Supplementary Material: Global climate impacts of country-level primary carbonaceous aerosol from solid-fuel cookstove emissions

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1 Supplementary Material

S.1 Forward model

The chemical transport model used in this study is GEOS-Chem. This model uses meteorological data from the Goddard Earth Observing System (GEOS) that has been regridded to the 2° × 2.5° resolution [Bey et al., 2001]. Anthropogenic emissions for the simulations are taken from the RCP Database [Moss et al., 2010] hosted by the International Institute for Applied System’s Analysis (http://www.iiasa.ac.at/). For this particular analysis, historical year 2000 emissions are considered [Lamarque et al., 2010]. Anthropogenic emissions are separated into the following sectors: energy, domestic, industry, transportation, landfill waste, agriculture, waste burning, solvents, shipping, grassfires and forest fires. Carbonaceous aerosol emissions from biofuel sources in the domestic sector are from Fernandes et al. [2007] and Bond et al. [2007]. Natural emissions of aerosols and aerosol precursors in GEOS-Chem include volcanoes, lightning NOx [Sauvage et al., 2007; Murray et al., 2012], ocean dimethyl sulfide, soil emissions [Hudman et al., 2012], and biogenic species (isoprene, terpenes) from MEGAN v2.1 [Guenther et al., 2012]. A fraction (20%) of the total biomass burning emissions from the inventory are also considered natural emissions.

GEOS-Chem tracks the formation and transport of the following aerosol species: sulfate, nitrate, ammonium, carbonaceous aerosol (organic and BC), sea salt and mineral dust [Park et al., 2003; 2004]. GEOS-Chem includes heterogeneous chemistry on the surface of aerosols [Evans and Jacob, 2005], aerosol phase partitioning using RPMARES [Binkowski and Roselle, 2003], and aerosol
effects on photolysis rates in the troposphere (Martin et al., 2003). Aerosol removal from the atmosphere includes both dry and wet deposition (Wesely, 1989; Jacob et al., 2000; Liu et al., 2001). Aerosols are considered to be formed or emitted with a fixed log-normal size distribution, while hygroscopic growth is calculated offline using relative humidity retrieved from the GEOS-5 meteorology in each grid cell (Martin et al., 2003). This offline calculation is used when considering surface area for heterogeneous chemistry along with aerosol optical properties. BC is emitted in a 4:1 ratio of hydrophobic to hydrophilic particles, while OC is considered to have a 1:1 ratio of hydrophobic to hydrophilic components. Both of these species have an e-folding time of 1.15 days for their conversion from being hydrophobic to hydrophilic (Cooke et al., 1999; Chin et al., 2002; Chung and Seinfeld, 2002).

Optical properties for aerosols in the model use refractive indices and size distributions retrieved from the Global Aerosol Data Set (GADS) (Koepke et al., 1997). These optical properties are then applied to the GEOS-Chem aerosol mass concentrations in order to calculate aerosol optical depths, single-scattering albedos (SSA) and phase functions (φ) using Mie scattering theory (Martin et al., 2004). For these calculations aerosols are assumed to be an external mixture, which results in underestimating total aerosol absorption (Jacobson, 2001). In order to accommodate for this we increase the absorption of BC aerosol by a factor of 1.5 following recommendations in Bond and Bergstrom (2006) such that the aerosol absorption matches published values. These optical parameters are inputs for the LIDORT radiative transfer model (Spurr et al., 2001) which is used to calculate the top of atmosphere (TOA) radiative flux over wavelengths from 315 to 1,667 nm (Bond and Bergstrom, 2006; Spurr et al., 2001; Spurr, 2001). Radiative flux calculations use surface albedos from remote sensing measurements (Koelemeijer et al., 2003) and cloud fractions from the NASA Global Modeling and Assimilation Office (GMAO).

### S.2 Cookstove emissions

The Bond et al. (2007) carbonaceous aerosol emissions inventory contains all biofuel emissions, which not only includes cookstoves, but other residential emissions sources and additional non-residential biofuel sectors (Fernandes et al., 2007; Bond et al., 2007). The total biofuel emissions
within a country, \( c \), are thus \( \sigma_{\text{biofuel}, c} \)

\[
\sigma_{\text{biofuel}, c} = \sigma_{\text{cs}, c} + \sigma_{\text{ncs}, c},
\]

where \( \sigma_{\text{cs}, c} \) is the carbonaceous aerosol emission from cookstoves and \( \sigma_{\text{ncs}, c} \) is the carbonaceous aerosol emission from non-cookstove sectors. In order to isolate the amount of biofuel aerosol emissions owing to cookstove use, \( \sigma_{\text{cs}, c} \), we must first determine the amount of carbonaceous emissions from other sources, \( \sigma_{\text{ncs}, c} \). We start by considering the percent of the year 2000 population in country \( c \) using solid fuels (\( SF_c \)) for cooking from [Bonjour et al. (2013)]. A number of countries have less than 5% of their population using solid fuel for cooking, or are assumed so in [Bonjour et al. (2013)] based on per-capita income greater than US$12,276. We can use these countries to calculate a non-cookstove use emissions fraction (\( \Theta_r \)) for each region (Africa, Asia, Europe, N. America, Oceanic Pacific, S. America and Central America),

\[
\Theta_r = \left( \sum_{c \in r} \sigma_{\text{biofuel}, c} \cdot (1 - u(SF_c - 0.05)) \right) \left( \sum_{c \in r} P_c \cdot (1 - u(SF_c - 0.05)) \right)^{-1}
\]

where \( P_c \) is year 2000 the population within a country and \( u \) is the Heaviside step function. The numerator therefore represents the sum of biofuel emissions for all countries in the region (\( r \)) that have less than 5% of the population using solid fuel for cooking and the denominator represents the sum of population across those same countries. This gives \( \Theta_r \) in the units of kilograms of non-cookstove biofuel carbonaceous emissions per capita. This equation assumes that \( \sigma_{\text{biofuel}, c} \) is approximately \( \sigma_{\text{ncs}, c} \) for countries with less than 5% of the population using solid fuel for cooking.

With this regional emissions factor calculated we can now calculate \( \sigma_{\text{cs}, c} \) based on the following equation,

\[
\sigma_{\text{cs}, c} = \sigma_{\text{biofuel}, c} - P_c [1 - SF_c] \Theta_r
\]

The inventory of carbonaceous aerosol emission from residential cookstove use per country calculated using this approach is shown in Figure [S.5]. We note that this inventory does not distinguish between solid fuels used for cooking with modern wood-burning stoves versus more traditional...
cookstoves. Non-cooking solid fuel used by the fraction of the population that also uses solid fuel for cooking will be ascribed to $\sigma_{cs,c}$ by this approach, which will thus may be biased high compared to the emissions from cooking alone. The inventory could also be biased low owing to exclusion of fossil-fuel coal used for cookstoves, which is considered as a solid fuel for cooking in Bonjour et al. (2013) but not the biofuel inventory of Bond et al. (2007). Based on national-scale surveys that have been compiled and made available by the World Health Organization (WHO) this source contributes less than 10% of solid fuel use for cooking in most countries, although values in China are notably higher (34%) (http://www.who.int).

Figure S.1: BC and OC emissions factors reported in various biofuel characterization studies: (1) Turn et al. (1997), (2) Sheesley et al. (2003), (3) Streets et al. (2003), (4) Venkataraman (2005), (5) Roden et al. (2006), (6) Johnson et al. (2008), (7) Li et al. (2009), (8) Roden et al. (2009) and the average $\Phi$ limits are calculated from the Bond et al. (2007) biofuel emissions inventory.

These cookstove emissions can be further refined in terms of BC ($\sigma_{BC}$) and OC ($\sigma_{OC}$) emissions using emissions factors for different types of stoves and fuels. A summary of emissions factors used for both OC and BC is shown in Figure S.1. These studies are based on a variety of different fuels, stoves, and measurement techniques. Crop residue fuels tend to have a statistically significant lower BC to total carbon ratio, $\Phi$, compared to woody fuels. While $\Phi$ is different between woody fuels and crop residue, the total carbonaceous emission factors are not statistically different for different fuel types, although some studies (e.g. Roden et al. 2009) have shown that differences between different
measurement techniques (e.g., Roden et al., 2009) can lead to differences in characterizations of both the total carbonaceous emissions and $\Phi$.

S.3 Adjoint model calculations

The GEOS-Chem adjoint model is used in combination with the analytical Jacobians computed by LIDORT to evaluate the impact of grid-scale perturbations to emissions of aerosol and aerosol precursors, $\sigma_{i,k}$, on direct radiative forcing, $J(\sigma_{\tau,i,k})$, where $i$ and $k$ are the spatial and species indices, respectively. We consider both the annual global radiative forcing and the annual regional radiative forcing within four different latitude bands ($\tau$), which are: the Arctic (60N to 90N), the northern hemisphere mid-latitudes (28N to 60N), the Tropics (28S to 28N) and the southern hemisphere extra tropics (90S to 28S). The adjoint sensitivities are calculated in several steps as shown in Equation $\text{S.4}$ (evaluated from right to left),

$$
\lambda_{\tau,i,k} = \left( \frac{\partial J(\sigma_{\tau,i,k})}{\partial \sigma_{i,k}} \right)^T = \left( \frac{\partial B_{\text{conc},i,k}}{\partial \sigma_{i,k}} \right)^T \left( \frac{\partial A_{\text{opt},i}}{\partial B_{\text{conc},i,k}} \right)^T \frac{\partial J(\sigma_{\tau,i,k})}{\partial A_{\text{opt},i}}. \tag{S.4}
$$

where $\lambda_{\tau,i,k}$ is the radiative forcing sensitivity for a given region, $\tau$, with respect emissions of species, $k$, within grid cell, $i$. $A_{\text{opt}}$ is the array of optical parameters (aerosol optical depth, single-scattering albedo and phase function) in GEOS-Chem, which is used as an input for LIDORT, and $B_{\text{conc}}$ is the array of aerosol mass concentrations within GEOS-Chem. In the first step, sensitivities of $J(\sigma_{\tau,i,k})$ with respect to aerosol optical parameters are calculated using LIDORT Jacobians and offline Mie calculations (Henze et al., 2012). These derivatives are then used to force an adjoint calculation, which propagates sensitivity information backwards through the model for all components of the GEOS-Chem governing equations including aerosol thermodynamics, chemistry, deposition and transport.

In order to efficiently calculate adjoint sensitivities of annual radiative forcing, yearly values were approximated using the average of weekly simulations of the first week of each month. All simulations used a one year forward model spin-up using the RCP historical emissions (Lamarque et al., 2010).
S.4 Radiative forcing scaling

Calculations described thus far are only for direct radiative forcing and are based on a single model. However, as discussed in the introduction, there is a range of uncertainties related to direct radiative forcing and other aerosol properties and mechanisms. Therefore, following UNEP and WMO (2011), here we use total forcing estimates calculated across a suite of chemistry-climate models to develop several scaling factors that are applied to our direct radiative forcing calculations to account for three major effects: direct radiative forcing ($SF_{k,DRF}$), aerosol semi and indirect effects ($SF_{k,SI}$), and the forcing from snow-ice albedo changes due to BC deposition ($\lambda_{BC,i,ALB}$). The direct radiative forcing scaling factors ($SF_{k,DRF}$) are created from a comparison of eight to eleven (depending on species) different chemistry-climate models [Myhre et al., 2013; Boucher et al., 2013] and the direct radiative forcing from GEOS-Chem. Many of these models include the radiative forcing for sulfate and nitrate [Myhre et al., 2013], while GEOS-Chem estimates the direct radiative forcing due to emissions of SO$_2$, NO$_x$ and NH$_3$, which lead to the formation of sulfate, nitrate and ammonium. Since separate RF values for ammonium-sulfate and ammonium-nitrate are not reported for the models used in Myhre et al. (2013), here we assume that the direct radiative forcing from emissions of SO$_2$, NO$_x$, and NH$_3$ combine to match the direct radiative forcing estimates of sulfate and nitrate from Myhre et al. (2013), giving a central estimate of $SF_{k,DRF}$ of 0.567. The values for the central estimate and upper and lower bounds for $SF_{k,DRF}$ of all aerosol species are shown in Table 1. Since sulfate acts chemically and physically similar to nitrate with respect to cloud interactions, the same $SF_{k,SI}$ is used for all secondary inorganic precursors, which is based on the effective radiative forcing aerosol-cloud interaction ($ERF_{aci}$) for sulfate from Boucher et al. (2013) divided by the effective radiative forcing aerosol-radiation interaction ($ERF_{ari}$) for sulfate from Myhre et al. (2013). This scaling factor, $SF_{SIA,SI}$, is used for all secondary inorganic aerosol precursors (SIA, Table 1).

Both BC and OC are directly emitted aerosols, therefore $SF_{k,DRF}$ for these species is calculated using the ratio of estimated direct radiative forcing from GEOS-Chem to the direct radiative forcing values in Myhre et al. (2013). This direct radiative forcing scaling is needed to correct for some of the aforementioned assumptions regarding mixing state and BC aging in the GEOS-Chem forward model. BC semi-direct effects ($\pm 0.4$ W m$^{-2}$) are larger in magnitude than the BC $ERF_{aci}$; the
latter are therefore assumed to be negligible. The BC semi-direct effects perturb the upper and lower bounds of the radiative forcing but not the central estimate. Unlike BC, OC has a large indirect component. Since the ERF\textsubscript{aci} is not reported for OC [Boucher et al., 2013], we infer a value from the difference between the modeled ERF\textsubscript{SIA,aci} and the modeled ERF\textsubscript{aci} for all species (Boucher et al., 2013). This assumes that all of the ERF\textsubscript{aci} comes from either OC or the secondary inorganic species, since the BC ERF\textsubscript{aci} is negligible.

Lastly, we implement \(\lambda_{BC,i,ALB}\) to account for the deposition of BC on snow and ice, which leads to a large change in radiative forcing through a decrease in surface albedo. The magnitude of this change (RF\textsubscript{BC,global,ALB}) is approximately 0.15 ± 0.1 W m\(^{-2}\) [UNEP and WMO 2011; Myhre et al., 2013; Bond et al., 2013]. Given the strong spatial dependency of this forcing mechanism, we spatially distribute RF\textsubscript{BC,global,ALB} using deposition sensitivities (\(\lambda_{BC,i,dep}\)) of BC onto snow and sea ice calculated with the GEOS-Chem adjoint model, where \(J_{BC,dep}\) is the global deposition of BC onto snow and sea ice,

\[
\lambda_{BC,i,dep} = \frac{\partial J_{BC,dep}}{\partial \sigma_{BC,i}}. \tag{S.5}
\]

Figure S.2 shows the seasonal variation of grid cells which have snow or ice cover according to the GMAO datasets retrieved from the NASA Langley Research Center Atmospheric Science Data Center (https://eosweb.larc.nasa.gov/). These sensitivities are used to redistribute RF\textsubscript{BC,global,ALB} into a spatially resolved radiative forcing sensitivity (\(\lambda_{BC,i,ALB}\)) by converting the sensitivity of global deposition onto snow and ice with respect to emissions in each grid cell (\(\lambda_{BC,i,dep}\)) as shown in Equation S.6

\[
\lambda_{BC,i,ALB} = \lambda_{BC,i,dep} \frac{0.15 \ \text{Wm}^{-2}}{J_{BC,dep}}. \tag{S.6}
\]

Due to the strong seasonality in \(\lambda_{BC,i,dep}\), these snow-ice albedo sensitivities are calculated for the first week of each month and averaged before being applied to Equation 1. Figure S.3 shows the resulting yearly average for the spatially resolved radiative forcing sensitivities (\(\bar{\lambda}_{BC,i,ALB}\)) in Wm\(^{-2}\) per kilogram BC emitted. This map shows not only the expected higher sensitivities near the poles but also the higher impacts from emissions near higher altitude mountainous regions such as the Himalayas, Alps and Andes. The high sensitivities over central Europe, the western coast of
South America and central Asia show that the radiative forcing per emission of BC in these regions is actually equal to or greater than the direct radiative forcing sensitivities.

Figure S.2: Grid cells containing over 50 cm H₂O equivalent snow depth for a weekly-average noon GMAO estimated snow depth for different months (a) January 2009, (b) April 2009, (c) July 2009, (d) October 2009.

S.5 Results

Table S.1: Model estimates of contribution to total temperature change and emissions metrics for all countries with model inputs of annual emissions from solid-fuel cookstove use (Column 1) and $\Phi$ (Column 2), cooling impact from removal of annual cookstove emissions (Column 3), or efficiency in terms of cooling effect per emission (Column 4 and 5).
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Figure S.3: Spatial distribution of BC radiative forcing sensitivities ($\lambda_{BC,i,ALB}$) for the albedo change from deposition onto snow and sea ice in the units of RF per kg of BC emitted in each grid cell.

Figure S.4: Sample model calculation showing grid-scale contributions to surface temperature change of a 100% removal of biofuel emissions using ARTP calculations. (a) Contributions from removal of BC emissions. (b) Contributions from removal of OC emissions.
Figure S.5: Country-level total annual carbonaceous (BC + OC) aerosol emission due to cookstove use (total emissions for India = 878 Gg C per year and China = 1080 Gg C per year). Countries in grey have less than 5% total population using solid fuels.
Figure S.6: Calculated Φ from cookstove emissions inventory (Fernandes et al., 2007; Bond et al., 2007; Bonjour et al., 2013) on a country-level basis. Lower Φ denotes smaller BC to TC ratio and higher Φ denotes larger BC to TC ratio. Countries in grey have less than 5% total population using solid fuels.
References


