Future anthropogenic land use change impacts on carbonaceous aerosol and implications for climate and air quality

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Key words:

Anthropogenic land use change; carbonaceous aerosol; fire; biogenic VOC; organic aerosol

Key Points:

- Future anthropogenic land use change (LUC) will perturb biogenic and fire emissions, thereby modulating atmospheric carbonaceous aerosol.
- The predicted change in carbonaceous aerosol loading results in moderate future aerosol direct radiative forcing.
- Anthropogenic LUC may be more important than anthropogenic emissions in determining future carbonaceous aerosol burden.

Abstract.

Future anthropogenic land use change (LUC) may alter atmospheric carbonaceous aerosol (black carbon and organic aerosol) burden by perturbing biogenic and fire emissions. However, there has been little investiga(Danabasoglu et al., 2020)tion of this effect. We examine the global evolution of future carbonaceous aerosol under the Shared Socioeconomic Pathways projected reforestation and deforestation scenarios using the CESM2 model from present-day to 2100. Compared to present-day, the change in future biogenic volatile organic compounds emission follows changes in forest coverage, while fire emissions decrease in both projections, driven by trends in deforestation fires. The associated carbonaceous aerosol burden change produces moderate aerosol direct radiative forcing (-0.021 to +0.034 W/m^2) and modest mean reduction in $PM_{2.5}$ exposure (-0.11 $\mu g/m^3$ to -0.23 $\mu g/m^3$) in both scenarios. We find that future anthropogenic LUC may be more important in determining atmospheric carbonaceous aerosol burden than direct anthropogenic emissions, highlighting the importance of further constraining the impact of LUC.

Plain Language Summary

Land is an important source for carbon-containing aerosols – fires emit soot and organics, while forests emit organic gases that form organic aerosols through chemical reactions in the atmosphere. Future anthropogenic activities, including reforestation and cropland expansion, may change the land use type and thus perturb the carbon-containing aerosols in the atmosphere. However, there has been little investigation on this topic. In this work, we study how the carbon-containing aerosol may change from the present to the end of the century under projected reforestation and deforestation pathways using a global climate model. We find that biogenic emissions follow the forest coverage, and fire emissions decrease in both projections due to a reduction in tropical anthropogenic deforestation by the end of the century. The resulting change in carbon-containing aerosol moderately cool or warm the climate at the end of the century because aerosols can scatter and/or absorb solar radiation. It also leads to a modest improvement in global air quality. We highlight the importance of further constraining the impact of future anthropogenic land use change, since we find that it may be even larger than the impact of anthropogenic emissions on the loading of carbon-containing aerosols in the atmosphere.

1 Introduction

Carbonaceous aerosols, including organics (both primary, POA and secondary, SOA) and black carbon (BC), make up 20% to 80% of the fine aerosol mass in the troposphere (Jimenez et al., 2009; Zhang et al., 2007). These particles alter the Earth's climate directly through scattering and absorbing radiation and indirectly through perturbing cloud properties (Chung & Seinfeld, 2002). They also contribute to surface fine particulate matter ($PM_{2.5}$, i.e., particulate matter with diameter smaller than 2.5 micrometers), exposure to which is harmful for human health (Burnett et al., 2018). It is therefore crucial to estimate how carbonaceous aerosol levels will evolve in a future atmosphere.

Human activities have substantially altered global landscapes, converting 37% of land area into cropland and grazing land (Klein Goldewijk et al., 2017). The terrestrial biosphere is an important source of trace gases and aerosols, and thus anthropogenic land use change (LUC), such as forestry management and agricultural development, may critically impact atmospheric composition (Heald & Spracklen, 2015; Ward et al., 2014; Ward and Mahowald, 2015). This represents an underexplored anthropogenic forcing on climate.

Changes to vegetation and fire activity associated with LUC can modulate the sources of carbonaceous aerosol and its precursors. Natural vegetation produces biogenic volatile organic compounds (BVOCs), which oxidize in the atmosphere to form lower volatility products which can condense to form SOA (Hallquist et al., 2009). The emissions of BVOCs strongly depend on the vegetation type. For example, broadleaf trees are stronger emitters than crops and grasses (Guenther et al., 2012). Therefore, deforestation (reforestation) has been found to be associated with decrease (increase) in biogenic SOA concentrations (Heald et al., 2008; Heald & Geddes, 2016; Unger, 2014, Weber et al., 2024). Wildfires emit BC, POA, and SOA precursors. LUC

75 influences fire emissions by altering the above-ground biomass and thus fuel availability (Kloster
76 et al., 2010, 2012) and through socio-economic changes linked to conversion between natural
77 and anthropogenic landscapes, such as savannah to cropland (Andela et al., 2017). In addition,
78 deforestation fires in the tropics are a critical and variable source of emissions (Li et al., 2018;
79 van der Werf et al., 2010), contributing ~ 20% of global carbon emissions in present day (PD)
80 (Li et al., 2013; van der Werf et al., 2010, 2017).

81 Anthropogenic LUC is projected to continue to the end of this century. Recent
82 predictions from the Shared Socioeconomic Pathways (SSPs) suggest that total cropland may
83 expand or contract in the future, accompanied by changes in forests and other natural lands
84 (Riahi et al., 2017). However, there has been little exploration of how future LUC itself will
85 impact carbonaceous aerosols and the attendant climate forcing and air quality. Previous studies
86 suggested that anthropogenic LUC will alter the SOA burden by -20% to 30% by 2100 from PD
87 (Heald et al., 2008; Lund et al., 2021; Wu et al., 2012; Scott et al., 2018; Ward et al., 2014). But
88 these studies either used LUC from old scenarios that only consider deforestation or applied
89 idealized land use perturbations. Kloster et al. (2012) tested different Representative
90 Concentration Pathway (RCP) scenarios and estimated that projected LUC and wood harvest
91 would decrease total fire carbon emissions by 5 to 35%, but they did not extend their results to
92 atmospheric aerosols. Ward et al. (2014) found that LUC could modify fire aerosol emissions by
93 -21% to 21% from 2010 to 2100, depending on RCP trajectories. However, the radiative forcings
94 associated with these LUC-driven fire aerosol changes were not estimated.

2 Methods

2.1 CLM5 and CAM6-Chem

We use the land and atmosphere components of the Community Earth System Model version 2.2 (CESM2.2; Danabasoglu et al., 2020) – Community Land Model version 5 (CLM5; Lawrence et al., 2019) and Community Atmosphere Model version 6 with chemistry (CAM6-Chem; Emmons et al., 2020).

The active biogeochemistry configuration of the CLM5 model used here prognostically calculates leaf area index (LAI). It includes 16 natural plant functional types and 8 active crops, which we grouped into seven land use types (i.e., tropical, temperate, and boreal tree, grass, shrub, crop, and bare soil) in the analysis for simplicity. BVOC emissions respond to light, temperature, leaf age, and CO_2 following the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 (Guenther et al., 2012). A prognostic fire model predicts smoke emissions (Li et al., 2012, 2013; Li & Lawrence, 2017), accounting for four types of fires: agricultural fires in cropland, deforestation fires in tropical closed forest, non-peat fires outside cropland and tropical closed forest, and peat fires. Fire burned area is determined by climate factors, vegetation properties, and human activities. The deforestation fire burned area is also related to the annual loss of tropical tree cover at each grid cell. Fire carbon emissions are estimated considering burned area and surface carbon density; emissions of trace gases and aerosol species are scaled from the fire carbon emissions using emission factors, which are dependent on the vegetation type. Peat fire emissions are unaffected by LUC and thus excluded from our analyses below (Figure S1). This fire model has been extensively evaluated and widely used by previous studies (see Texts S1.1 and S1.2), but we note that further work is needed to

evaluate how well the model responds to LUC perturbations. Species emitted from biogenic and fire sources are tabulated in Table S1.

The CAM6-Chem model includes the MOZART-TS1 chemistry (Model of Ozone and Related chemical Tracers with tropospheric and stratospheric chemistry; Emmons et al., 2020) and a 4-mode version of the modal aerosol model (MAM4; Text S2 and Liu et al., 2016). Three carbonaceous aerosols (i.e., BC, POA, and SOA) are represented in MAM4. BC and POA are emitted from both fire and anthropogenic sources. All the fire emissions are injected into the model surface layer without considering the plume injection height (Val Martin et al., 2010), which is highly uncertain; previous studies disagree on the importance of this factor (Carter et al., 2020; Lu et al., 2024; Tang et al., 2022; Veira et al., 2015). SOA formation from the oxidation of VOCs is parameterized through the volatility basis set that considers NO_x-dependent yields (Hodzic et al., 2016; Jo et al., 2021; Tilmes et al., 2019). VOCs precursors for SOA formation are also shown in Table S1.

2.2 Experiments

We conducted model experiments in two steps: (1) estimating biogenic and fire emissions with evolving anthropogenic LUC using CLM5 and then (2) exploring the climate and the air quality impacts of the LUC-driven carbonaceous aerosol changes using CAM6-Chem.

For the first step, two CLM5 experiments were conducted using land use data following the SSP1-2.6 and SSP3-7.0 pathways (hereafter SSP1 and SSP3; Riahi et al., 2017). These two pathways are chosen because they bracket the range of projected global reforestation (SSP1) and deforestation (SSP3). But we note that in SSP1 (SSP3) there may be local deforestation (reforestation) (Figure S2). Initially, we spun up CLM5 to steady state in 1850 for about 1200 years to reach equilibrium. Subsequently, a spin-up from 1850 to 2004 was conducted using

historical data. The two future simulations were run from 2005 to 2100, with meteorology cycling from 2001 to 2010 following offline dataset, CO_2 concentrations fixed to year 2005, and population density fixed to year 2010. Simulated biogenic and fire emissions were averaged over the first (2005-2014) and last (2091-2100) ten years of the simulations to generate climatologically monthly emission files for PD and future (F) conditions. We note that both SSP1 and SSP3 simulations have the same PD emissions because the land use data are identical before 2015. The annual total fire carbon emission for PD (2720 Tg/yr) falls within the range of Global Fire Emission Database version 4.1s (GFED4.1s; 2027 Tg/yr; Randerson et al., 2018) and Fire Inventory from NCAR version 2.5 (FINNv2.5; 3399 Tg/yr; Wiedinmyer et al., 2023) over the same years with few regional exceptions (Table S2; see Figure S3 and Text S1.3 for comparisons in spatial patterns), which indicates a reasonable representation of the fire emission in the CLM5 fire model.

For the second step, we performed three CAM6-Chem experiments (one PD and two F) driven by the biogenic and fire emissions generated from the CLM5 experiments. Anthropogenic emissions were set to PD climatology (Hoesly et al., 2018) in all the three experiments. By comparing the two F experiments with the PD experiment, we estimate the climate and air quality impacts of LUC-driven carbonaceous aerosol change. In addition, to investigate the relative importance of LUC and anthropogenic emissions, another two CAM6-Chem experiments were conducted using future SSP1-2.6 and SSP3-7.0 anthropogenic emissions (Riahi et al., 2017) but keeping biogenic and fire emissions and climate at PD (see Section 3.4). Anthropogenic emissions related to carbonaceous aerosols include those for BC, POA, and anthropogenic VOCs (Table S1). All simulations were performed for 11 years with the first year used for spin-up at 0.9° × 1.25° horizontal resolution and 32 vertical layers.

In each CAM6-Chem experiment, we added diagnostic radiative calls that remove total carbonaceous aerosol and each carbonaceous aerosol species, respectively, to estimate the effective radiative forcing due to aerosol-radiation interaction (ERF_{ari}; also known as direct radiative forcing) from PD to F. We first calculated the direct radiative effects by taking the difference between default radiative fluxes and those from diagnostic calls that remove corresponding aerosol species. Then, the ERF_{ari} was defined as the direct radiative effect in F with respect to PD. We note that we do not account for the radiative forcing related to aerosol-cloud interactions (i.e., indirect forcing).

3 Result

3.1 Emissions response to LUC

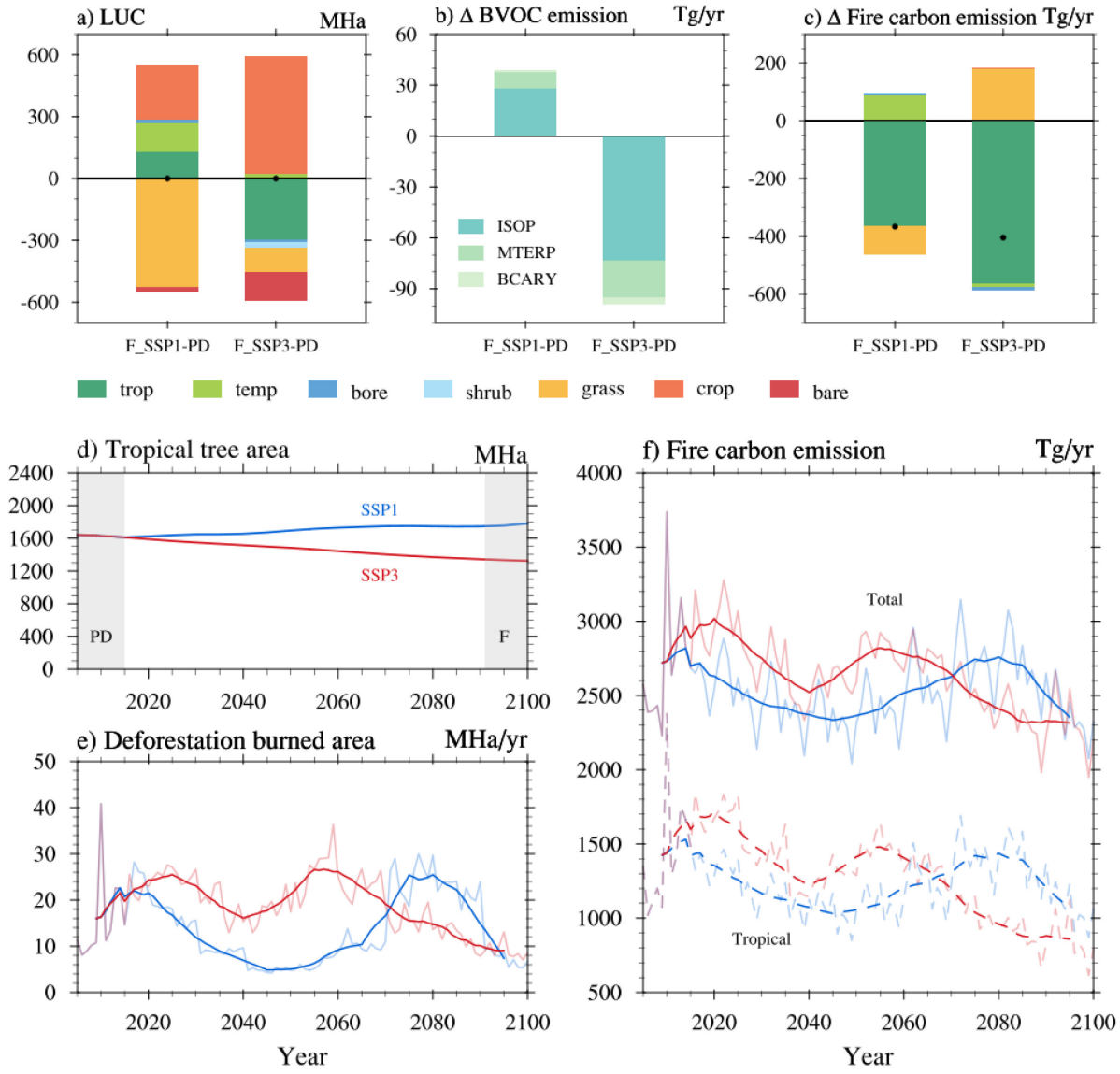


Figure 1. a) LUC b) BVOC (ISOP: isoprene, MTERP: monoterpene, and BCARY: sesquiterpene) emission change, and c) total fire carbon emission change from PD to F projected by SSP1 and SSP3. LUC and fire carbon emission change are shown in land use classes (trop: tropical tree, temp: temperate tree, bore: boreal tree, shrub, grass, crop, and bare: bare soil) and

178 categorized by colors. Black dots on panel a) and c) are global total changes. Panels d-f show
179 SSP1 (blue lines) and SSP3 (red lines) projections of tropical tree area, deforestation burned area,
180 and fire carbon emission, respectively. Gray shading on panel d) marks the time periods treated
181 as PD (2005-2014) and F (2091-2100). Light red and blue lines on panel e) and f) are yearly time
182 series, while darker lines are ten year running means. On panel f), solid and dashed lines are total
183 fire carbon emission and that from tropical tree, respectively. Figure S4 reproduces this figure in
184 a color-blind friendly version.
185
186 We first explore the LUC from PD to F as well as the corresponding changes in BVOC
187 and fire emissions using the CLM5 simulations (Figure 1a-c). The SSP1 scenario projects
188 reforestation over tropical and temperate forest accompanied by cropland expansion at the
189 expense of grassland (Figure 1a). Tropical reforestation is predominantly concentrated in Eastern
190 Brazil and tropical Africa except the Congo Basin, while temperate tree reforestation occurs in
191 the Eastern US, Europe, and Eastern China (Figure S2a-b). In contrast, SSP3 forecasts a
192 deforestation of tropical trees, primarily in tropical Africa and the western Amazon Forest
193 (Figure S2h), along with a moderate global reduction in grassland and bare soil and a substantial
194 increase in cropland (Figure 1a). We observe that, in SSP3, the change in grassland area is not
195 globally uniform – in particular, the African grassland change exhibits a dipole feature,
196 characterized by a significant increase in the Congo Rainforest and the Sahel but a decrease
197 elsewhere (Figure S2l).
198 The global total emissions for BVOCs, BC, and POA in PD and F are shown in Table S3.
199 In response to the reforestation in SSP1 and the deforestation in SSP3, total BVOC emissions,
200 including isoprene, monoterpenes, and sesquiterpenes, increase by 39 Tg/yr (5.6%) and

decreases by 99.3 Tg/yr (14.2%) in SSP1 and SSP3, respectively (Figure 1b). The percentage change is consistent among the three classe(Hodzic et al., 2016)s of biogenic VOCs. The spatial distribution of the change in BVOC emissions is generally positively correlated with the alteration of tree coverage (Figures S2 and S5). In the SSP1 scenario, emissions increase in Eastern US, Southern Africa, Europe, and Eastern China. The Amazon forest also shows slight BVOC emissions increase (mostly < 2%) but has little LUC, which is related to LAI increases (Figure S6). In the SSP3 scenario, the emissions decrease in the Southern Hemisphere (SH; South America and Africa), India, Southeast Asia, and the East Coast of the US, whereas emissions increase in the Midwest and South US, Europe, and Eastern China.

The total fire carbon emissions decrease by 367 Tg/yr in SSP1 and by 404.8 Tg/yr in SSP3 (Figure 1c). This converts to a BC emission decrease of 0.4 TgC/yr (3.7%) in both F projections and a POA emission decrease of 3.4 TgC/yr (5.6%) and 5.5 TgC/yr (9.1%) in SSP1 and SSP3, respectively (Table S3). The fire carbon emissions decrease is dominated by tropical forest in both projections, despite the opposite changes in tropical forest extent (Figure 1c-d). The fire emissions decrease in SSP1 (reforestation scenario) reflects the decrease in tropical deforestation fires, outweighing the growth in fuel availability caused by tropical reforestation (Figure 1e-f). Although global tropical tree coverage increases persistently in SSP1, regional tropical tree loss exists, peaking around 2080 (Figure S7). In SSP3 (deforestation scenario), the tropical tree loss and associated deforestation fires is stronger in the first half of the century, peaking around 2020 and 2055. In the latter half of the century, deforestation fires decline and the overall reduction in forest biomass over the tropics generate less fire carbon loss. Thus, the total F (2091-2100) fire emissions are less than in PD. Our results indicate that around half of the emissions originate from tropical forests (Figure 1f). This may be an overprediction, because

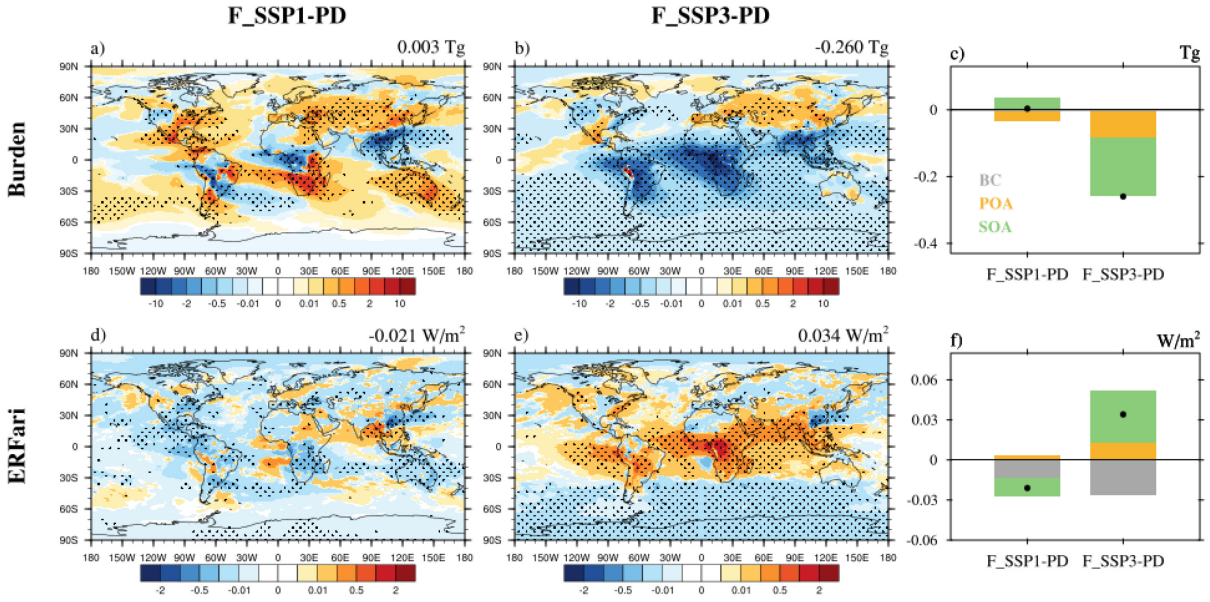
GFED4.1s suggests that tropical savannas account for 62% of total fire carbon emissions (van der Werf et al., 2017). But it is worthwhile to note that satellite-based fire emission products also rely on uncertain factors, like biogeochemical models, land cover datasets, and emission factors. In addition, in the SSP3 projection, the global fire carbon emission from grassland increases despite the global grassland coverage decreases (Figure 1a and 1c). This reflects the heterogeneous response of fire carbon emission to LUC over grassland, which is related to vegetation density (Figure S8).

The fire emissions projected with the SSP1 LUC scenario show declines in the tropics but growth in subtropics and the mid-latitudes (Figure S9). In SSP3, the fire emissions are projected to mostly decrease in the tropics and the SH (Figure S9), in response to the tropical deforestation and grassland shrink. However, there is an emission increase in the Congo Basin, which is the combined result of both deforestation fire and grassland coverage increase. The conversion from grassland into cropland decreases local fire emissions over South Africa and India in SSP3 (Text S1.4). We note that the LUC in the Eastern US, Europe, and Eastern and Southern China are relatively strong in both predictions but the changes in fire emission are minor, which reflects the suppression of fire in these highly populated regions (Text S1.5). Also, both scenarios show emission perturbations in East Siberia (in total, less than 2 Tg/yr) that may be related to disturbance in soil moisture and other factors rather than LUC.

3.2 Carbonaceous aerosol burden change and climate forcing

In the SSP1, the global carbonaceous aerosol burden increases marginally by 0.003 Tg (0.15%), which results from an increase in SOA (0.038 Tg; 4.4%), driven by biogenic emission increases, mostly compensated by decreases in the fire-driven BC (-0.005 Tg; -3.9%) and POA (-0.03 Tg; -3.9%) (Figure 2c). In the SSP3, the burden of all the three aerosol species decreases

247 (SOA: -0.175 Tg, -20.2%; BC: -0.006 Tg, -4.7%; POA: -0.079 Tg, -10.2%), following the
248 decreases in both biogenic and fire emissions (Figures 2c). This leads to a substantial total
249 carbonaceous aerosol burden decrease (-0.26 Tg, -12.6%). The two scenarios result in
250 particularly different trajectories over the SH (Figure 2a-b) – the carbonaceous aerosol burden
251 decreases significantly and consistently in the SH in SSP3, whereas in SSP1, the change is
252 largely positive. Given the relatively short lifetime of aerosols, the spatial distribution of the
253 burden change generally follows local emission changes (Figure S10a-f). We notice that the
254 aerosol lifetime changes slightly from PD to F due to changing geographical distribution of
255 emissions (Table S4).
256



257

258 **Figure 2.** Annual change in total carbonaceous aerosol burden (top row; unit: mg/m^2) and the
259 ERF$_{ari}$ from PD to future (bottom row; unit: W/m^2). Dotted regions on panel a-b and d-e are
260 where the changes are significant to the 0.05 levels. Numbers on the top right of each panel are
261 global total burden (a-b) and global average ERF$_{ari}$ (d-e). Also shown are global total burden (c)

and global averaged ERF_{ari} (f) with contribution from individual aerosol species classified by colors. Black dots on the two panels (c and f) are changes in total carbonaceous aerosols.

Carbonaceous aerosols impact the climate through direct interactions with shortwave radiation – POA and SOA have net cooling radiative effect through scattering shortwave radiation, while BC is a light-absorbing aerosol that has a strong warming effect. We note here that given uncertainties in the sources and photochemical evolution of brown carbon, we neglect the warming effect of this contribution to POA and SOA. This may be somewhat compensated by fire aerosols that are too strongly absorbing (Brown et al., 2021). For SSP1, the global averaged ERF_{ari} due to LUC-driven total carbonaceous aerosol change is -0.021 W/m^2 (Figure 2d), which is dominated by a cooling due to the BC burden decrease and the SOA burden increase that overweighs the warming caused by the POA burden decrease (Figure 2f). In SSP3, the decrease in POA and SOA burden results in a strong warming ERF_{ari} whereas there is a moderate cooling caused by BC burden decrease. As a result, the global ERF_{ari} for total carbonaceous aerosol is +0.034 W/m^2 in SSP3. The warming predominantly occurs over the tropical and subtropical regions in the SH, with regional forcing that exceeds 0.1 W/m^2. We note that the decreasing aerosol burden at mid and high southern latitudes leads to a cooling due to preponderance of POA and SOA above cloud during the poleward transport in the SH. Overall, the future LUC-driven ERF_{ari} of the carbonaceous aerosol in both projections are on the order of ~10% of the total aerosol ERF_{ari} (-0.3 ± 0.3 W/m^2) from 1750 to 2014 estimated by IPCC AR6 (Forster et al., 2021).

We find that SOA has the largest burden change among the three aerosol species in both future predictions. Moreover, the ERF_{ari} from the fire aerosols (i.e., BC and POA) oppose each

other and produce a forcing that is smaller in magnitude compared to that of SOA. Therefore, we conclude that the LUC-driven biogenic emissions have larger impact on carbonaceous aerosol burden and climate forcing than the LUC-driven fire emissions in these simulations.

3.3 Surface $PM_{2.5}$

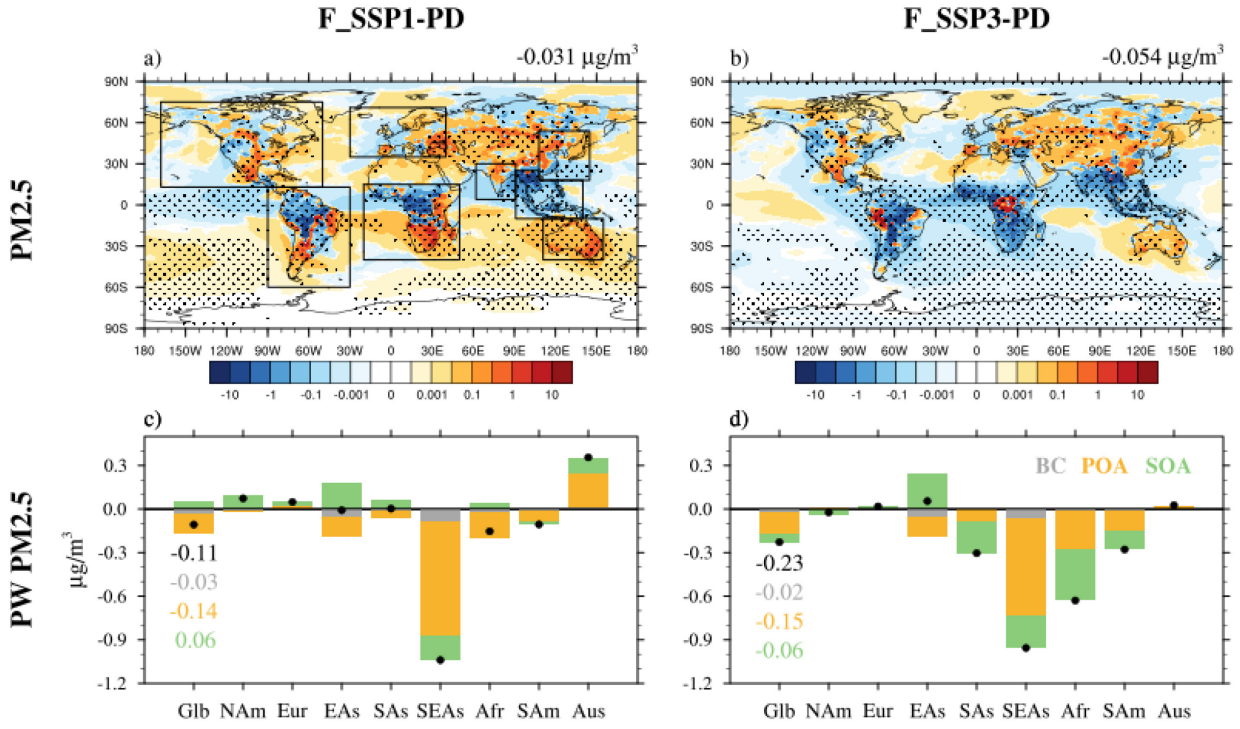


Figure 3. Top row: same as panel a) and b) in Figure 2, except for total carbonaceous aerosol $PM_{2.5}$ surface concentrations. Global (Glb) and regional (NAm: North America, Eur: Europe, EAs: East Asia, SAs: South Asia, SEAs: Southeast Asia, Afr: Africa, SAm: South America, Aus: Australia) average surface population weighted (PW) $PM_{2.5}$ concentrations are shown on the bottom panels. Borders of regions are shown by black rectangles in panel a). Contributions from individual species are classified by different colors, while the black dots represent total changes. Global averaged total PW $PM_{2.5}$ concentrations are given in black numbers, with contribution from each aerosol species shown by numbers in corresponding colors.

LUC-driven changes in carbonaceous aerosol, which contribute to surface $PM_{2.5}$, have implications for human health. Figure 3a-b shows the global distributions of surface carbonaceous aerosol $PM_{2.5}$ concentration change from PD to F. In contrast to the burden increase, global averaged surface $PM_{2.5}$ concentrations decrease by -0.031 $\mu g/m^3$ (-5.1%) in the SSP1 projection. This difference is due to the change in SOA, whose global burden increases (Figure S10c) but surface concentration slightly decreases (Figure S11c). The strongest decrease occurs over the tropics, including the Amazon Forest, mid-Africa, and Southeast Asia. These regions have dense forest and therefore large BVOC emissions (Guenther et al., 2012). As a result, surface OH is depleted and the SOA formation timescale is extended, making surface SOA levels rather insensitive to VOC emission increases. Instead, the fire-driven decrease in POA concentration has a larger impact on SOA by limiting the condensation of precursor gases. Therefore, the change in surface SOA concentration mostly follows the POA change in the tropics (Figure S11b-c). In the SSP3 projection, the surface total $PM_{2.5}$ concentration decreases by -0.054 $\mu g/m^3$ (-8.9%), because of reductions in all three aerosol species. The change in surface SOA is due to changes in both BVOC emissions and surface POA concentrations.

To estimate the impact of the change in carbonaceous aerosols on human $PM_{2.5}$ exposure, we calculate the global and regional population weighted (PW) $PM_{2.5}$ changes (Figure 3c-d). We use 2010 from the Gridded Population of the World, Version 4 dataset (SEDAC, 2018) for both PD and F. Globally, the total surface PW $PM_{2.5}$ concentration decreases in both SSP1 (by -0.11 $\mu g/m^3$) and SSP3 (by -0.23 $\mu g/m^3$). The reduction in both projections is dominated by the change in POA concentrations driven by decreases in fire emission. The global averaged improvement in air quality in both projections is modest comparing to the World Health Organization air quality

guideline level (5 μg/m^3; WHO, 2021), with larger regional improvements (e.g. nearly 1 μg/m^3 over Southeast Asia).

Globally, the fire aerosols drive the reductions in PW $PM_{2.5}$ surface concentrations, both directly and via changing the partitioning of SOA. Therefore, from our simulations we conclude that the air quality response to anthropogenic LUC is dominated by the trajectory of fire emissions. We note that our conclusions here and in Section 3.2 are both subjected to uncertainties related to fire plume injection heights – injecting smoke emissions aloft will reduce the LUC-driven fire PW $PM_{2.5}$ decrease at the surface and may impact aerosol transport and climate radiative effects (See Figure S12 and Text S3).

3.4 Relative importance of LUC and anthropogenic emissions changes

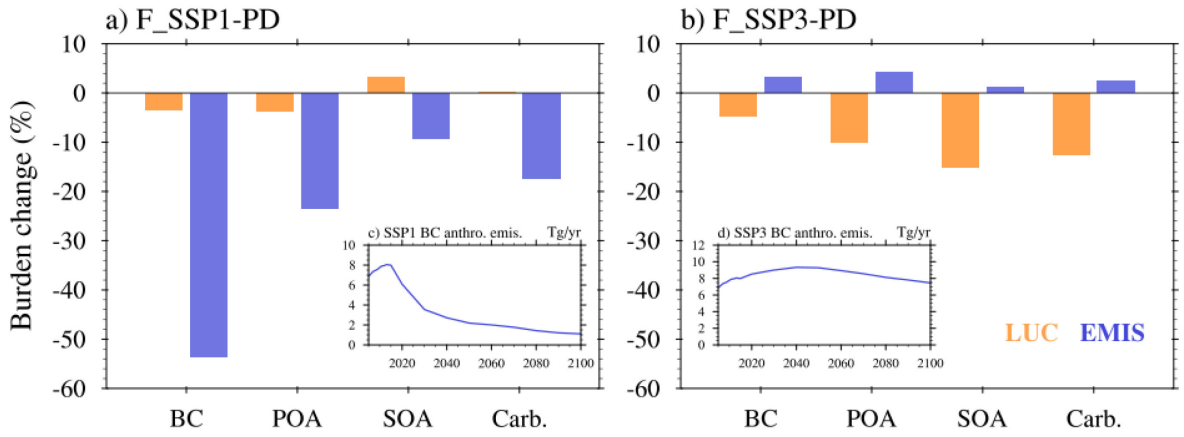


Figure 4. Percentage change in atmospheric carbonaceous aerosol burden in the two future scenarios compared with PD, for a) F_SSP1 and b) F_SSP3. Bars represent the impact of anthropogenic LUC (orange) and anthropogenic emission (EMIS, blue) changes. Inset panels are time series for anthropogenic BC emissions following c) SSP1 and d) SSP3.

Anthropogenic emissions of carbonaceous aerosol are projected to change in the next century in response to socio-economic factors, including air quality regulation. The SSP1 is a strong pollution control scenario, which exhibits a rapid decline globally in anthropogenic emissions (Figures 4c and S13-S14). In contrast, the SSP3 scenario assumes that the implementation of pollution controls is delayed. Therefore, anthropogenic emissions increase slightly over the short-term, except over developed regions, and declines in the second half of the century when strong emission reductions also occur in Asia (Figure S13). As a result, the end of century emissions in SSP3 are comparable to the level at PD (Figures 4d and S14).

Figure 4 compares the percentage change in atmospheric carbonaceous aerosol burden due to anthropogenic LUC and anthropogenic emissions change. In the SSP1 projection, the reduction in anthropogenic emissions has a more substantial impact on carbonaceous aerosol burden than LUC. In contrast, the LUC-driven emission change plays a more important role in the SSP3 projection. Therefore, depending on which pathway we take, the impact of anthropogenic LUC may outweigh the impact of anthropogenic emission changes on the abundance of carbonaceous aerosol and, consequently, the climate forcing and air quality.

Previous studies suggest that climate change will increase future atmospheric carbonaceous aerosols (Heald et al., 2008; Kloster et al., 2012; Sporre et al., 2019). We conducted two additional CLM5 experiments and find that the magnitude of emission changes due to LUC is 25-128% of that due to climate change (Figure S15 and Text S4), highlighting that both climate change and LUC have a substantial impact on carbonaceous aerosol sources.

4. Conclusions

In this study, we investigate the response of carbonaceous aerosols (i.e., BC, POA, and SOA) to future anthropogenic LUC using CESM2. We test two future LUC projections (SSP1-2.6 and SSP3-7.0), which bracket the range of reforestation and deforestation. In response to the forest coverage change, BVOC emissions increase in SSP1 and decrease in SSP3. However, fire emission decrease in both projections. Dynamically varying tropical fires dominate the fire emission trends. Therefore, the fire emissions do not monotonically follow LUC, which emphasizes that the time horizon selected to quantify the effect of LUC is critical. We find that the LUC-driven carbonaceous aerosol emission change by end of century results in a direct radiative forcing that is cooling in SSP1 and warming in SSP3, the magnitude of which are moderate as compared to the total historical ERF_{ari} for aerosol in IPCC AR6. Both future LUC scenarios result in modest improvements in air quality. Overall, with the two future pathways tested in this study, we conclude that the LUC-driven biogenic emissions may have a larger impact on climate forcing, while the LUC-driven fire emissions may be more important for air quality. We note that our study relies on existing land (including fire parameterizations and their relationship to land use change) and atmospheric models and commonly used SSP projections, all of which introduce uncertainties to our results. Given that we find that LUC represents a considerable perturbation to future carbonaceous aerosol concentrations, future research is needed to improve these model schemes and datasets to reduce the associated uncertainties.

Biogenic and fire aerosols are generally overlooked when quantifying aerosol climate forcing (e.g., in IPCC AR6 and the modeling studies it builds upon). Our work demonstrates that LUC is an anthropogenic mechanism that can alter these aerosols, regardless of whether their source is natural or anthropogenic. We find that this pathway may be an even more important

driver than direct anthropogenic emissions in modifying atmospheric carbonaceous aerosol burden and thus in determining the future climate forcing and air quality impact of carbonaceous aerosols. This highlights the importance of including these effects in future climate simulations and improving our understanding of how fire and biogenic emissions respond to changing land use as well as climate.

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Open Research

Datasets used for model evaluation include the GFED4.1 fire emission data (Randerson et al., 2018), the FINNv2.5 fire emission data (Wiedinmyer et al., 2023), and the global population dataset (SEDAC, 2018). This work is based on CESM2.2 (NCAR, 2023). The data shown in the figures is available on Zenodo (Shi et al., 2025).

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