and part of South America. CAM5-Chem does not explicitly simulate nitrate aerosols, which might, to some extent, lead to uncertainties in surface  $PM_{2.5}$  concentrations for solid biofuel stove emissions (Huang et al., 2020; Liu et al., 2012). However, we expect such effects to be marginal due to relatively low  $NO_x$  emissions associated with the solid biofuel stove sector.

Global PM2.5-induced and O3-induced premature deaths attributable to solid biofuel stove emissions for the year 2010 were about 382,000 (95CI: 349,000-409,000), with the corresponding YLL of 8.10 million years (95CI: 7.38-8.70 million years), which agrees reasonably well with existing literature (Butt et al., 2016; Chafe et al., 2014; Lacey et al., 2017). However, a few studies have estimated much higher global or regional annual total PM2.5-induced and O3-induced premature deaths associated with the residential energy sector (Conibear et al., 2018a, 2018b; Kodros et al., 2018; Lelieveld et al., 2015; Silva et al., 2016; Zhao et al., 2018). This is partially driven by the inclusion of residential coal combustion in the residential energy sector in the above studies. In addition, there are other important factors that likely contribute to the estimated differences in the health outcomes: exposure-response functions, the adoption of different emission inventories, inclusion of household PM<sub>2.5</sub> exposure, differences in model spatial resolution and chemistry schemes, accuracy of model simulated PM2.5 and O3, as well as specific year of health impact assessment. In our study, global PM2.5-induced premature deaths [342,900 (95CI:320,200-364,000 dominate the combined annual total PM<sub>2.5</sub>-induced and O<sub>3</sub>-induced premature deaths for the global solid biofuel stove sector, with mean fractional contribution of 89.8% from the PM<sub>2.5</sub>-induced premature deaths relative to the global annual total PM<sub>2.5</sub>-induced and O<sub>3</sub>-induced premature deaths. Regionally, the highest annual total premature deaths associated with  $PM_{2.5}$  and  $O_3$  from solid biofuel stove emissions are found over China, followed by India, ROA, and SSA, which account for 36.9%, 28.8%, 20.2%, and 5.6 %, respectively, relative to the global totals. YLLs associated with PM2.5-induced and O3-induced premature deaths for the year 2010 in China were the highest among 11 defined regions, which were 2.65 million years (95CI: 2.44-2.82 million years), followed by India [2.55 million years (95CI: 2.35-2.71 million years)], ROA [1.73 million years (95CI: 1.59-1.85 million years)], and SSA [0.55 million years (95CI: 0.43-0.66 million years)]. In this study, we do not quantify the contribution of interannual variability of model simulated surface PM<sub>2.5</sub> and O<sub>3</sub> concentrations during the period of 2006–2010 to the associated annual total premature deaths, which introduces additional uncertainty for the disease burden of global solid biofuel stove emissions.

Replacements of traditional and outdated cookstoves with advanced ones, such as pallet-fed gasifiers and liquefied petroleum gasifiers (LPG), can reduce aerosol and trace gas emissions (Champion & Grieshop, 2019; Jetter et al., 2012) as well as GHG emissions (Bailis et al., 2015). For example, Champion and Grieshop (2019) have reported that an in-field test of pallet-fed gasifiers in Rwanda substantially decreased PM<sub>2.5</sub> and CO emissions, compared with the traditional cookstoves for wood burning. Nevertheless, in terms of human health effects associated with advanced stove emissions, studies often find no significant reductions in air pollution exposures and their associated health impacts (Aung et al., 2018; Hanna et al., 2016; Mortimer et al., 2017; Smith et al., 2011; Tielsch et al., 2016). Despite the fact that advanced solid fuel cookstoves can reduce air pollutant emissions and the associated air pollution exposures, however, emissions from this type of cookstoves are often not reduced down to levels achievable by nonsolid fuels, such as LPG. In addition, PM<sub>2.5</sub> exposure-outcome associations are highly supralinear where health impacts are only substantially reduce at the lowest exposures (Smith & Sagar, 2014). Therefore, we recommend to promote clean household energy programs in low-income and middle-income countries to substantially reduce air pollution exposure and the associated health impacts.

Our study further highlights the cobenefits for air quality and human health of mitigating emissions associated with the global solid biofuel stove sector, especially over populous regions from low-income and middle-income countries (e.g., China, India, and other developing countries in Asia and SSA), where a large share of residential energy relies on solid biofuel stoves for household cooking, heating, and lighting. Our study could provide scientific evidence for policy makers on promoting clean energy associated with the solid biofuel stove sector in order to achieve multifold benefits.



# **Conflict of Interest**

The authors declare no conflicts of interest relevant to this study.

# **Data Availability Statement**

Gridded surface  $PM_{2.5}$  and  $O_3$  concentrations from CESM CAM5-Chem modeling output and the corresponding premature deaths associated with global solid biofuel stove emissions are publicly available at https://doi.org/10.6084/m9.figshare.13356197.v1.

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