Projected increases in wildfires may challenge regulatory curtailment of $PM_{2.5}$ over the eastern US by 2050

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Abstract

Anthropogenic contribution to the overall fine particulate matter ($PM_{2.5}$) concentrations has been declining sharply in North America. In contrast, a steep rise in wildfire-induced air pollution events with recent warming is evident in the region. Here, based on coupled fire-climate-ecosystem model simulations, summertime wildfire-induced $PM_{2.5}$ concentrations are projected to nearly double in North America by the mid-21st century compared to the present. More strikingly, the projected enhancement in fire-induced $PM_{2.5}$ (~ 1-2 μg/m^3) and its contribution (~15-20%) to the total $PM_{2.5}$ are distinctively significant in the eastern US. This can be attributed to downwind transport of smoke from future enhancement of wildfires in North America to the eastern US and associated positive climatic feedback on $PM_{2.5}$ i.e. perturbations in circulation, atmospheric stability and precipitation. Therefore, the anticipated reductions in $PM_{2.5}$ from regulatory controls on anthropogenic emissions could be significantly compromised in the future in the densely populated eastern US.

Key points:

1) Wildfire-$PM_{2.5}$ associations studied based on unprecedented two-way coupled fire-climate-ecosystem model simulations
2) A steep rise in wildfire-induced air pollution events with recent warming is evident in the region
3) The transported smoke from enhanced wildfires in North America can severely affect air quality over Eastern US

Keywords: wildfire emissions, climate change, air quality, smoke transport, wildfire-climate-ecosystem interactions

1. Introduction

Wildfires are widespread burning events in forests, shrub lands, and grazing lands. In North America (mainly Canada and the US), particulate matter emissions from wildfires are a significant source of regional air pollution (Shi et al., 2019; McClure and Jaffe, 2018; Van Der Werf et al., 2010; Jaffe et al., 2008). Since the 1980s, the number of large wildfires and the length of wildfire season have been increasing, and the trends are projected to continue in the future over the western US, Alaska and Canada (Kitzberger et al., 2017; Kirchmeier-Young et al., 2017; Abatzoglou and Williams, 2016; Partain et al., 2016; Jolly et al., 2015; Westerling et al., 2006; Gillett et al., 2004). Accordingly, particulate emissions from wildfires are also anticipated to increase in North America in the 21st century (Knorr et al., 2017; Liu et al., 2016; Val Martin et al., 2015). Human exposure to high concentrations of wildfire-emitted airborne particulate matter of diameter ≤2.5 μm ($PM_{2.5}$) is known to have substantial adverse effects on pulmonary and cardiovascular functioning (Anjali et al., 2019; Black et al., 2017), which contribute significantly to global and regional all-cause mortality (Zhang et al., 2020; Hong et al., 2019; Yang et al., 2019; Ford et al., 2018; Johnston. et al., 2012). Therefore, a better understanding of the future changes in wildfire-induced $PM_{2.5}$ and its contribution to the total surface $PM_{2.5}$ is essential.

In the last two decades, ambient air quality in the US has substantially improved due to a decline in $PM_{2.5}$ by ~ 40 % (US EPA, 2018). The decrease in $PM_{2.5}$ is primarily due to curtailment of anthropogenic emissions resulting from US-based efforts to meet regulations such as the Clean Air Act (US EPA, 2009), Cross-State Air Pollution Rule, Regional Haze Rule, and the motor vehicles emissions standards. Consequently, air quality over the contiguous US (CONUS) and Canada has improved steadily such that it is predicted to achieve the targeted National Ambient Air Quality Standards in the future (Nolte et al., 2018). Under this promising scenario, the influence of wildfire-emissions on the total $PM_{2.5}$ becomes even more crucial. Depending on the competition between climate-induced increase in wildfires and the regulatory control on anthropogenic emissions, future enhancement in wildfire-induced $PM_{2.5}$ may compromise the reduction in anthropogenic $PM_{2.5}$ concentrations in certain regions. In agreement, recent studies have highlighted the potential for future enhancement in wildfire-induced pollution to diminish the reducing trend in $PM_{2.5}$, primarily over the western US (O'Dell et al., 2019; Ford et al., 2018; Val Martin et al., 2015; Yue et al., 2013).

While the fractional wildfire burnt area and fire intensities are the greatest over the western US and Canadian regions within North America, anthropogenic emissions dominate the ambient $PM_{2.5}$ concentration over the eastern US. The inherent geographical separation between the regions with large wildfire emissions and anthropogenic emissions leads to a pertinent question: will future enhancement in wildfires over the western US and Canada have significant effects on $PM_{2.5}$ over the eastern US? Addressing this question is crucial because the declining trend in $PM_{2.5}$ over the eastern US is the major contributor to the observed 40% decrease in $PM_{2.5}$ over the US in the last two decades (US EPA, 2018). Eastward advection of wildfire smoke from Canada and the western US has been found to severely hamper the surface air quality of the central and eastern US under the influence of the prevailing westerlies during the summer months (Brey et al., 2018; Wu et al., 2018; Gunsch et al., 2018; Kaulfus et al., 2017; Dempsey, 2013). The transported wildfire smoke can influence the meteorology and climate via the radiative impact of carbonaceous emissions, changes in land albedo and cloud system perturbations (Ward et al., 2012; Liu et al., 2014). These fire-weather interactions can have positive feedback on the locally-emitted

$PM_{2.5}$ in the eastern US by surface cooling and boundary layer suppression(Guan et al., 2020). At the same time, fire-triggered ecosystem changes can induce negative feedback on PM2.5 by reducing the future wildfires over North America (Zou et al., 2020). Thus, two-way interactions between fires and climate that are important for predicting future changes in wildfire locations, intensities, and durations (Harris et al., 2016) as well as associated particulate emissions is essential. However, past studies have mostly employed simple statistical models based on statistical regressions of present-day fire burnt area on the meteorological fields (Liu et al., 2016; Spracklen et al., 2009; Yue et al., 2013; Val Martin et al., 2015), and more recently, one-way coupled modelling (Ford et al., 2018; O'Dell et al., 2019).

Here, based on new two-way coupled fire-climate-ecosystem simulations, we demonstrate the significance of wildfire-induced contributions to ambient $PM_{2.5}$ over the eastern US due to enhanced wildfire smoke transportation and smoke-induced changes in weather in eastern US. This enhancement in wildfire-induced $PM_{2.5}$ may potentially challenge the targeted policy-driven reduction of $PM_{2.5}$ in the eastern US. Next, our model setup, experiments and methodology are explained in Section 2, followed by results and discussion in Section 3. The study is summarized in Section 4.

2. Materials and Methods

2.1. RESFire-CESM Model description

We employ the open-source REgion-Specific ecosystem feedback fire (RESFire) model coupled with the Community Land Model version 4.5 and the Community Atmosphere Model version 5 (CAM5) of the Community Earth System Model (CESM) version 1 (Zou et al., 2019; Neale et al., 2013) to perform two-way coupled simulations. RESFire provides state-of-the-art capabilities to simulate the complex fire-climate-ecosystem interactions globally for fires occurring over wildland, cropland, and peatland. Although wildfires dominate in the North American region, RESFire simulates both wildfires and prescribed fires. Moreover, this integrated setup includes climatic feedback from fire-induced aerosol direct and indirect radiative effects and associated weather changes. It also includes feedback from fire-induced vegetation distribution changes and associated biophysical processes such as evapotranspiration and surface albedo. Sofiev et al. (2012) described the fire plume rise parameterization. Other features in CLM4.5 and CAM5, such as the photosynthesis scheme (Sun et al., 2012), the MAM3 aerosol module (Liu et al., 2012), and the cloud macrophysics scheme (Park et al., 2014), allow for more comprehensive assessments of the climate effects of fires through their interactions with vegetation and clouds. Fire-ecosystem interactions are modelled by simulating fire-induced vegetation mortality and regrowth (and associated land cover change) in RESFire. This approach has been introduced in Zou et al. (2019) and the simulated ecological and climatic effects of wildfires have been evaluated in two sets of sensitivity experiments in Zou et al. (2020). Although fire-climate-ecosystem interactions are considered in this study, our focus is on the fire-induced changes in $PM_{2.5}$ over Canada and the US, so the two vegetation-focused sensitivity experiments reported in Zou et al. (2020) are not included in this paper. Please refer to Zou et al. (2019) and Zou et al. (2020) for more details about the simulation of fire-ecosystem interactions.

2.2 Numerical Experiment and Methodology

We designed two sets of simulations for the present day and future scenarios to quantify the impacts of fire-climate-ecosystem interactions (Table 1). The spatial resolution is 0.9° (lat) × 1.25° (lon) with a time step of 30 min. In each set of simulations, we conducted a default all emission included control run (X_{ALL}, where x=2000 or 2050 indicates the present day or future, respectively) and a sensitivity run with no wildfire emissions to the atmosphere

(X_{WEF}, where X is the same as for the control runs). The ALL runs are designed to simulate fully interactive fire disturbances such as fire emissions with plume rise and fire induced land cover changes of the present day (representative of the 2000s, 2000_{ALL}) and a moderate future emission scenario (representative of the 2050s, 2050_{ALL}) via the RCP4.5. The only difference between the ALL and WEF scenario is that wildfire emissions are absent in the WEF scenario. Specifically, in the WEF runs, the online simulated fire emissions are not passed to the CAM5 atmosphere model so that the difference between the ALL and WEF runs can be used to isolate the atmospheric impacts of fire-climate interactions.

Table 1: Summary of the sensitivity simulations performed

Scenario	Present-day		Future	
Experiment Name	2000_{ALL}	2000_{WEF}	2050_{ALL}	2050_{WEF}
Simulation years	2001-2010	2001-2010	2051-2060	2051-2060
Atmosphere	CAM5	CAM5	CAM5	CAM5
Land	CLM4.5	CLM4.5	CLM4.5	CLM4.5
Ocean	Climatology	Climatology	RCP4.5	RCP4.5
Sea ice	Climatology	Climatology	RCP4.5	RCP4.5
Non-fire emissions	ACCMIP	ACCMIP	RCP4.5	RCP4.5
Fire emissions	Online fire aerosols with plume rise	—	Online fire aerosols with plume rise	—
Land cover	Fire disturbances on present-day condition	Fire disturbances on present-day condition	Fire disturbances on RCP4.5 condition	Fire disturbances on RCP4.5 condition

For the present-day experiments, we used the spun-up states from Zou et al. (2019) as initial conditions for both meteorological and chemical variables. Sea surface temperature (SST) for the present day was obtained from the Met Office Hadley Centre (HadISST). Present-day non-fire emissions from anthropogenic and other sources were based on ACCMIP (Lamarque et al., 2010) for the year 2000. We replaced the prescribed GFED2 fire emissions (van der Werf et al., 2006) in the default setting of CESM with the online-coupled fire emissions generated by the RESFire model. Zou et al. (2019) provided more details of the physics parameterizations and modeling experiment settings used in these simulations. Land use and land cover data for 2000 and 2050 from the Land-Use History A product (Hurtt et al., 2006) are used to initialize the $2000_{ALL}/2000_{WEF}$ and $2050_{ALL}/2050_{WEF}$ simulations, respectively. Following the above setup, the future scenario 2050_{ALL} experiment accounts for both fuel load changes associated with the projected land use and land cover change (LULCC) in the 2050s and fire weather changes driven by the SST and sea ice forcing from a coupled CESM simulation following the greenhouse gas (GHG) forcing of the RCP4.5 scenario. The global mean GHG mixing ratios in the CAM5 atmosphere model were fixed at the year 2000 levels in all the present-day experiments and they were replaced by those of the RCP4.5 scenario with the well-mixed assumption and monthly variations. However, the future population and socioeconomic conditions were identical to those of the present day so there was no explicit impact of human-induced mitigation/enhancement effects on wildfires in the future projection in all the future experiments. Future human impacts were considered implicitly in LULCC-induced fuel load changes in the RCP4.5 scenario.

The net projected changes by 2050s in emissions, meteorology and air quality during summer (JJA: June, July, August) months are estimated by comparing decadal-mean values simulated by 2000_{ALL} with 2050_{ALL}. Wildfire-induced enhancement in $PM_{2.5}$ concentration in the present day and mid-21st century is estimated by comparing 2000_{ALL} with 2000_{WEF} and 2050_{ALL} with 2050_{WEF}, respectively. Further, the projected increase in wildfire-induced $PM_{2.5}$ in the future is calculated by comparing the simulated wildfire effect of the 2050s (2050_{ALL}-2050_{WEF}) with that of the 2000s (2000_{ALL}-2000_{WEF}). With large spatiotemporal variability, the projected changes in transported fire-emissions from the western US and Canada to the eastern US by the 2050s and the corresponding impacts are summarized using probability distribution functions. The latter provide information not only for the mean but also variability and extreme values to quantify the simulated changes for the three subregions.

3. <u>Results and Discussion</u>

3.1 Model Evaluation

Zou et al. (2019) performed comprehensive evaluation of the RESFire simulated wildfire burnt area distribution, associated carbon emissions and terrestrial carbon balance to demonstrate reasonable model skill. Zou et al. (2020) compares global fire simulations by CESM-RESFire with modeling results reported in the literature to show better agreement with the GFED4.1s benchmark data and predicts more prominent changes in the future than those predicted by Kloster et al. (2010, 2012). These differences might come from differences in the climate sensitivities of the fire models and scenarios and other input data used to make future projections.

Here, we evaluate the simulated surface $PM_{2.5}$ against satellite-estimates (Figure 1) over North America. The $PM_{2.5}$ concentration is calculated as the sum of sulfate, nitrate, fine sea salt (first 2 size bins), fine dust (first size bin), black carbon (BC), and organic aerosol (OC) at the surface-level of model. OC is the sum of primary organic matter (POM) and secondary organic aerosol (SOA), and SOA is the sum of secondary species formed from toluene, monoterpenes, isoprene, benzene, and xylene. Figure 1 compares the observed and simulated mean annual $PM_{2.5}$ averaged over 2001-2010. The 10-year average satellite AOD-derived annual mean surface $PM_{2.5}$ concentrations (Van Donkelaar et al., 2018) are regridded to the model grid (Figure 1A) and then compared with the RESFire simulations in the 2000_{ALL} present-day run (Figure 1B). The spatial distribution of annual surface $PM_{2.5}$ is reasonably well simulated but also have some biases. To quantify the biases, we also estimated the correlation coefficient as well as normalized mean biases (NMB) of the simulated values compared against the satellite retrieved values over two subregions. Quantitatively, the NMB values over the western US (WUS) and eastern US (EUS) are 18% and 7%, respectively (Figure 1C-D). In addition, the spatial variability of the 2001-2010 averaged annual AOD distribution (Supplementary Figure 1) is also well represented in our simulation, although the model underestimates high AOD values. Similar spatial variability and biases in AOD and $PM_{2.5}$ were also found when a comparison was performed for only summer months (June through August; JJA). The simulated $PM_{2.5}$ has also been evaluated against the ground-based Interagency Monitoring of Protected Visual Environments (IMPROVE) data, showing similar spatial pattern and biases (10-25%) (Supplementary Figure 2). The biases are smaller over Eastern US and Southwestern US region. The simulated PM2.5 values over California matches quite well with the observed annual mean values. However, the biases over Northwestern US region are ~30-40%, a portion of which could be attributed to possible biases in model's meteorology in northwestern US region. Nonetheless, both satellite and in situ evaluation indicate that our simulation biases are largely within the uncertainty range among the various satellite and ground-based datasets, which have normalized mean biases ranging from -3.3% to 33.3% when benchmarked against the ground-based IMPROVE data over the contiguous US (Diao et al., 2019; Val Martin et al. (2015)).

Discrepancies between the simulated and observed $PM_{2.5}$ values may be attributed to several potential reasons. First, the satellite-derived data has a non-zero lower bound of $PM_{2.5}$ concentrations, so the ambient background concentrations for relatively cleaner regions such as the western US may be overestimated (Figure 1C), also the sampling frequency between these datasets are different. Second, year 2000-based constant non-fire emissions were used in the RESFire simulation, which may result in overestimation of the $PM_{2.5}$ concentrations from non-fire sources during 2001-2010 when anthropogenic emissions and $PM_{2.5}$ concentrations continue to decrease (US EPA, 2018). This overestimation is prominent in regions dominated by non-fire sources such as the southeastern US. Third, large uncertainties in fuel consumption and emission factors preclude an accurate estimation of the primary fire emissions in the model, especially for the eastern US where large fractions of low-intensity prescribed fires consume only under-canopy fuels such as litter and duff layers. The fire model may fail to capture the subtle distinctions between low-intensity prescribed fires and forest fires, so more fuels are consumed and result in higher emissions. Lastly, comparison of a coarsely resolved simulation against in-situ observations also contributes to uncertainty. Differences in the degree to which fire-climate interactions and other physical processes and feedbacks are represented by the models can explain the slights differences in estimating the present day mean wildfire-induced change in $PM_{2.5}$ over local and downwind regions between our simulations and previous studies. Nonetheless, reasonable simulation of the spatial distribution of wildfire burnt area, AOD, and near surface particulate concentration (mean bias of ~10-20 %) instills confidence about the fidelity of our model setup in particulate pollution simulation, which is the focus of this study.

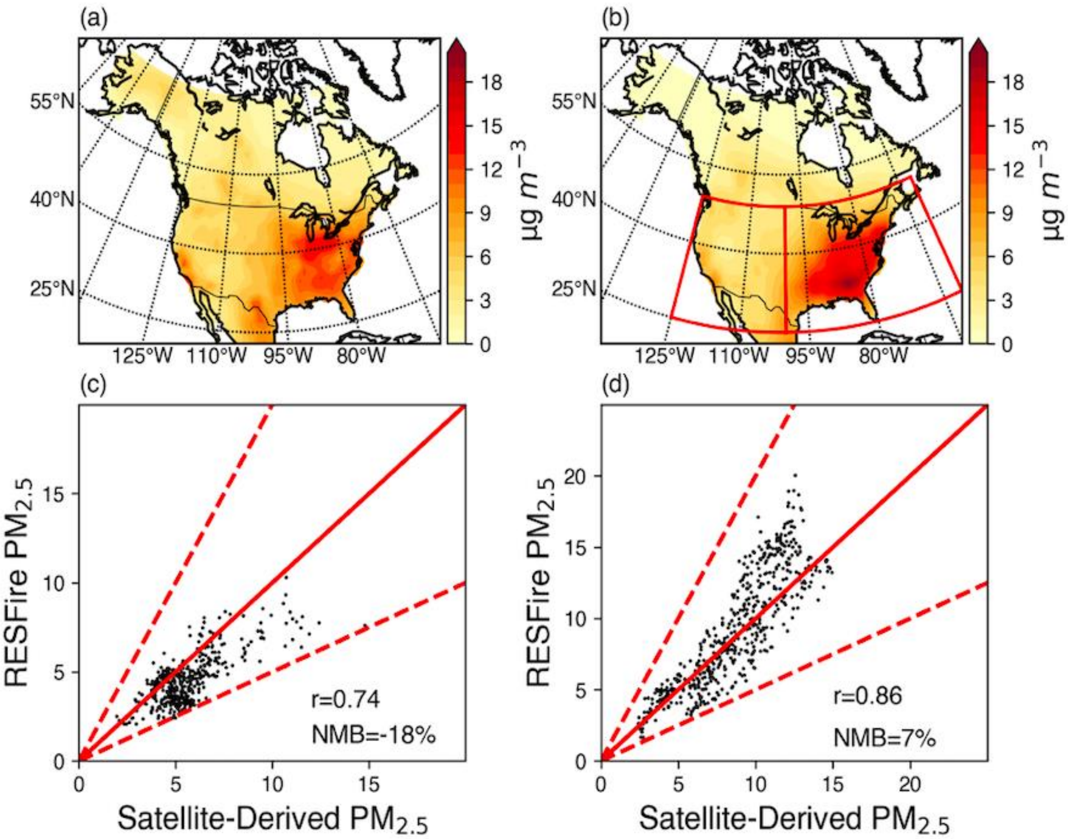

Figure 1: Comparison of the 10-year (2001-2010) averaged annual mean surface $PM_{2.5}$ concentration between observations and RESFire simulations. (a) Satellite-derived surface $PM_{2.5}$ concentrations (with dust and sea-salt removed) estimated by Donkelaar et al., 2018 (available at https://sedac.ciesin.columbia.edu/data/set/sdei-global-annual-gwr-pm2-5-modis-misr-seawifs-aod; last access: 5 November, 2021); (b) 2000_{ALL} Simulated surface $PM_{2.5}$ concentrations (with dust and sea-salt removed) averaged over 2001-2010; The red boxes denote the two subregions (EUS and WUS) shown in Fig. 2 in the main text. (c) comparison of simulated and satellite based gridded surface $PM_{2.5}$ concentrations in the WUS subregion; Number of samples is equal to the number of land grids ~450 (d) same as (c) but in the EUS subregion. Number of samples is equal to the number of land grids ~375 The red solid and dashed lines denote the 1:1 ratio line and ±100% biases, respectively. The correlation coefficients and NMB values are shown at the lower-right corner of each subplot.

3.2 Fire-induced changes in burnt area and $PM_{2.5}$

The decadal-mean annual fire burnt area simulated for the present day shows widespread wildfires over the entire North America (Figure 2A). Specifically, Canada and the forested areas of the northwestern (> 36 N latitude) and southeastern (< 36 N latitude) US are most intensely affected by wildfires in the present day. By the mid-21st century, a striking increase of 2-5 times in fire burnt area is projected over Canada, Alaska, the Pacific Northwest and portions of the western US by the 2050s (Figure 2B). A distinct positive shift in the probability density function (PDF) of annual fire burnt area is evident in the future, with the decadal-mean difference statistically significant at the 99% confidence level (Zou et al., 2020). A small and statistically insignificant change in interannual variability (~ 0.4 Mha yr^{-1}) of fire burnt areas is also simulated between the present and future. Specifically, our model predicts more than a doubling of burnt area in boreal regions of Canada in the future, in line with a previous projection for Canada (Wotton et al., 2017). Future enhancement in fire burnt area is ~ 20-50% in most fire grids over the western coast of US, which is higher than that over the eastern US (Figures 2A and 2C). The increase over the western US is closer to the lower bound of that derived from statistical model ensemble projections for the western US in the mid-21st century (Yue et al., 2013). The statistics-based projections of future burnt area over North America were likely too high because fire-induced land cover change, fuel load reduction and factors could induce a negative fire feedback, which was not considered in previous fire projection studies (Zou et al. 2020).

Annual fire burnt area in the southeastern US shows a decline in the future (Figure 2C), as precipitation is projected to increase in that region (discussed later). Note that all future fire changes between 2050_{ALL} and 2000_{ALL} are primarily associated with climate warming in response to the increase in greenhouse gas (GHG) concentrations in the RCP4.5 scenario. No direct impacts of population and socioeconomic changes on wildfires are included in our simulations, although these factors contribute to changes in GHG emissions (via the RCP scenario) that influence the climate simulated in 2000_{ALL} and 2050_{ALL}. As about 80% of the projected fire changes in the future is restricted to the summer season (June through August; JJA) (Figure 2D), we focus on analysis of the summer-mean wildfire-induced $PM_{2.5}$ and its projected future changes over North America.

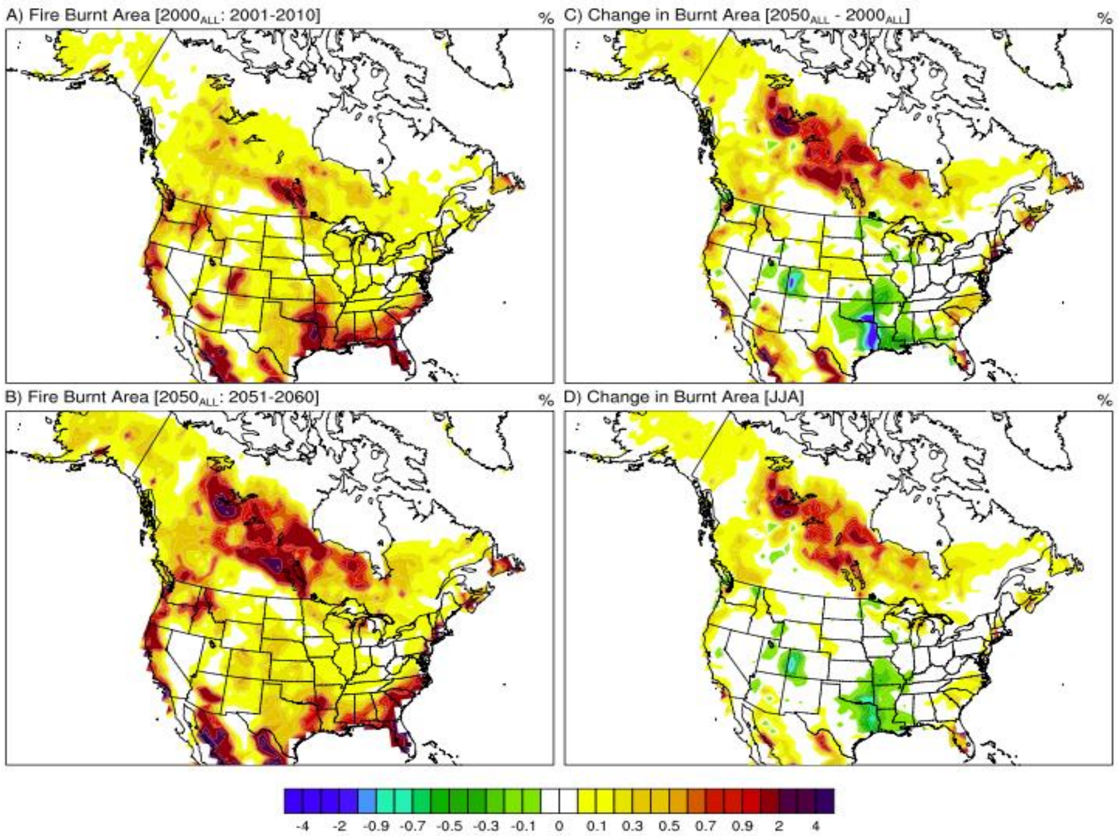

Figure 2: **Spatial distribution of fire burnt area. A-D**, Spatial distribution of simulated decadal-mean annual burnt area (as percentage) over North America for present day (A), mid-21st century (B) and the net change between the 2050s and the 2000s (C). **D**, same as (C), but for wildfire burnt area during summer only (June through August; JJA). The colorbar illustrate grid fraction of area burnt.

The simulated 10-year averaged summer-mean wildfire-induced $PM_{2.5}$ values in 2000_{ALL} are more than 0.5 µg/m^3 over a large part of North America in the present day, with noticeably larger values (> 1 µg/m^3) in Canada and the northwestern, central, and southeastern US (Figure 3A). Interestingly, the spatial distribution of wildfire-induced $PM_{2.5}$ > 1 µg/m^3 resembles an inverted horse-shoe shape. The inverted horse-shoe shaped spatial distribution is also consistent with the wildfire-smoke climatology derived from the satellite-guided operational smoke product of the Hazard Mapping System (HMS) during 2005-2015 (Brey et al., 2018; Kaulfus et al., 2017). By the mid-21st century, the spatial extent of the horse-shoe shape for areas with wildfire-induced $PM_{2.5}$ enhancement > 1 µg/m^3 expands significantly to span most regions of North America, with the most pronounced enhancement occurring over Canada (Figure 3B). The PDFs of the spatial distribution for the three regions can be seen in Figure 3C-E. Specifically, wildfire induced $PM_{2.5}$ in the 2000s over Canada, WUS and EUS during summer is ~ 1-3 $\mu g/m^3$, 1-3 $\mu g/m^3$ and 0.6-1.2 $\mu g/m^3$, respectively. Maximum values within the WUS region are found over the Pacific Northwest, with most areas having wildfire induced $PM_{2.5}$ values of ~ 2-3 $\mu g/m^3$. Similarly, the southern states have relatively high wildfire induced $PM_{2.5}$ concentrations of ~ 2-4 $\mu g/m^3$ within the EUS in the present-day simulation.

Compared to the 2000s, the wildfire induced JJA-averaged $PM_{2.5}$ values are almost doubled to ~ 3-6 $\mu g/m^3$ over Canada in the 2050s (Figure 3B and Figure 3C). Consistently, the values of wildfire induced PM2.5 over WUS (mainly coastal) also doubled in 2050s compared to 2000s, with modal values of ~ 2-2.5 $\mu g/m^3$ (Figure 3D). Most interestingly, the enhancement in wildfire-induced summer-mean $PM_{2.5}$ over the northern EUS is also significant by the 2050s (Figures 3B). Largely, the summer-mean wildfire-induced $PM_{2.5}$ concentration over EUS increases from ~0.8 to ~2 $\mu g/m^3$ in the mid-century to values of 1.2-3.0 $\mu g/m^3$ (Figure 3E). The summer-mean wildfire-induced $PM_{2.5}$ is thus projected to double in North America by the 2050s compared to the 2000s, with a substantial coverage over the EUS. An important finding from these PDFs appears to be that there are fewer grids with < 1 $\mu g/m^3$ wildfire induced PM2.5, or alternatively, that more regions are being influenced by $PM_{2.5}$, and many areas that were already seeing wildfire impacts are seeing enhanced impacts. Such enhancement is found not only at the surface but also in an elevated atmospheric layer over EUS between 900 and 700 hPa. This is nonintuitive given the fact that the increase in fire-burnt area by mid-century over the EUS is not substantial.

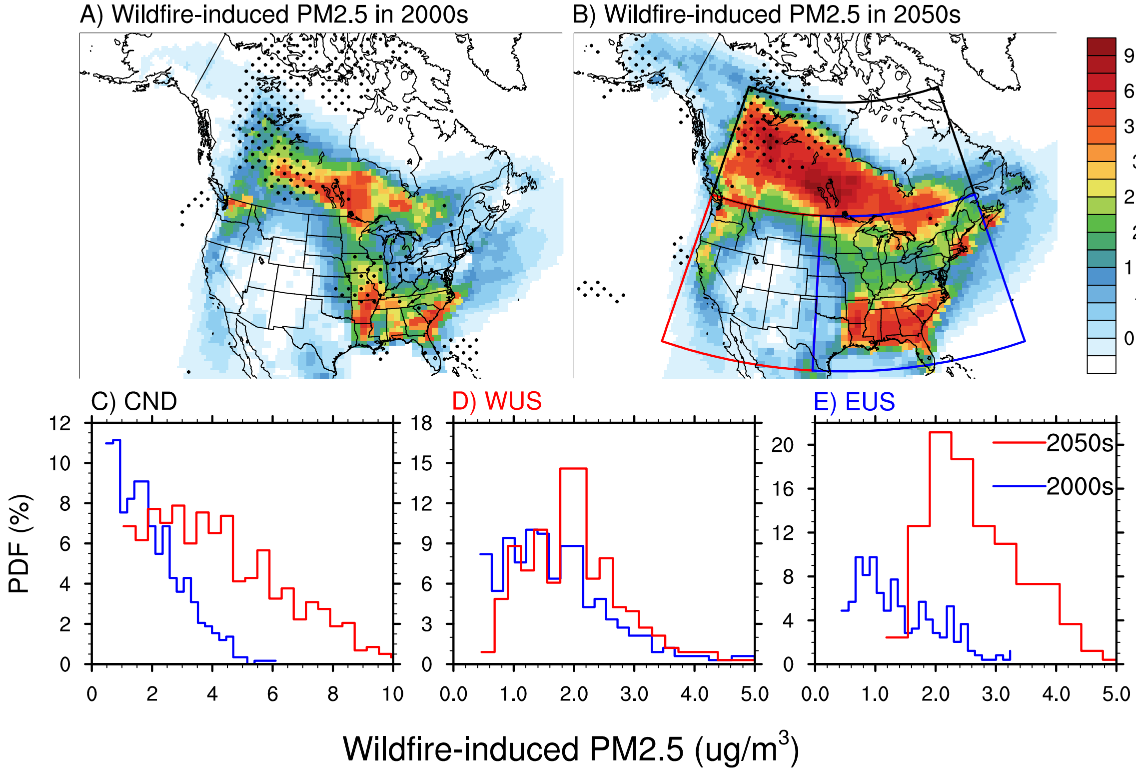

Figure 3: **Spatial distribution of** $PM_{2.5}$ **concentrations.** Spatial distribution of decadal-mean wildfire-induced enhancement in summer (June through August; JJA) $PM_{2.5}$ concentration over North America for present day (A, 2000_{ALL}-2000_{WEF}) and future (B, 2050_{ALL}-2050_{WEF}). The grids with statistical significance of 90% is identified by black dots. **C-E**, Probability density functions (PDFs) of wildfire contribution within the three regions shown in Figure 2B for Canada (CND: black box) (C), WUS (red box) (D), and EUS (blue box) (E), respectively, for 2000s (blue) and 2050s (red). Only grids over land in North America are used to generate the PDFs. The y-axis indicates the probability of occurrence of different $PM_{2.5}$ values shown in the x-axis. The colorbar illustrates PM2.5 in ug/m^3.

As anthropogenic- and wildfire-induced $PM_{2.5}$ concentrations may change differently with time across North America, next, we investigate the relative contribution of wildfire-induced $PM_{2.5}$ to the total $PM_{2.5}$ in the future. Prominent enhancement of the wildfire contribution is apparent in the entire domain by the 2050s (Figures 4A-B). Largely, during the 2000s, the simulated fractional contribution of wildfires to $PM_{2.5}$ is ~15-50 % in Canada (Figure 4A). Specifically, a bi-modal distribution is simulated over Canada with modal values around 18% and 30% (Figure 4C). Over WUS, the present day simulated percentage contributions of wildfire-induced values are 5-25% (Figure 4A), with modal values between

10-20% (Figure 4D). Note that many areas located in the Pacific Northwest have higher values of ~30-40% (Figure 4A). At the same time, the fractional contribution by wildfire-induced $PM_{2.5}$ is ~5-10% in most areas of EUS in present day (Figure 4F). Nevertheless, some areas in the central US also have higher values of ~10-25% (Figure 4A).

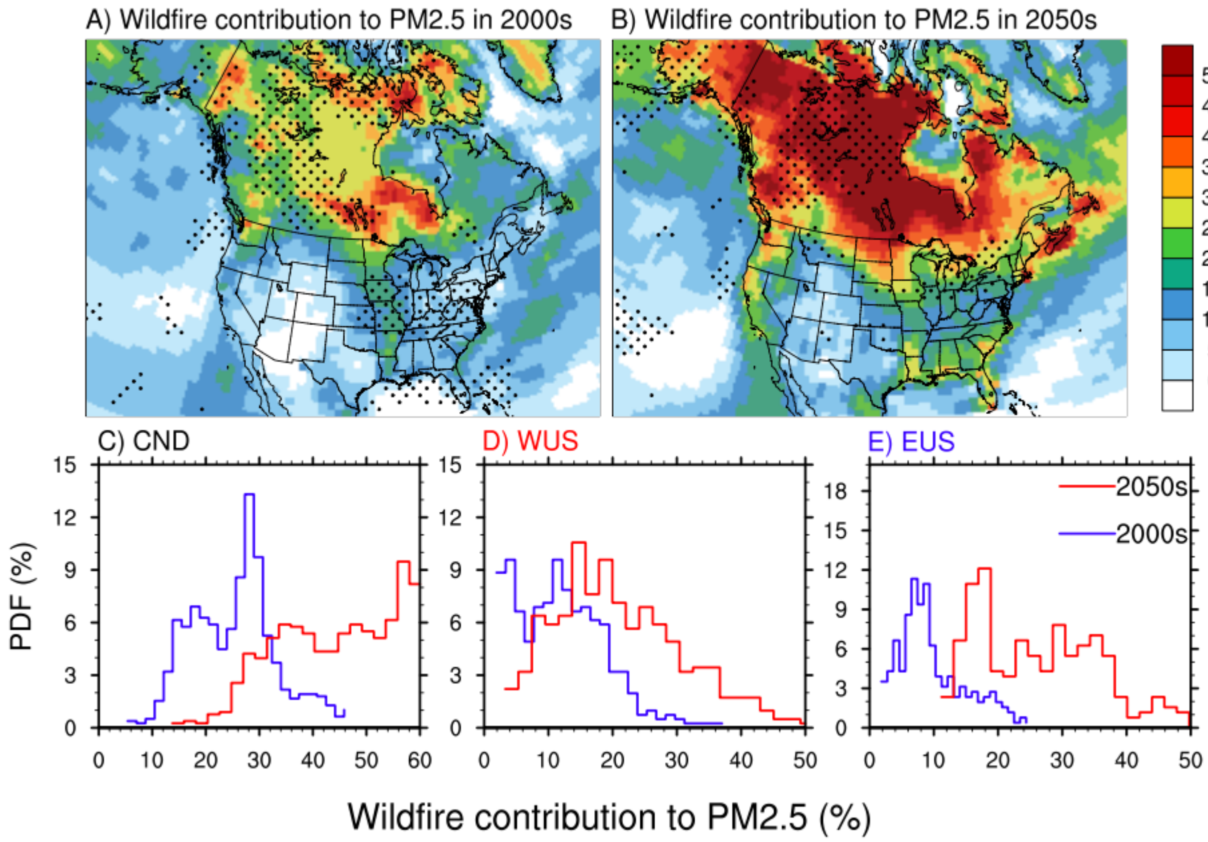

Figure 4: **Spatial distribution and probability density function of the percentage contribution of wildfire emissions. A-B**, Spatial distribution of the percentage contribution of wildfire emissions to decadal-averaged summer (June through August; JJA) mean $PM_{2.5}$ concentrations over North America during present day (A) and future (B). The percentage contribution of wildfire-induced $PM_{2.5}$ to the total $PM_{2.5}$ concentrations is calculated as ($[2000_{ALL}-2000_{WEF}]/2000_{ALL}$) and ($[2050_{ALL}-2050_{WEF}]/2050_{ALL}$) for the present and future, respectively. The grids with statistical significance of 90% is identified by black dots. **C-E**, Probability density functions (PDFs) of the percentage wildfire contribution within the three regions shown in Figure 2D for Canada (CND: black box) (C), WUS (red box) (D), and EUS (blue box) (E), respectively, for the 2000s (blue) and the 2050s (red). Only grids over land in North America are used to generate the PDFs. The y-axis indicates the probability of occurrence of different $PM_{2.5}$ values shown in the x-axis.

The wildfire contributions in the 2050s show a clear shift towards higher values in all sub-regions compared to the 2000s (Figure 4B). Over Canada, the values shifted from 15-30 % in the 2000s to ~30-60% in the 2050s, a nearly two-fold increase in the fractional contribution of wildfire emissions to the total $PM_{2.5}$ concentration is simulated (Figure 4B and corresponding PDF in Figure 4C). Similarly, the contribution values increased to ~ 10-35 % in the 2050s, compared to 10-20% in the 2000s over WUS (Figure 4B), thereby featuring a broadening of the bi-modal distribution of wildfire contribution (Figure 4D). The shift in the percentage contribution is most prominent for the higher values, corresponding to some areas located in the Pacific Northwest and west coast of the US (Figure 4B). Consistent with Figure 3B, the shift in the contribution values over EUS is also very distinct, revealing an increase in the mode values from 6-10% in the 2000s to ~16-20 % by the 2050s (Figure 4B and Figure 4E). Thus, not only in absolute values, but our results also underscore a large increase in the contribution of wildfire emissions over EUS in the future.

3.3. Mechanistic understanding of the underlying processes

The larger enhancement in the relative contribution of wildfire emissions to the total surface $PM_{2.5}$ in EUS in the 2050s can be explained by three mechanisms. First, due to the increase in Canadian and western US wildfires, downwind transport of wildfire smoke plumes to EUS will be enhanced by the 2050s. This long-range transport to the atmospheric column of EUS can happen within a few days of the fire occurrence (Supplementary Figures 3A and 3B). Using Hazard Mapping System (HMS)-detected smoke plumes, recent studies identified a strong positive association between the transported smoke plumes in the atmospheric column and collocated surface PM2.5 enhancement in EUS (Brey et al., 2018; Wu et al., 2018; Gunsch et al., 2018; Kaulfus et al., 2017; Larsen et al., 2017; Dempsey, 2013). Hazard Mapping System (HMS) is an operational smoke detection product over North

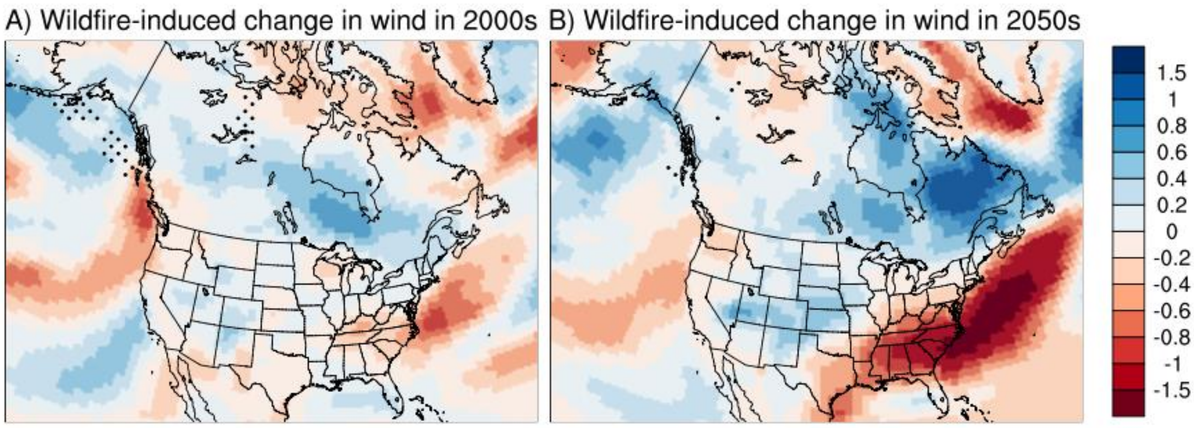

Figure 5a: Spatial distribution of decadal mean summer (June through August; JJA) wildfire-induced future changes [(2050_{ALL}-2050_{WEF}) – (2000_{ALL}-2000_{WEF})]. A) Wind speed below 850 hpa for [(2050_{ALL}- – 2000_{ALL}], B) Wind speed below 850 hpa [(2050_{WEF}) – (2000_{WEF})]. The unit is m/s. The grids with statistical significance of 90% is identified by black dots.

America known as developed by the National Oceanic and Atmospheric Administration (NOAA) and operated by National Environmental Satellite, Data, and Information Service (NESDIS), available at http://satepsanone.nesdis.noaa.gov/FIRE/fire.html. Specifically, these studies found that the smoke plumes transported from Canada are located at an altitude of ~ 1-3 km over EUS (Colarco et al., 2004; Wu et al., 2018). Due to mixing by the daytime boundary layer and deposition, the smoke plumes enhance the surface $PM_{2.5}$ concentration over EUS (Wu et al., 2018; Colarco et al., 2004; Rogers et al., 2020; Dreessen et al., 2015). Hence HMS smoky days may be a useful proxy for wildfire-induced surface PM2.5 over North America. In agreement, Brey et al. (2018) showed that the HMS-based smoke plumes observed over EUS is significantly aged, suggestive of their long-range transport origin. Consistent with the observed temporal change in HMS pattern, Xue et al. (2021) estimated using the mid-visible Multi Angle Implementation of Atmospheric Correction (MAIAC) satellite-derived Aerosol Optical Depth (AOD) that Canadian and western US fires have caused an increase in the daily $PM_{2.5}$ over Montana, North Dakota, South Dakota and Minnesota by 18.3, 12.8, 10.4 and 10.1 $\mu g\ m^{-3}$, respectively, between August 2011 (a low fire month) and August 2018 (a high fire month). In summary, the visually apparent satellite-based signatures of wildfire-smoke across Canada and EUS provide a necessary, though not sufficient, support for the influence of Canadian smoke plumes on EUS air quality. Although, the change in burnt area over northeastern EUS is negligible compared to the western US and Canadian regions, however, there are some enhancements seen over east coast of US, which can also contribute to enhanced fire emissions.

In future, the wildfire-induced change in speed of the westerly jet flows over Canada wildfire regions is increased (Figure 5 A-B). It indicates that the westerly-induced transported wildfire emissions from Canada boreal forests to the eastern half of Northern America and EUS will be enhanced in future compared to that in present era. On the one hand, the wildfire-induced changes in wind speed over the EUS is reduced in future, which implies that the local emissions over EUS are less dispersed. Simultaneously, this will also cause the transporting smoke plumes to slow down and be subjected to relatively more boundary layer mixing over the EUS and dry deposition/settling enhances, thereby contributing to the enhanced PM2.5 values at surface. The westerly winds over western US below 40^{o} N is also strengthened in future (Figure 5 A-B) compared to present day which indicates more advection flux wildfire emissions to EUS. Thus, the net effect is more removal of wildfire-emitted PM2.5 from WUS and more influx of wildfire-emitted PM2.5 in EUS.

Along with this dynamical changes, other climatic feedbacks simulated can also contribute to enhancement of EUS pollution. Specifically, the enhancement of wildfire-induced smoke aerosols increases solar absorption and scattering in the future (Figure 6A). This reduces the incoming solar radiation reaching at the surface (Figure 6B) and induces surface cooling. With atmospheric warming and surface cooling, lower-tropospheric stability is enhanced by wildfire aerosols in the future (Figure 6C). The smoke plumes which reaches eastern US are at an elevated altitude due to self-lofting property of absorbing aerosols as they travel downwind but the smoke over western US is at near surface elevation as it is at its source region. This can explain the more significant atmospheric stability simulated over the eastern US compared to the source regions in western US and boreal forests of Canada. Relatively stronger atmospheric stability over eastern US impose a stronger thermal capping that traps more anthropogenic aerosols and particulate matter near the surface over EUS (already an emission hotspot). At the same time, future increase in wildfire emissions also leads to greater reduction of monthly rainfall (Figure 6D) over EUS, which may additionally strengthen the positive feedback to surface PM2.5 over EUS by reducing wet scavenging of transported wildfire smoke to EUS. Thus, wildfire-emitted aerosols induce positive feedback on the surface PM2.5 concentration over EUS through fire-climate interactions that vary on a regional scale. Moreover, the above discussed dynamical changes in future can also feedback these simulated thermodynamical and precipitation changes, exaggerating the enhancement in PM2.5 values over EUS in future. However, due to computational constrains, no direct quantification of the magnitude of these feedback (with aerosol-radiation and aerosol-cloud interactions turned off) on PM2.5 is performed and would be taken up in future studies.

Lastly, the reason of why the contribution of wildfire emissions to the total surface $PM_{2.5}$ in EUS is so substantial in the 2050s is the drastic reduction of anthropogenic contribution to the surface $PM_{2.5}$ over EUS in the future primarily due to policy-driven reduction in anthropogenic emissions under the RCP4.5 scenario. Specifically, the simulated ambient summer mean $PM_{2.5}$ concentration exhibits widespread declines in the future (Supplementary Figure 4), with reduction in $PM_{2.5}$ concentration over eastern US in the range of 4-15 $\mu g/m^3$, which is greatest within North America. Thus, large reduction in anthropogenic contribution combined with increased downwind advection of Canadian smoke and WUS to EUS and the associated positive feedbacks can explain the projected dominance of wildfire emissions over EUS in future.

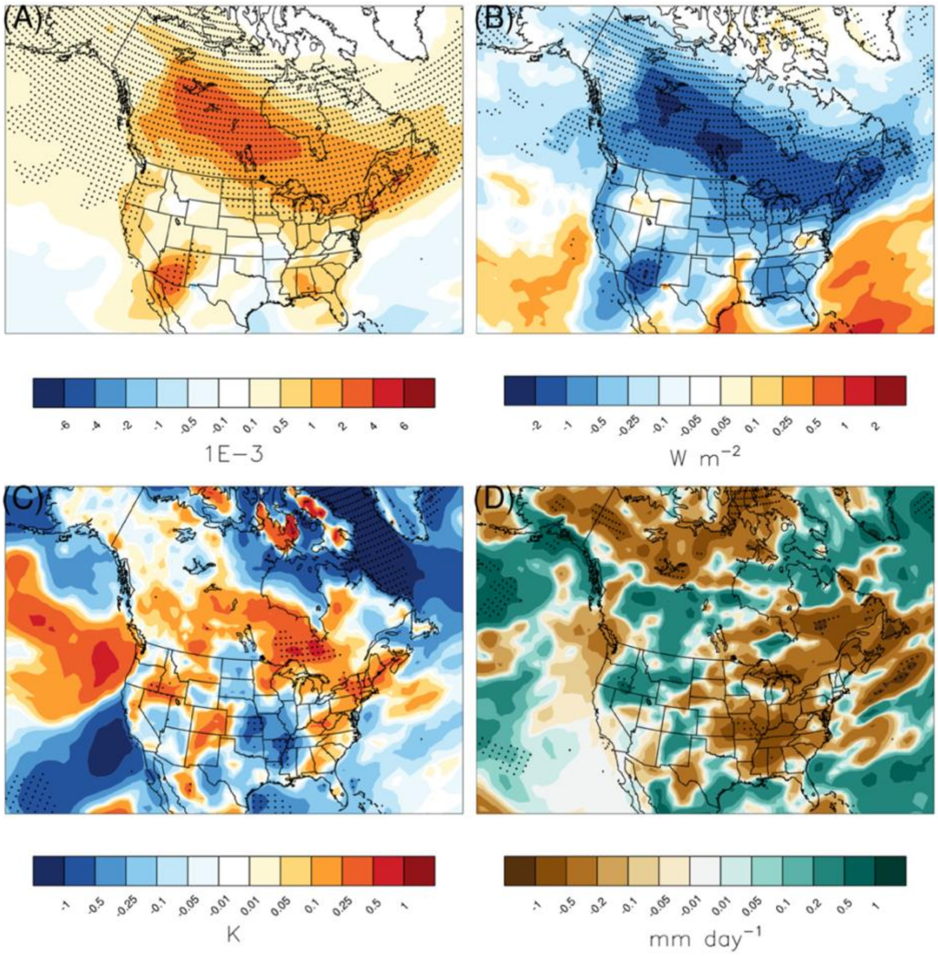

Figure 6: Spatial distribution of decadal mean summer (June through August; JJA) wildfire-induced future changes [(2050_{ALL}-2050_{WEF}) – (2000_{ALL}-2000_{WEF})]. A) aerosol absorption optical depth at 550 nm, B) aerosol direct radiative forcing at surface, C) lower-tropospheric stability calculated as the difference between the potential temperature at 900 hPa and 1000 hPa, D) summer averaged precipitation rates, over North America. Areas marked with black dots indicate grids where changes are significant at the 95% confidence level.

3.4 Future Implications and uncertainties

However, is the simulated future enhancement in wildfire contribution over EUS substantial enough to affect the surface $PM_{2.5}$ values over EUS in future? The World Health Organization (WHO) air quality guidelines for annual and daily $PM_{2.5}$ concentration are 10 μg m^{-3} and 25 μg m^{-3}, respectively. As no specific guideline for seasonal-mean $PM_{2.5}$ in the summer is available, we use the annual guideline value as a reference to understand the implication of wildfire emissions on ambient $PM_{2.5}$ concentration in the future. Interestingly, the mean summertime $PM_{2.5}$ concentration in the wildfire emission free (WEF) scenario is projected to remain within 10 µg m^{-3} over most of North America, except for the southeastern US (~15% of the domain) (Figure 7A). However, the ALL-scenario projects an increase in the exposure concentration level such that values > 10 µg/m^3 are common in Canada and EUS in the future (Figure 7B). Quantitatively, over Canada, the entire PDF of $PM_{2.5}$ concentration shifts towards higher values by ~5-6 µg/m^3. Specifically, the modal value shifts from ~ 6 µg/m^3 in 2050_{WEF} to 11-12 µg/m^3 in 2050_{ALL} (Figure 7C), so $PM_{2.5}$ concentration is projected to surpass the WHO guidelines over a large fraction of Canada in the future. Similarly, the entire PDF of $PM_{2.5}$ concentration shifts towards higher values by ~2-3 µg/m^3 over EUS, with the mode of the PDF increasing from ~ 7-8 µg/m^3 in 2050_{WEF} to ~ 10-11 µg/m^3 in 2050_{ALL} (Figure 7E). The modal value of summer mean $PM_{2.5}$ over WUS increases from ~ 6 µg/m^3 in 2050_{WEF} to ~ 7-8 µg/m^3 in 2050_{ALL} (Figure 7D), although a few grid cells show $PM_{2.5}$ values greater than 10 µg/m^3 (Figure 7B).

Clearly, the climate-induced enhancement in fires and its influence via the advected wildfire smoke to EUS can have significant implications for air quality management in the future. The PM2.5 enhancement in future over the southern states within EUS is large (Figure 7A-B), which is consistent with Figure 3 and 4 results. However, the future change in burnt area over the same region is negligible or mostly reducing (Figure 1C-D). Thus, it can be argued that the simulated enhancement is mostly related with the dynamic perturbations and thermodynamical feedbacks due to wildfire emissions (Figure 6). As the rate of anthropogenic emissions is also regionally highest over the Southeastern states, the impact of these wildfire-induced climatic feedbacks on local air quality is distinctly seen over the EUS.

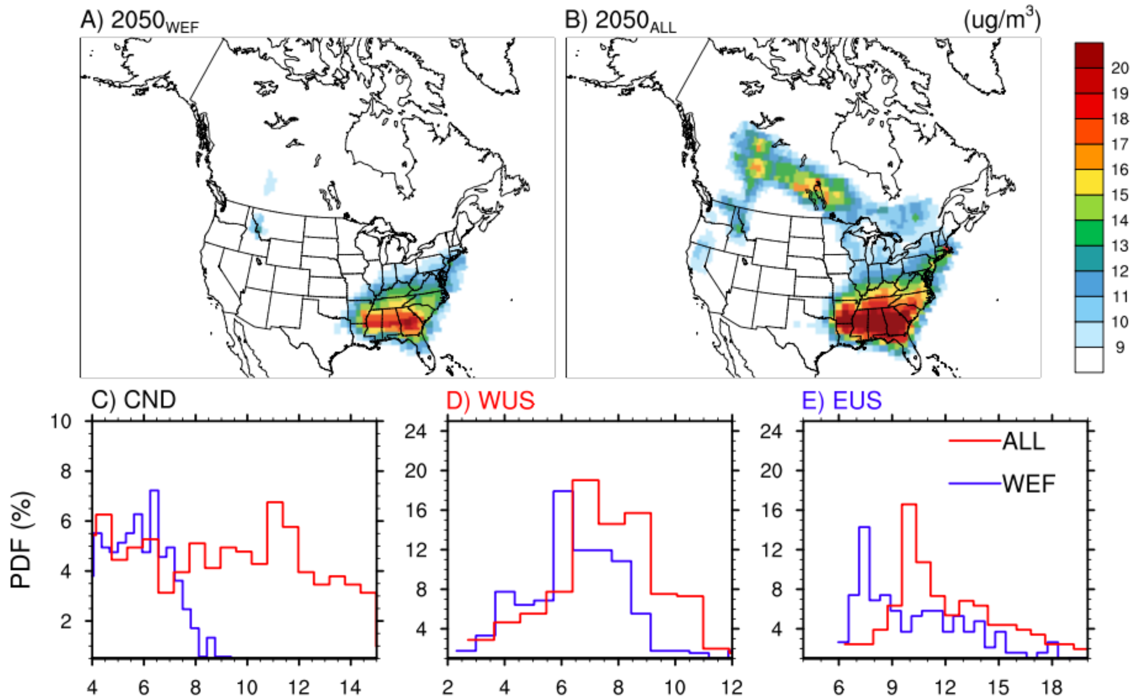

Figure 7: **Spatial distribution and probability density function of** $PM_{2.5}$ **concentration in 2050s. A-B**, Spatial distribution of decadal-average summer (June through August; JJA) mean $PM_{2.5}$ concentration over North America in mid-21st century from 2050_{WEF} (wildfire emission-free) (A) and 2050_{ALL} (wildfire emission-inclusive) (B). **C-E**, Probability density functions (PDFs) of the same within the three regions shown in Figure 2B for Canada; CND (C), western US; WUS (D), and eastern US; EUS (E), respectively, for the 2050_{WEF} (blue) and 2050_{ALL} (red) runs. The y-axis indicates the probability of occurrence of different $PM_{2.5}$ values shown in the x-axis. Only grids over land in North America are used to generate the PDFs. Note the different ranges of values shown in the y- and x-axis in **C-E**. The colorbar and the x-axis for Panel C-E indicates $PM_{2.5}$ values.

Note that our simulated present-day estimates of wildfire induced $PM_{2.5}$ values as well as the percentage contribution of wildfire emissions are within the range of reported values in previous studies over the domain, which augment the fidelity our future projections. Specifically, our simulated present-day estimates of wildfire induced $PM_{2.5}$ values are also within the range of reported values in previous studies over the domain. Reported values of wildfire-induced $PM_{2.5}$ over WUS during summertime vary from ~1 $\mu g/m^3$ (Jaffe et al., 2008) to ~2 $\mu g/m^3$ (Park et al., 2007) and ~3 $\mu g/m^3$ (Ford et al., 2018), with the highest values documented over the Pacific Northwest and west coast regions (~1-4 $\mu g/m^3$) (O'Dell et al., 2019). The wildfire-induced $PM_{2.5}$ over EUS during summertime varies from ~1 $\mu g/m^3$ (Park et al., 2007) to ~2.5 μg/m^3 (~ 3 μg/m^3 in the southeastern US) (Ford et al., 2018). Consistently, our simulated present-day estimates of wildfire contribution values are also within the range of reported values in previous studies. For example, Meng et al. (2019) found that wildfires can be the largest sectoral contributor (~18-59%) to the population-weighted $PM_{2.5}$ in various subregions of Canada. Over WUS, the present-day percentage contribution of wildfire induced $PM_{2.5}$ to the total $PM_{2.5}$ is reported to be ~ 12% (Liu et al., 2017), ~ 15% (Park et al., 2007) and ~30% (Ford et al., 2018), with higher values of ~40% in the Pacific Northwest (O'Dell et al., 2019). Over EUS our simulated values are also within the range of previously reported values of ~ 5% (Park et al., 2007) and ~15-18% (Ford et al., 2018). However, our two-way coupled simulations illustrate that future enhancement in the wildfire associated $PM_{2.5}$ over the EUS could be greater compared to the western US, which is not emphasized explicitly in any of the previous studies (although Ford et al., 2018 illustrated increase in PM2.5 over mid and central US from Canadian fires). These could be since inclusion of the wildfire-induced climatic feedbacks in our simulation is an unprecedented exercise. Please also note that our study is focused on JJA period and the wildfires in western US mainly occurs during August-September months, so the results should be compared consciously.

Nonetheless, inherent limitations in our simulations may introduce uncertainties in the projected future changes. For example, our reported changes in $PM_{2.5}$ concentrations based on relatively coarse resolution simulations and decadal averages likely represent a low-end estimate compared to changes at regional and daily/weekly scales. Moreover, our experiments do not consider the direct human influences such as population change and socioeconomic development on wildfires, which may aggravate the increase in $PM_{2.5}$ concentrations over the densely populated EUS in the future. Common sources of uncertainty in modeling burnt area and fire emission and fire aerosol and smoke are also present in our model. Fire smoke, in particular, is extremely hard to measure and evaluate. Lastly, inherent uncertainties in the physics parameterizations used in the model, sensitivity of climate to GHGs emissions, and the RCP scenarios should also be noted. Thus, ensemble modeling considering different emissions scenarios, population and future time periods, and the use of a finer spatial resolution may provide a more robust and better quantification of the wildfire-induced impact on policy regulated improvements in $PM_{2.5}$ over EUS.

4. Conclusion

In summary, online coupled fire-climate-ecosystem simulations project a nearly twofold increase in wildfire-induced summer-mean surface $PM_{2.5}$ concentration by the mid-21st century over the entire North America. In a wildfire-emission free future, a large portion of North America will have $PM_{2.5}$ values below the WHO guidelines. But in a future with wildfire emissions, the improvements from policy-driven reductions in anthropogenic $PM_{2.5}$ will be compromised by the projected doubling of $PM_{2.5}$ from wildfires. More strikingly, wildfire-induced enhancement in surface $PM_{2.5}$ values and percentage contribution of the wildfire emissions over EUS could be substantial by mid-century. This is mainly because of the large enhancement in fires over Northern America by 2050s and associated increase in amount of downwind transport of smoke to EUS. In addition, enhancement of smoke transport induces a positive climate feedback to $PM_{2.5}$ concentrations over EUS by increasing the lower-tropospheric stability and reducing wet scavenging rates. Despite the inherent limitations, this study highlights the natural versus anthropogenic contributions and the non-local nature of air pollution that can complicate regulatory strategies aimed at improving air quality over the eastern US in a warmer future.

Data availability statements

The HMS data used in this paper are available free through the link https://www.ospo.noaa.gov/Products/land/hms.html. The model simulations data are available at https://portal.nersc.gov/project/m1660/yang560/wildfire

Code availability statements

The model code and scripts are available at https://portal.nersc.gov/project/m1660/yang560/wildfire

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Authors Contribution

YQ, CS and RL designed this study. CS did the model and satellite analysis and wrote the first draft of the manuscript. YZou performed the simulations. All authors provided inputs throughout the study and helped in the drafting and submission process.

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