Supplementary Information for
Tripling of Western US Particulate Pollution from Wildfires in a Warming Climate

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Appendix S1. Cross validation of the MLR model

For each grid d with n years of available observations, we train the model using n-1 years of data and predict over the remaining year. The resulting leave-one-out cross validation results for both the mean PM$_{2.5}$ ($r^2$=0.56–0.61, root mean square error, RMSE =4.0–5.6 μg/m$^3$) and the 95th percentile PM$_{2.5}$ ($r^2$=0.51–0.66, RMSE=11.3–14.1 μg/m$^3$) in August–September averaged over western US grids are statistically significant (p<0.05). The results are close to the standard model fit ($r^2$=0.76–0.82, RMSE=2.3–7.7 μg/m$^3$), indicating robustness of the MLR model.

Appendix S2. Meteorological dataset

Monthly mean surface temperature, precipitation and relative humidity are obtained from the European Centre for Medium-Range Weather Forecasts Reanalysis version 5 (ERA5) (1). The relative humidity is calculated from the dew point temperature in ERA5 as

$$RH = 100\% \times \frac{e_s(T_d)}{e_s(T)},$$

where $T_d$ is the dewpoint temperature and $T$ is the temperature, and $e_s$ is the saturation vapor pressure, which can be empirically calculated using Bolton’s method (2) in the form of

$$e_s(T) = 6.112 \exp\left(\frac{17.67T}{T+243.5}\right).$$

Air stagnation in the U.S. National Centers for Environmental Information dataset is defined as days when there is no precipitation, sea level geostrophic winds are lower than 8 m/s (or <10 m/s when there is a surface to 850 mb temperature inversion), and 500 mb wind speeds are lower than 13 m/s (3). Air stagnation in the CMIP6 models is defined as days with precipitation smaller than 1 mm, surface wind speed lower than 3.2 m/s and 500 mb wind speeds lower than 13 m/s (4).

Appendix S3. Fire modules in the three CMIP6 models

The fire module in CESM2 includes four components: non-peat fires outside croplands and tropical closed forests, agricultural fires in croplands, deforestation and degradation fires in the tropical closed forests, and peat fires (5-7). The burned area fraction is determined by climate and weather conditions, vegetation composition and structure, and human activities. Among them, human influence is represented by anthropogenic ignitions (increase with population density), fire suppression (increase with population density and gross domestic product per capita human), agricultural waste burning, and deforestation rate. After the calculation of burned area fraction, fire impacts are estimated, including fire emissions due to biomass and peat burning as well as plant-tissue mortality, which lead to adjustment of terrestrial ecosystem structure and functioning. We use the coupled simulations from CESM2-WACCM, a version of CESM2 with the same land model but with comprehensive chemistry extending to 130 km vertically (8, 9).

The fire module in GFDL-ESM4.1 is developed within a global dynamic vegetation and land surface model that comprises two sub-models for simulating agricultural and non-agricultural fires (10-12). The area burned for agricultural fires on cropland and pasture at each grid cell is forced with satellite observations of fire seasonality and frequency, depending on grid-scale crop and pasture area but not environmental changes. Simulation of non-agricultural fires follows the process-based fire model in CESM2, which predicts area burned at each grid cell as a product of the number of fires and burned area per fire, depending on grid-scale fuel availability, fuel moisture, and ignition source. Human fire suppression is represented as a function of population density. An enhanced fire rate of spread is introduced to better represent crown fires with high intensity. Non-agricultural fires are simulated on a daily basis, allowing multiday burning with a maximum duration of 30 days. CO$_2$ emissions from fires are estimated by applying combustion completeness factors for different vegetation types on the aboveground biomass within the burned area.

CNRM-ESM2-1 represents natural wildfires over forests and grasslands in a land model that uses prescribed land use and land cover change files (13, 14). The grid cell fire fraction calculation was
adapted to a daily timestep and depends on availability of fuel and fuel moisture, approximated by soil moisture and temperature. Fire fraction is set to zero when the surface litter carbon content is below 200 gC/m² and when soil temperature is lower than 0°C. It is also set to zero when more than 20% of the grid cell is covered by croplands. Except for this limitation on cropland, human fire suppression is not represented. Fire-induced emissions and fire effects on living and dead biomass are Plant Functional Type-dependent.
Supplementary Figures S1 to S14:

Figure S1. Observed correlations between fires and surface PM$_{2.5}$ air quality. Correlation $r^2$ of mean PM$_{2.5}$ averaged over each 2°x2° grid with regional total CO$_2$ emissions from fires in May–November during 1997–2020, derived from simple linear regression (left panels) and multiple linear regression (middle panels) with consideration of meteorological variables, and the variance explained over the western US (black box on maps) by each predicting variable (right panels). The $r^2$ values are color-coded for sites with significant correlations, with gray indicating sites with insignificant correlations ($p > 0.05$). The width of the box (in degrees), within which regional total fire emissions are best correlated with PM$_{2.5}$ at that site, is given in the right corner in the top panel.
Figure S2. Sample sizes of surface PM$_{2.5}$ observations. Number of samples for calculating monthly means and q95 PM$_{2.5}$ at each $2^\circ \times 2^\circ$ grid in August and September averaged over 1997–2020. Only grids with more than 20 samples are shown.
Figure S3. Evaluating model simulations of fires over western North America. (a) The August–September total burned area over western North America (black box in Fig. S4) from 1980 to 2020 from MODIS satellite observations (black) and from three CMIP6 land-only experiments (solid lines). (b) same as (a) but for fire CO$_2$ emissions from two satellite observation-based inventories (black for GFED4s; gray for QFED2.5) and from three CMIP6 land-only experiments (solid lines). Means and standard deviations (sd) during 2000–2014 are reported.
Figure S4. Spatial distribution of burned area from observations and land-only simulations. Mean (left panels) and standard deviation (right panels) of the August–September total burned area during 2000–2014 from MODIS satellite observations and from three CMIP6 land-only simulations. Black box represents western North America where fire CO₂ emissions are integrated for analysis in Figs. 2-4. Global and Northern Hemisphere total burned area (Mha) are reported at the right corner.
Figure S5. Spatial distribution of fire emissions of CO\(_2\) from observations and land-only simulations. Mean (left panels) and standard deviation (right panels) of the August–September total CO\(_2\) emission from fires during 2000–2014 from two satellite observation-based emission inventories (GFED4s and QFED2.5) and from land-only experiments of three CMIP6 fire models driven by observation-based meteorological forcings. Black box represents western North America where fire CO\(_2\) emissions are integrated for analysis in Figs. 2-4. Global and Northern Hemisphere total fire CO\(_2\) emissions from fires (TgC) are reported at the right corner.
Figure S6. Same as Fig. S4, but with model results from the CMIP6 coupled model simulations.
Figure S7. Same as Fig. S5, but with model results from the CMIP6 coupled model simulations.
**Figure S8.** Uncertainties in MLR PM$_{2.5}$ predictions based on percentage versus absolute change of fires. (Left panels) August–September mean PM$_{2.5}$ in the late 21$^{st}$ century (2080–2100) at western US sites (averaged over a 2$^\circ$×2$^\circ$ grid) predicted by MLR driven by the percentage change of fire CO$_2$ emissions under SSP5-8.5 relative to the respective 1990–2010 average from each CMIP6 fire model. (Right panels) same as the left panels but driven by the absolute changes in each model. Only grids with MLR correlation $r^2$>0.5 are shown.
**Figure S9.** Changes in burned area over northern mid-latitudes by three CMIP6 Earth system models. Changes in the August–September total burned area during the late 21st century (2080–2100) compared to present day (1990–2010) under SSP5-8.5 simulated by three CMIP6 models. Number of ensembles for each model is shown in the parentheses at the bottom-left corner. The black box represents western North America. Stippling indicates grids where the changes are not statistically significant at 95% confidence level from >50% of the available ensembles. For multi model mean, the results are first averaged across the available ensemble members from each model (3 for CESM2, 1 for GFDL-ESM4.1 and 5 for CNRM-ESM2-1), and then averaged across the models. Stippling indicates grids with less than two models show statistically significant ($p < 0.05$) changes or the three models do not agree in sign. For each model, a change is defined significant if >50% of the ensemble changes are statistically significant ($p < 0.05$).
Figure S10. Same as Fig. S9, but for fire CO₂ emissions.
Figure S11. Historical evolution of fires over western North America. Temporal evolution of August–September total fire CO₂ emissions (10-year running average) over western North America during 1900–2014 from CESM2 pre-industrial control simulation (pi-Control, black), coupled simulation with all forcings and historical land use and population density (HIST, blue), and coupled simulation with all forcings but with land use and population density held constant at 1850 level (HIST_NoLU, orange). Linear trends (in absolute and percentage relative to 1960 level) during 1960–2014 as well as the 95% confidence limits are reported.
Figure S12. Projected changes in August–September mean PM$_{2.5}$ from each predicting variable. The MLR predicted 10-yr running average of changes in August–September mean PM$_{2.5}$ averaged over the US Pacific Northwest (black box in Fig. 5) based on the CMIP6 Earth system model projections of each predicting variable: fire CO$_2$ emissions (orange), air stagnation index (blue), surface temperature (green), precipitation (cyan) and relative humidity (red).
Figure S13. Same as Fig. S9, but for air stagnation frequency.
Figure S14. Prescribed organic carbon emissions used for chemistry-climate model PM$_{2.5}$ predictions. Shown are August–September total organic carbon emissions over western North America from biomass burning (color-coded solid lines) and anthropogenic sources (color-coded dashed lines) during 2015–2100 under four SSPs. Prescribed fire emissions from GFED4s for historical simulations (before 2014; black) are also shown.
Supplementary Tables 1 and 2

Table S1 List of CMIP6 fire models, and number of ensembles from each experiment.

| Models                  | Land-only | Coupled Historical | Coupled SSP1-2.6 | Coupled SSP2-4.5 | Coupled SSP3-7.0 | Coupled SSP5-8.5 |
|-------------------------|-----------|--------------------|------------------|------------------|------------------|------------------|
|                         | 1850–2014 | 2015–2100          |                  |                  |                  |                  |
| CESM2                   | 1         | 3                  | 1                | 3                | 1                | 3                |
| (1.25°×0.94°)           |           |                    |                  |                  |                  |                  |
| GFDL-ESM4.1             | 1         | 3                  | 1                | 3                | 1                | 1                |
| (1.25°×1°)              |           |                    |                  |                  |                  |                  |
| CNRM-ESM2-1             | 1         | 5                  | 5                | 5                | 5                | 5                |
| (1.4°×1.4°)             |           |                    |                  |                  |                  |                  |
Table S2  Multi-ensemble means of changes in fire CO$_2$ emissions (in percent relative to 1990–2010) over western North America and the MLR-predicted PM$_{2.5}$ (µg/m$^3$; percentage changes in parenthesis) averaged over US Pacific Northwest sites by late 21st century (2080–2100).

| Models            | SSP1-2.6 | SSP2-4.5 | SSP3-7.0 | SSP5-8.5 |
|-------------------|----------|----------|----------|----------|
|                   | Fire CO$_2$ emissions, August–September |
| CESM2             | +113%    | +145%    | +254%    | +242%    |
| GFDL-ESM4.1       | +70%     | +121%    | +202%    | +262%    |
| CNRM-ESM2-1       | +60%     | +100%    | +110%    | +133%    |
|                   | Mean PM$_{2.5}$ (US Pacific Northwest), August–September |
| MLR (CESM2)       | 15.4 (+62%) | 18.4 (+93%) | 22.7 (+138%) | 23.9 (+150%) |
| MLR (GFDL-ESM4.1) | 12.7 (+34%) | 17.8 (+87%) | 22.4 (+135%) | 24.8 (+160%) |
| MLR (CNRM-ESM2-1) | 13.2 (+39%) | 16.6 (+74%) | 18.9 (+99%) | 18.9 (+99%) |
|                   | q95 PM$_{2.5}$ (US Pacific Northwest), August–September |
| MLR (CESM2)       | 46.4 (+91%) | 55.8 (+129%) | 73.0 (+201%) | 76.9 (+217%) |
| MLR (GFDL-ESM4.1) | 39.1 (+61%) | 57.2 (+136%) | 73.6 (+203%) | 81.6 (+236%) |
| MLR (CNRM-ESM2-1) | 38.1 (+57%) | 50.4 (+108%) | 56.4 (+132%) | 58.0 (+139%) |
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