The influence of wildfire smoke on ambient chemical species concentrations in the contiguous US

Emma Krasovich Southworth[1,5], Minghao Qiu[2,3], Carlos F. Gould[4], Ayako Kawano[1,5], Jeff Wen[5], Sam Heft-Neal[6], Kara Kilpatrick Voss[7,8], Alandra Lopez[5], Scott Fendorf[5], Jennifer Burney[7,8], and Marshall Burke[5,6,9,*]

[1]Emmett Interdisciplinary Program in Environment and Resources, Stanford University, Stanford, California 94305, United States.
[2]School of Marine and Atmospheric Sciences, Stony Brook University, Stony Brook, New York, 11794, United States.
[3]Program in Public Health, Department of Family, Population and Preventive Medicine, Stony Brook University, Stony Brook, New York, 11794, United States.
[4]School of Public Health, UC San Diego, La Jolla, California 92093, United States.
[5]Doerr School of Sustainability, Stanford University, Stanford, California 94305, United States.
[6]Center on Food Security and the Environment, Stanford University, Stanford, California 94305, United States.
[7]School of Global Policy and Strategy, UC San Diego, San Diego, California 92093, United States.
[8]Scripps Institution of Oceanography, UC San Diego, San Diego, California 92093, United States.
[9]National Bureau of Economic Research, Cambridge, Massachusetts 02138, United States.
[*]To whom correspondence should be addressed: mburke@stanford.edu & emmars@stanford.edu

Abstract

Wildfires are a significant contributor to ambient air pollution and pose a growing public health threat in many parts of the world. Increased wildfire activity over the past few decades has exacerbated smoke exposure across the US, yet our understanding of how wildfire influences specific chemicals and their resulting concentration in smoke remains incomplete. We combine 15 years of daily measures of species-specific $PM_{2.5}$ concentrations from 700 air pollution monitors with satellite-derived estimates of ambient wildfire smoke $PM_{2.5}$, and use panel regression to estimate the contribution of wildfire smoke to the concentrations of 27 different chemical species in $PM_{2.5}$. We find that wildfire smoke drives detectable increases in the concentration of 25 of the 27 species, with the largest increases observed for chemicals previously associated with biomass burning: organic carbon, elemental carbon, and potassium. We find that smoke originating from wildfires that burned structures had higher concentrations of copper, lead, zinc and nickel relative to smoke from fires that did not burn structures. Wildfire smoke is responsible for an increasing share of ambient species concentrations for multiple species, especially in the Western US. Using existing estimated relationships between ambient chemical exposure and cancer risk, we find that wildfire enhancement of carcinogenic species concentrations could be enough to cause small increases in cancer risk, but these increases are very small relative to other risk factors. Our results demonstrate that fixed ground monitors in combination with satellite-derived data can be used to understand how wildfire smoke influences chemical concentrations at large scales and measure population-level exposures.

Significance Statement

Wildfire risk is growing in the US and many other parts of the world, with demonstrable impact on surface air quality. We use daily measurements from 700 monitors around the US over 15 years to characterize the chemical species present in wildfire smoke, quantify how changing wildfire smoke concentrations are affecting observed species concentrations, and estimate health risks from exposure. We find that increases in wildfire smoke lead to measurable increases in over two dozen chemical species' concentrations and that this influence has grown over time for many species. We show that the burning of structures significantly elevates specific chemical concentrations in smoke and that wildfire enhancement of carcinogenic species could lead to small increases in population cancer risk.

Introduction

Wildfires are a major source of ambient air pollution and represent a growing environmental health hazard in the United States (US) and globally. Over the last few decades, increased wildfire activity has exacerbated smoke exposure across the US and begun to reverse decades of hard-won progress in improving air quality[1]. Studies suggest that, in recent years, wildfire smoke has contributed 25% of all ambient fine particulate matter ($PM_{2.5}$) in the US[2,3], and substantially more in some areas of the Western US. This contribution is projected to grow in the coming years due to higher wildfire risks under a warming climate[4], the expansion of at-risk areas from a growing wildland-urban interface (WUI)[5,6], and increasing policy stringency around reducing other sources of ambient $PM_{2.5}$ pollution[7].

A wide body of epidemiological research has demonstrated that exposure to ambient $PM_{2.5}$ - from smoke or otherwise - is associated with a number of negative health impacts that vary in severity and timing of onset from exposure[8–12]. Documented impacts of exposure to wildfire smoke in particular include adverse birth outcomes[13,14], respiratory exacerbations[12,15], cognitive impairment[16], emergency department visits and hospital admissions[12,17–19], and cardiovascular mortality and morbidity among others[20,21]. While most previous epidemiological research has focused on exposure to total $PM_{2.5}$ pollution, toxicological evidence indicates that exposures to specific chemical constituents of $PM_{2.5}$ may shape how specific health outcomes manifest and evolve[22]. Limited epidemiological evidence also supports species-specific impacts[23–26] and explicitly argues for the consideration of species composition in studies of the health effects of $PM_{2.5}$[27,28].

While existing evidence documents the effects of wildfire smoke on total $PM_{2.5}$ concentrations, a comprehensive understanding of the types and amount of different chemical species in smoke and the corresponding population exposure remains elusive. Most previous research on the chemical composition of wildfire smoke $PM_{2.5}$ comes from the collection of air samples during controlled burns in laboratory settings or at field locations near specific active wildfires[29–34]. These studies find that wildfire smoke is a complex mixture of gases (e.g., carbon monoxide, carbon dioxide, nitrogen oxides) and particles (e.g., trace metals, elemental carbon, organic compounds), and that the mixture of gases and particles emitted by wildfires depends on the materials burned, size of the fire, combustion efficiency, distance the smoke has traveled, and environmental conditions like wind speed, temperature, and humidity[35]. Some recent observational studies use air quality monitoring station data across broader spatial and temporal scales and find that wildfire smoke increases the fractions of elemental and organic carbon during “smoke waves” in the western US[36] and elevates concentrations of trace metals (copper and lead) on “smoke-impacted” days from specific fires in California[37]. While these studies offer important insights into the composition and potential toxicity of wildfire smoke in specific settings, they are often limited in spatial scale (e.g., one sampling location, one region) and thus may not be representative of the types of exposures that a population experiences. In turn, they can neither be used to characterize how overall temporal and spatial variation in wildfire smoke exposure is altering species concentrations, nor to assess the potential population health burden from these alterations.

Here, we characterize the chemical composition of wildfire smoke $PM_{2.5}$ at a broad temporal and spatial scale by combining 15 years of daily measurements of species-specific $PM_{2.5}$ concentrations from 700 air pollution monitors across the continental US, with satellite-derived concentrations of ambient wildfire smoke $PM_{2.5}$[2] from 2006 to 2020 (**Figure 1**). We estimate the impact of variation in wildfire smoke on the concentrations of 27 different species of $PM_{2.5}$ using a panel regression approach that measures how a one unit change in smoke $PM_{2.5}$ affects the mass concentrations of different chemical species (**Materials & Methods**). Our approach relates daily measurements of species concentrations at each monitor to daily variation in $PM_{2.5}$ from wildfire smoke, using high-dimensional fixed effects (dummy variables) to isolate the impact of wildfire $PM_{2.5}$ from a range of other time-invariant, seasonal, and time-varying factors that could also contribute to variation in species concentrations. We estimate both pooled national models as well as region-specific models to quantify how regional factors might influence smoke-species relationships. Building on earlier work that links observed smoke $PM_{2.5}$ concentrations to specific source fires[38], we study whether characteristics of those source fires – in particular, whether structures were burned in the fire – influence measured species concentrations in the resulting smoke. We then combine our statistical estimates of the smoke-species relationships for each species with estimates of nationwide trends in smoke $PM_{2.5}$ and calculate the overall change of species concentrations due to increasing wildfire $PM_{2.5}$ around the country. Finally, we integrate our predicted changes in species concentrations due to wildfire smoke with established relationships between ambient chemical exposure and cancer risks to estimate the increase in cancer cases resulting from wildfire-induced elevations in ambient species concentrations[39,40].

Results

Chemical composition of wildfire smoke $PM_{2.5}$

We find that wildfire smoke significantly increases the concentration of the majority (25 of 27) of chemical species measured by US EPA's CSN and IMPROVE monitoring networks in our sample (**Supplementary Table 1**). The largest increases in concentration relative to an average non-smoke day concentration in our sample (i.e., background species-specific ambient pollution) are for organic carbon (OC), elemental carbon (EC), potassium (K), and phosphorus (P) (**Figure 2**). An additional 1 μg/m^3 of wildfire smoke PM$_{2.5}$ significantly increases the concentrations of OC by 0.39 μg/m^3 (39% increase relative to 0.99 μg/m^3 background, 95% confidence interval (CI): 0.36 to 0.41 μg/m^3); EC by 0.06 μg/m^3 (25% increase relative to 0.25 μg/m^3 background, 95% CI: 0.05 to 0.06 μg/m^3); K by 0.007 μg/m^3 (14.3% increase relative to 0.05 μg/m^3 background, 95% CI: 0.006 to 0.007 μg/m^3); and P by 0.00005 μg/m^3 (7.3% increase relative to 0.0007 μg/m^3 background, 95% CI: 0.00004 to 0.00006 μg/m^3). Findings are robust to alternative model specifications, covariate inclusion/exclusion, and an alternative measure of monitor-captured total PM$_{2.5}$ concentration (**Supplementary Figure 1**).

While the composition and concentration of chemicals can significantly vary based on what is burning, the temperature of the burn, and surrounding atmospheric conditions[41,42], studies have consistently shown that certain species are disproportionately likely to be present in wildfire smoke. For instance, K is emitted from biomass burning even with varying fuel type and burn conditions[43,44], and elevated OC and EC have been associated with increasing wildfire activity as well as as biomass combustion from prescribed fires, wildfires, or residential wood combustion[10,45,46]. Our large observed enhancements in EC and OC during smoke events are consistent with this earlier work[47,48]. We further confirm this consistency by calculating the potassium-to-elemental carbon and the potassium-to-organic carbon ratios – common measures of biomass burning. We estimate that these are are 0.11 ± 0.14 for K/EC and 0.017 ± 0.027 for K/OC in our data, which is similar to published estimates[49].

For the majority of the other species in our sample, we find that variation in wildfire smoke drives smaller (in terms of mass) but statistically significant increases in concentration relative to the average ambient background species concentrations (**Figure 2**; **Supplementary Table 2**). While small, these mass concentration increases could have meaningful impacts on ecosystems and human health, particularly as some are essential for the functioning of different organisms (e.g., zinc)[50]. Other factors, such as the age of smoke and reactivity of the chemical compound in smoke, are also important considerations that may matter more for downstream impacts beyond changes in mass concentration but are not considered here[51].

We detect only small or negligible effects of wildfire-smoke-driven concentration changes for chromium (Cr) and sodium (Na). The null result is expected for Na because it is highly reactive and unlikely to be captured in monitoring readings, which aligns with previous work that does not consistently find elevated Na on smoke days[37]. Previous research shows that Cr production in soil is highly dependent on fire severity and burn temperature, where low severity fires lead to minimal differences in the amount of reactive and toxic forms of Cr between burned and unburned soils, which could in turn impact how much is mobilized in the air[52]; our finding of a small average effect of smoke on Cr concentrations could mask important heterogeneity by fire type.

Variation in the chemical composition of wildfire smoke $PM_{2.5}$

Our main results – using a nationwide model that accounts for regional differences – show that wildfire smoke increases concentrations of several chemicals of interest. However, this approach averages over potentially important regional differences in smoke composition; we therefore also estimate models that allow the effect of wildfire smoke on species concentrations to vary by region. While this approach does not directly link monitor measurements to specific fires (and therefore information about what burned), it does allow the relative compositional relationships to vary across the country. We find regional heterogeneity in the composition of wildfire smoke that is consistent with existing evidence (**Supplementary Figure 2**)[36]. There are large and significant differences in the concentration of EC and OC in smoke $PM_{2.5}$ between the western regions (Rocky Mountain, Pacific, and Southwest) compared to the eastern regions (Northeast and Southeast), whereas for K, the biggest differences are between the Midwest and Southeast regions (**Supplementary Table 3**). These regional differences could be because each region's smoke is a combination of local burned material and transported smoke from burns elsewhere. Given the prevailing burn and circulation patterns, atmospheric mixing is stronger in the eastern regions, with likely strong influence from Canadian fires across our study area and period.

Beyond regional variation in the chemical composition of smoke $PM_{2.5}$, we explore how overall species concentrations are affected by growing wildfire activity by predicting the concentration of each species that is attributable to wildfire over time **(Figure 3)**. Given the increased wildfire activity during this period, we would expect the concentration of chemicals associated with wildfire activity (e.g., from the burning of biomass) to also increase over time. We predict the concentration of each chemical species attributable to wildfire smoke in total $PM_{2.5}$ (see **Materials and Methods**) and find that this quantity is significantly trending upward for the majority of species even as the background species-specific concentrations are decreasing for some chemicals (**Supplementary Figure 3**). By dividing the predicted daily wildfire attributable concentrations by the observed species-specific concentrations in total $PM_{2.5}$ measured at ground monitors, we calculate the share (in %) of each species concentration attributable to wildfire and determine if this share is significantly increasing over time. We find the largest rate of increase in the concentration attributable to wildfire for species linked to biomass burning, with EC and OC both increasing at 0.5% per year and P increasing at 2.7% per year on average. For OC, the 2020 annual average wildfire attributable concentration is 5.2x that of the 2006 concentration (from 0.10 $\mu g/m^3$ in 2006 to 0.52 $\mu g/m^3$ in 2020). This jump is even larger for EC: from an annual average concentration attributable to wildfire in 2006 of 0.01 μg/m^3 to 0.07 μg/m^3 in 2020 (6.1x higher). For P, the 2020 annual average concentration of wildfire-attributable particles is 5.4x that of the 2006 concentration, increasing from 0.00001 μg/m^3 in 2006 to 0.00007 μg/m^3 in 2020 (**Supplementary Table 4**).

Notably, we find that the wildfire-attributable shares of chemicals known to have no safe level of exposure — As, Ni, and Pb[53] — are also increasing over time. On average, the concentration attributable to wildfire is increasing 0.16% per year for As (2020 concentration is 1.9x that of 2006), 0.15% per year for Ni (2.6x), and 0.14% per year for Pb (3x) (**Figure 3**). In other words, wildfire is responsible for an increasing amount of total exposure to many chemicals in our sample. For some of these chemicals (e.g., As, V, SO_4 among others), exposure to non-wildfire sources of a chemical is decreasing even as wildfire driven exposures are increasing (**Supplementary Figure 3**).

Comparison of species concentrations in smoke from wildfires that burn structures

More man-made structures are burning in wildfires in recent years (**Supplementary Figure 4**), which existing work suggests could play a role in the concentration of chemicals in smoke[37]. We leverage our larger temporal and spatial sample to test this hypothesis at scale (i.e., more chemicals across multiple fires), building on earlier work[38] to link smoke exposure at monitors to source fires and then using administrative data to link these fires to the number of structures burned (**Materials & Methods**).

We find that an additional structure burned per fire per day significantly increases the concentration of Ni, Cu, Zn, and Pb in smoke compared to smoke from fires that did not burn structures in our sample (assuming significance at the p ¡ 0.01 level). (**Figure 4A**, **Supplementary Table 5**). Multiplying our estimates by 1,000 for interpretability, an additional 1,000 structures burned per fire day increases the concentration of Cu by 1.7x10^{-4} μg/m^3 (0.009% increase relative to a 0.002 μg/m^3 average concentration in smoke $PM_{2.5}$ when no structures burned, 95% CI: 6.8x10^{-5} to 2.7x10^{-4}μg/m^3) and Pb by 2.5x10^{-4} μg/m^3 (0.02% increase relative to 0.0014 μg/m^3, 95% CI: 1.8x10^{-5} to 3.2x10^{-4} μg/m^3). We find that the largest increase in concentration due to an additional structure burning is for Zn, with an average increase of 1.2x10^{-3} μg/m^3 (0.02% increase relative to 0.006 μg/m^3 , 95% CI: 6.5x10^{-4} to 1.8x10^{-3} μg/m^3). For some elements that are more common in vegetation than structure building materials (OC, Se, P, and Mg), we find a decreasing association between structures burned and species concentrations, consistent with the fact that fires that burn structures are often not those that burn the most vegetation (**Supplementary Figure 5**). Our results are robust to a randomization inference test in which structures are randomly re-assigned to alternate (incorrect) fires (**Figure 4B**, **Supplementary Ta-**

ble 6), which helps rule out that our results are being driven by region- or time-period-specific characteristics of nearby fires rather than by burned structures themselves.

When applied to specific fires that burned many structures, our estimates suggest that these fires can result in substantial downwind increases in key species. For example, we estimate the excess concentrations in smoke $PM_{2.5}$ from the Camp Fire, which was the most destructive fire in California's history, burned over 18,000 structures, and lasted for about 18 days[54]. We find that smoke from the Camp Fire increased species concentrations in smoke $PM_{2.5}$ relative to if no structures had burned, with increases in Pb concentration by 17.0% per fire day or 0.00025 $\mu g/m^3$ (95% CI: 0.00018 to 0.00032 $\mu g/m^3$) and Ni by 7.2% or 0.00003 $\mu g/m^3$ (95% CI: 0.0000031 to 0.00005 $\mu g/m^3$).

Cancer risk assessment from exposure to wildfire attributable concentrations

To understand potential risk to public health from wildfire-driven changes in exposure to specific chemical species, we combine model-estimated changes in country-wide ambient exposure to three carcinogenic species – As, Ni, and Pb – with existing estimates of changes in cancer risk that result from changes in ambient exposure to these chemicals. We estimate that increasing wildfire activity has generated substantial increases in ambient concentrations of these species between the first five years of our sample (2006-2010) to the last five years (2016-2020), with particularly large regional increases in areas where changes in wildfire smoke exposure have been the largest (**Figure 5A**). We then estimate how many additional cancer cases might occur as a result of this exposure, using existing estimates of changes in lifetime cancer risk due to changes in chronic exposure, and assuming that our estimated species concentrations represent average long-term (lifetime) residential exposures (**Materials and Methods**). Total excess cases are then the product of the estimated change in exposure in a given location, the change in cancer risk for that exposure, and the number of people who were exposed in that location.

We find that increases in wildfire activity have likely increased lifetime cancer risk over our study period, perhaps substantially in percentage terms, but that the overall increase in attributable cases is likely very small (**Figure 5B**, **Supplementary Table 7**). For both Pb and Ni, we estimate that chronic exposure to wildfire-driven species concentrations in the recent 2016-2020 period would lead to zero to four additional cancer cases across the US population. For As, chronic exposure to recent concentrations would lead to roughly 45 additional cases. The largest total number of attributable cases are in populated centers in Midwest and Eastern parts of the US, while the largest increase in attributable cases is in the Western US. These increases are very small relative to other risk factors (e.g. exposure to outdoor air pollution causes over half a million deaths due to lung cancer each year, in comparison[55]), suggesting that wildfire-driven changes in ambient concentration of these particular species is unlikely to be a meaningful source of cancer risk. However, this does not mean that other chemicals not measured here or that other exposure scenarios not considered (e.g., acute exposures) would not lead to meaningful increases in risk, nor that exposure to these chemicals could not result in other non-cancer health burdens (e.g., autoimmune disease).

Discussion

Our study characterizes the chemical composition of wildfire smoke using daily concentration of 27 chemicals measured at EPA ground monitors and statistical techniques that can isolate the contribution of wildfire smoke to species concentrations from a broad range of other seasonal, trending, or time-invariant factors that could also affect concentrations. Our research complements previous research that has used a variety of techniques (e.g., chemical transport models[29,56,57], field collected air quality samples[29–33], and laboratory techniques[29–34]) to understand the impact of wildfire on ambient air quality. While these alternate methods offer important insights and detail into the composition of smoke and the mechanisms that drive compositional changes, they are often limited spatially or temporally and/or are highly computationally or resource intensive. Our work enables characterization of the contribution of wildfire smoke to species concentrations at a broad scale and very low computational and resource cost, complementing these other approaches that can provide mechanistic and fire-specific detail.

We detect the signature of wildfire smoke across a broad array of chemical species concentrations and show that this signature has strengthened over time. Our work provides comprehensive new insight into the composition and toxicity of wildfire smoke, with implications for population exposure and corresponding health impacts. We find that wildfire drives significant increases in 25 of the 27 chemical species in our sample, with the largest increases in the concentration of OC, EC, K, and P, consistent with the abundance of these elements in biomass. Given the upward trend in wildfire activity and that the majority of historical wildfires burned in vegetated areas, our results align with previous research that has identified these chemicals as markers of biomass combustion[10,45–47,49]. We find that wildfire also influences many other chemical species' concentrations in our sample, albeit sometimes to a lesser degree, yet these changes may still be meaningful to ecosystem function and human health outcomes given that some of the chemicals are considered essential nutrients (e.g., zinc) or are harmful toxins (e.g., arsenic).

For many of the chemicals in our sample, the *share* of each species' concentration attributable to wildfire has significantly increased over time, with wildfire-attributed concentrations of multiple carcinogenic metals significantly higher by the end of our sample. This trend is consistent with the observed increase in wildfire activity over our sample period and will likely continue to grow given the projections of future wildfire under climate change scenarios. When assessing the risk that these concentration increases pose to human health, we estimate that exposure to wildfire attributable concentrations of three of these metals – As, Ni, and Pb – could result in a combined ~50 excess cancer cases. Though these risks are quite small compared to other environmental risks, there are likely many other chemicals in smoke not measured in our study that are cancer- and health-relevant. Our approach could be readily extended to other settings where consistent monitoring of these other chemicals is available. Our approach could also be extended to study the chemical composition of smoke from prescribed burning, a key fire risk management tool where information on the quantity and toxicity of generated smoke is a key open scientific and public health question.

Our work offers insight into one specific channel through which wildfires can impact the chemical components of ambient air quality: the burning of infrastructure. In line with previous work, we find that wildfires that burn structures result in higher concentrations of certain metals, including Cu, Pb, Zn, and Ni, compared to fires that do not burn structures, and that fires that burn many structures (such as California's 2018 Camp Fire) can lead to large compositional changes in air quality, including substantial enhancement of known carcinogens. Some of these species are considered trace elements in soil (Zn and Cu) and are present naturally but at relatively low concentrations compared to other soil components (like Al, Si, and Fe). Thus, enhancements in the concentrations of Zn and Cu points to smoke from burned structures as an important contributor to ambient concentrations for these species[37,58]. As houses are increasingly constructed in the wildland-urban interface, risk of structure-driven changes in air quality and toxicity will also likely increase, potentially posing significant health risks for nearby populations. However, burned structures are only one among many fire-specific features that could affect the toxicity and health impacts of smoke. Differences in soil type or vegetation as well as the distance the smoke traveled are a few among many factors that could affect how wildfire activity translates into downwind air quality. Future work could again build on existing efforts to link smoke back to its source fire[38], and hone in on the drivers of observed air quality differences and their importance for exposure mitigation and health outcomes.

Our approach has additional limitations. We focus on $PM_{2.5}$ pollution due to its known link to harmful health outcomes, but PM of other size bins and ultrafine particles could also be harmful and are an important avenue to explore in future work. Furthermore, our analysis does not determine the effect that wildfire smoke has on the concentration of sub-species variants (e.g., trivalent and hexavalent chromium), as this information is not captured at monitoring stations.

Better measurement of sub-species concentrations will likely be important for health risk analyses. Similarly, we did not explicitly quantify compound chemical exposures here (simultaneous increases in exposure to multiple chemicals) or try to anticipate their health impacts. Finally, over the course of our sample of air quality measurements, there were changes to air quality monitoring[59], such as changes to detection limits, management, or monitoring methods that were not always clearly documented. We identified a few of these changes (**Figure 1C**), and while our fixed-effects approach will account for many differences in management or measurement error between monitors and any common changes to monitoring across monitors, it is possible we did not account for others.

We contribute to a growing body of literature that demonstrates the impact that wildfires have on a range of chemical concentrations in ambient air. Our findings underscore the growing significance of wildfire as a multidimensional threat to air quality and health. This threat is projected to grow under a warming world. However, wildfires' contribution to air quality is currently not considered in conventional regulatory frameworks at the US federal level. Currently, national air quality standards are focused on six principal pollutants ("criteria" air pollutants), which are regulated by the National Ambient Air Quality Standards in the Clean Air Act, and are intended to provide public health protection[60]. Notably, wildfires and their emissions are excluded from these standards as they are considered "exceptional events," which are defined as natural or unusual events that can overwhelm existing strategies to control anthropogenic pollution. Yet as we show here, wildfires are a significant contributor to the concentration of chemicals in ambient air and may lead to meaningful health impacts, only some of which we can currently document. We take an important step towards demonstrating that not all $PM_{2.5}$ is created equal in terms of composition and toxicity. Given the measurable, detectable, and significant impact that wildfire has on ambient chemical concentrations, our findings could contribute to future policy and regulatory efforts to protect human health from this growing exposure risk.

Materials and Methods

Data

All of the data used in our study are from publicly available sources.

Total $PM_{2.5}$ chemical speciation data. We retrieve chemical speciation data (concentration measurements) from the Federal Land Manager Environmental Database (FED), which is an online repository of air quality data and metadata sponsored by the National Park Service and the U.S. Forest Service.[61]. The FED contains and regularly updates ground-based measurements with standardized metadata from dozens of monitoring networks as well as other air quality outputs. We select annual data from the “Pre-Generated Data” webpage (the same data are also available via query for more customized downloads). Here, we retrieve daily monitor-level chemical concentration measurements from 2006–2020 from two national air monitoring networks in the US: the Environmental Protection Agency’s (EPA) $PM_{2.5}$ Chemical Speciation Network (CSN) (consisting of Speciation Trends Network (STN) sites and supplemental speciation sites) and the Interagency Monitoring of Protected Visual Environments (IMPROVE) program. We select only the chemical species that are measured at both monitoring networks and are in both datasets, which include: aluminum (Al), arsenic (As), bromine (Br), calcium (Ca), organic carbon (OC), elemental carbon (EC), chlorine (Cl), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), magnesium (Mg), manganese (mg), nickel (Ni), nitrate (NO_3), phosphorus (P), potassium (K), rubidium (Rb), selenium (Se), silicon (Si), sodium (Na), strontium (Sr), sulfate (SO_4), sulfur (S), titanium (Ti), vanadium (V), zinc (Zn) (**Supplementary Table 1**).

Measurements from ground monitors, such as those utilized here from CSN and IMPROVE air monitoring networks, are the most accurate available source of information on ambient surface pollutant concentration. However, the monitors also have limitations particularly with respect to measuring wildfire smoke. Monitors are not uniformly spread across the US and monitor density varies dramatically across space. Additionally, monitors have their own set of measurement biases and those biases may vary over space and time. Both CSN and IMPROVE monitoring networks underwent significant changes in the methods and operations of measuring air quality over our sample duration, including: management shifts of who operates specific monitors, technological improvements that enhance the ability of monitors to detect lower concentrations or measure new chemicals, and methodological changes that correct previous biases in measuring concentration of different species[62–64]. In our data cleaning process, we retain any measured negative concentrations values as these are reported in both monitoring networks data. Both the changes in methods as well as the inclusions of negative concentration values are unlikely to impact our main results because our regression approach should capture any baseline changes between monitors in reporting or any changes in the methods or detection limits in a given year. Lastly, wildfire smoke $PM_{2.5}$ is only one of many source that contribute to ambient total $PM_{2.5}$ and these monitors do not distinguish between $PM_{2.5}$ from wildfires and that from other sources of ambient $PM_{2.5}$. Additional steps must therefore be taken to estimate the part of monitor measured $PM_{2.5}$ that comes from wildfire smoke (see **Wildfire Smoke $PM_{2.5}$ Data**).

Total $PM_{2.5}$ data. In addition to chemical concentration measurements, we retrieve total $PM_{2.5}$ concentration data for both monitoring networks from the annual-level pre-generated chemical speciation data files as well as additional data located on the EPA’s AQS site called “EPA $PM_{2.5}$

Mass FRM (88101) - Daily" under the "EPA Air Quality System (AQS)." For our analysis, we select a measure for total $PM_{2.5}$ called "$PM_{2.5}$ - Local Conditions" (MF, AQS code: 88101). There can be many measures of total $PM_{2.5}$ within the same monitoring networks as well as between networks, and previous studies have shown that there can be large differences in the reported concentration of total $PM_{2.5}$ at the same monitor. We explore how using the two different measures of total $PM_{2.5}$ may impact our results, and we show that our results are very similar regardless of the total $PM_{2.5}$ measure (**Supplementary Figure 1**).

In the chemical speciation data, neither monitoring network records the coordinate reference system (CRS) for the coordinate pairs (latitude and longitude) that indicate the location of a given monitoring site. To ensure that the CRS is consistent across all our data, we retrieve spatial information about each site from the Air Quality System (AQS) website[65] and standardize the coordinate reference system across monitoring networks. The location of the monitors used in our analysis along with the duration that each monitoring site is online is shown in **Figure 1A**. The majority of monitors are online for 10-15 years (our full sample period is 15 years).

Our dataset includes chemical speciation data for total $PM_{2.5}$ from both IMPROVE and CSN monitors from 2006-2020 at the daily-monitor level in the contiguous US. To produce a clean dataset, we adjust values that are "Inf", "NaN", "-999", or "-499" to "NA." We leave negative concentration values in our dataset as they could indicate that a measurement was below the method detection limit, except for those mentioned above, which likely indicate a data input error or missing data[66]. We drop any monitor-dates that are missing data for all of our selected species to ensure the composition of our monitoring stations is not changing dramatically over our sample. All chemical concentration measurements as well as total $PM_{2.5}$ are reported in units of $\mu g/m^3$. To explore seasonality (**Figure 1C**) and regional variation (**Supplementary Figure 2**) in the chemical composition of wildfire smoke $PM_{2.5}$, we classify each monitor-date in one of six regions (Midwest, Northeast, Pacific, Rocky Mountain, Southeast, and Southwest) and one of four seasons (autumn [September-November], winter [December-February], spring [March-May], summer [June-August]).

Wildfire smoke $PM_{2.5}$ data. For daily smoke $PM_{2.5}$ concentrations, we use a dataset of gridded daily wildfire smoke $PM_{2.5}$ predictions on smoke days at 10 km resolution from January 1, 2006 to December 31, 2020 for the contiguous US from Childs et al., 2022[2]. Predictions of wildfire smoke $PM_{2.5}$ are non-zero numbers, such that predictions on non-smoke days are by construction zero. When there are predictions of zero for wildfire smoke $PM_{2.5}$ on smoke days, this indicates that a given grid cell-day was determined to be a "smoke-day" but did not have elevated total $PM_{2.5}$. For further details on the daily wildfire smoke $PM_{2.5}$ dataset, see Childs et al., 2022.

For every monitor day, we construct a measure of total $PM_{2.5}$, smoke $PM_{2.5}$, and non-smoke $PM_{2.5}$. We match the gridded daily wildfire smoke $PM_{2.5}$ predictions with the daily chemical species concentration data from the air quality monitors to construct a measure of smoke $PM_{2.5}$ at monitoring stations. We assume that predicted smoke $PM_{2.5}$ in the same grid cell as monitoring station(s) is representative of what measured smoke $PM_{2.5}$ would be at that station(s). For monitor-days that do not have a wildfire smoke $PM_{2.5}$ prediction (i.e., missing), we assume these are non-smoke days, and assign values of zero for smoke $PM_{2.5}$ on these days. For days without wildfire smoke (non-smoke days), we set total $PM_{2.5}$ equal to non-smoke $PM_{2.5}$. For days with a predicted concentration of wildfire smoke $PM_{2.5}$, we calculate non-smoke $PM_{2.5}$ by subtracting smoke $PM_{2.5}$ from the measure of total $PM_{2.5}$ from the air quality monitors. We expect that the concentration of total $PM_{2.5}$ should be at least equal to or greater than the measure of wildfire smoke $PM_{2.5}$, given that total $PM_{2.5}$ should be the total of both smoke and non-smoke $PM_{2.5}$ components. For any total $PM_{2.5}$ concentrations that are either missing or lower than our measure of wildfire smoke $PM_{2.5}$ at a given monitor and day, we adjust the total $PM_{2.5}$ values to be equal to the wildfire smoke $PM_{2.5}$ at a given monitor-date. This is a reasonable assumption given that wildfire smoke $PM_{2.5}$ may comprise the majority of total $PM_{2.5}$ on smoke-days.

Estimating the effect of wildfire smoke $PM_{2.5}$ on chemical species concentration. To assess wildfire smoke's influence on different chemicals in ambient air pollution, we estimate changes in species' concentrations that come from changes in the level of wildfire smoke $PM_{2.5}$, while accounting for any other co-varying emission sources that could also change chemical concentrations in ambient air. A simple comparison of the concentration of chemical species on days with wildfire smoke as compared to days without wildfire smoke would be biased because, in our sample, total $PM_{2.5}$ pollution is about 5 μg/m^3 higher on wildfire smoke days, conditional on controlling for unobserved variation at a given monitor-month and year. Therefore, if wildfire smoke $PM_{2.5}$ and non-smoke $PM_{2.5}$ have the same proportion of a given species, total concentration will be higher on a wildfire smoke day. Here, we define (and calculate) non-smoke $PM_{2.5}$ as the ambient pollution represented by the difference of total $PM_{2.5}$ and wildfire smoke $PM_{2.5}$. Thus, a comparison of the relative species concentrations on wildfire smoke versus non-smoke days will not be informative for per-unit toxicity. Similarly, most of our selected chemical species have background seasonal variation (Fig 1C), since the majority of wildfire smoke days occur between June–September in the US. Accordingly, a comparison of species' concentrations may mischaracterize true seasonal differences as differences in smoke and non-smoke $PM_{2.5}$, though the direction of this seasonal bias is unknown ex ante and could be species-specific.

To do this, we combine observed ambient species concentration at ground monitors with gridded smoke $PM_{2.5}$ predictions at a monitor-day level and estimate a regression framework that compares within-monitor changes over time after flexibly accounting for seasonality and time trends. We do not compare across monitoring stations because monitors in different regions experience different levels of wildfire smoke $PM_{2.5}$ and so observed differences in species concentration could be due to baseline differences in wildfire smoke exposure. We also do not make comparisons for a given monitor across years, because wildfire smoke and some chemical species are trending over time (**Figure 1B and C**). Instead, our regression framework compares monitors to themselves within the same month-of-year after controlling flexibly for time trends. Namely, we relate monitor-specific deviations in average species concentration to monitor-specific deviations in the average level of wildfire smoke, after accounting for baseline differences across monitors and any year-specific shocks experienced by all monitoring stations. Using a fixed effects panel regression, we estimate:

$$spec_{idmt} = \beta_s PM_{idmt}^{smoke} + \beta_{ns} PM_{idmt}^{non-smoke} + \alpha_{im} + \delta_t + \varepsilon_{idmt} \tag{1}$$

via ordinary least squares (OLS) where the outcome is the concentration of a given species at monitor i in month m on day d in year t. Our estimate of interest is β_s, which can be interpreted as the average effect of a one μg/m^3 increase in daily wildfire smoke $PM_{2.5}$ on the same-day concentration of the chemical species of interest in ambient air. We dived our estimates by baseline non-smoke species-specific concentrations to get the change in concentration relative to the average concentration of that species on non-smoke days. We estimate these impacts both in percentage (**Figure 2A**) and in absolute terms (**Figure 2B**).

Our preferred model regresses daily wildfire smoke $PM_{2.5}$ on non-smoke $PM_{2.5}$ with fixed effects for monitor by month-of-year α_{im} (i.e., one intercept for each monitoring station's location in each of the 12 months, e.g., a monitoring station in July in CA), and fixed effects for each year. The monitor-by-month-of-year fixed effects account for any baseline differences between monitoring stations and local seasonality. Year fixed effects account for year-specific shocks common to all monitoring stations (e.g., a particularly bad wildfire year, changes in monitoring methods or equipment, etc). Our approach assumes that day-to-day within monitor variation in wildfire smoke after accounting for seasonality and time trends is plausibly exogenous with respect to chemical species concentrations.

We show that our estimated coefficients are robust to alternative model specifications (**Supplementary Figure 1**). To check whether sources of emissions other than those from wildfire smoke $PM_{2.5}$ change species' concentrations differentially we estimate versions of our models with and without non-smoke $PM_{2.5}$ concentration included as a covariate. We find that our estimates are consistent regardless of whether our models include non-smoke $PM_{2.5}$ and we include it in our preferred model because it is both highly predictive of our outcome and independent of our treatment, and thus reduces our error variance and corresponding standard errors (**Supplementary Figure 1**). We cluster our standard errors at the monitoring station-level to account for potential serial autocorrelation. Main model results can be seen in **Supplementary Table 2**.

Estimating the contribution of wildfire smoke $PM_{2.5}$ to a chemical species' concentrations over time. We predict the contribution of smoke $PM_{2.5}$ to daily chemical species' concentrations over our sample period of 2006–2020. Using the coefficients from our regional model, we multiply the β_s by the predicted smoke $PM_{2.5}$, which provides an estimated concentration of a given species at a monitoring station on a given date. We compare these predictions to the observed species concentration measured at monitoring stations by dividing our predictions by the observed concentration to get the fraction (%) of a given species concentration attributable to wildfire smoke $PM_{2.5}$. We test if the trend in the wildfire attributable fraction of each species is significantly increasing over our sample period by estimating:

$$AttFrac_{imt} = \beta_y year_{imt} + \alpha_{im} + \varepsilon_{idmt} \tag{2}$$

via ordinary least squares (OLS) where the outcome is the wildfire attributable fraction of a given species concentration at monitor i in month m on day d in year t. The estimate of interest is β_y, which can be interpreted as the average annual change (%) in the attributable fraction of the chemical species of interest. We include the monitor-by-month-of-year fixed effects to account for monitor-specific changes and local seasonality. Standard errors are clustered at the monitoring station-level.

We then average the daily concentration predictions by region, species, year, and month to get the average concentration at a given monitor, month, and year, and then we average again by species, year, and month, to get the average concentration of a given chemical species across all monitors in a given year and month. We calculate the average observed concentration in total $PM_{2.5}$ measured at monitoring stations for each species, month, and year. We plot this as percent of concentration due to wildfire smoke $PM_{2.5}$ (**Figure 3**).

Supplementary Figure 3 shows our predicted attributable concentration in comparison to observed species. **Figure 3** condenses this information only showing the attributable fraction of concentration due to wildfire smoke $PM_{2.5}$ for each species, and species that demonstrate significant annual increases in the concentration attributable to wildfire smoke $PM_{2.5}$ have their percent increase year-over-year included in the plot in black. Chemical species in which no detectable significant trend was detected are shown, but the percent increase is greyed out.

Estimating regional chemical species concentration in wildfire smoke PM$_{2.5}$. Using our main model specification (Equation 1), we run a regression for each region (Midwest, Northeast, Pacific, Rocky Mountain, Southeast, and Southwest) to estimate the β_s, or the average effect of a one μg/m^3 increase on the concentration of each chemical species within smoke within that region. Similar to our full sample model, we normalize the regional coefficients by dividing the β_s for each chemical species within a given region by the regional concentration average on non-smoke days as a way to understand how much concentration is changing relative to a baseline ambient species-specific concentration in that region. Our results can be interpreted as the average percent change of a given chemical species' concentration due to a one unit increase in smoke PM$_{2.5}$ within a region relative to a non-smoke day (**Supplementary Figure 2, Supplementary Table 3**).

Estimating the effect of fires that burn structures on species' concentrations in smoke PM$_{2.5}$. We first match wildfires to the smoke PM$_{2.5}$ that they emit. We recover the incident name and number of structures burned for fires in our sample period by processing and combining two datasets: Globfire and monitoring trends in burn severity (MTBS) burned area perimeter data. We combine Globfire's shapefiles of the final area of fires from 2006-2020 with the MTBS burned area perimeter data for the same overlapping period of 2006-04-19 – 2020-12-31. We join Globfire and MTBS data by area, calculate area of intersection, and only keep if area of intersection is >75% of the burned area. We remove fires that have MTBS ignition date more than one week before Globfire start or end dates.

We then incorporate the National Interagency Fire Center (NIFC) data to address some spatial inconsistencies in the burned structures data. Some fires in the burned structure data have integer lat/lon values. To resolve this, we merge the NIFC data with the burned structures data by filtering to the relevant year and correct coordinates. In cases where the lat/lon from the burned structures dataset is an integer, we use the NIFC lat/lon. In the NIFC data, there were 515 duplicated unique identifiers, where some were actual duplicates and others appeared to have slightly different county, FIPS, etc. This is likely because this dataset aggregates reports of fires to different agencies so there could be duplicates. We solve for the duplicate issues by grouping by unique ID, year, incident number, state, county, FIPS, and averaging the lat/lon coordinates. After this, there were still about 35 fires with duplicates IDs and some structures with integer lat/lon, so we remove these from the dataset.

We combine the burned structures dataset with the Globfire data and only keep incidents in which the start date from the structures burned dataset is within two weeks of the Globfire ignition date. To account for the fact that fires burn multiple days and the data provides only the final total number of structures destroyed, we divide the number of structures destroyed by the duration of the fire, using the end-start date of a given fire from Globfire to get the number of structures a particular fire burned in a given day. We aggregate all data to the 10x10km grid cell to match the resolution of our smoke dataset. However, one grid cell may experience multiple fires on the same date, we leverage a method from Wen et al., 2023, where we use a share value to distribute the number of structures destroyed within a grid cell to a specific fire's smoke. We sum up the total structures destroyed from each contributing fire for the given date and grid cell.

To understand if chemicals species' concentrations are changing due to fires that that burn structures, we construct a dataset that matches the concentration of each chemical species (from monitoring stations in that grid cell) in wildfire smoke $PM_{2.5}$ to the number of burned structures per day per fire in that smoke. We filter to fires that have burned at least one structure. We are essentially asking if smoke with structures burned in a fire has different levels of species concentrations than smoke that did not burn structures. We estimate if the effect that an increase in wildfire smoke $PM_{2.5}$ has on species' concentrations differs as a function of the number of structures burned in the source fire. Using a panel regression with fixed effects, we estimate:

$$spec_{idmt} = \beta_s PM^{smoke}_{idmt} + \beta_{bs} PM^{smoke}_{idmt} * BurnStruc_{idmt} + \beta_b BurnStruc_{idmt} + \alpha_{im} + \delta_t + \varepsilon_{idmt} \quad (3)$$

via ordinary least squares (OLS) where the outcome is the concentration of a given species at monitor i on day d in month m in year t in smoke that has burned at least one structure. The estimate of interest is the interaction term β_{bs}, which is a weighted measure of the number of structures burned in the attributed smoke, and can be interpreted as the effect that one additional structure burning in a fire has on the same-day concentration of a given chemical species in wildfire smoke $PM_{2.5}$ (**Figure 4, Supplementary Table 5, Supplementary Figure 4**). We cluster our standard errors at the monitor-level, and we include fixed effects for monitor by month-of-year and year to account for any baseline differences between monitoring stations, seasonality of smoke, and any year-specific shocks common to all monitoring stations. Our approach holds PM constant but does not directly control for fire-specific differences (e.g., low smoke and high structures burned versus high smoke no structures burned), nor does it differentiate timing of structures burned within a fire event.

Permutation/randomization-style inference. We allocate a share of burned structures per day to smoke associated with specific fires, but there may be cases of mis-allocation of our treatment. Mis-allocation could impact our estimated effect of structures burning on species' concentration. Therefore, randomization style inference allows us to test whether the effect we observe is unlikely by chance. Using a sharp null hypothesis that our estimated effect of an additional structure burning is zero, we permute the allocation of structures to specific fire's smoke by explicitly breaking the potential temporal autocorrelation of a given fire burning multiple days (i.e., same fire is overhead multiple days) and spatial autocorrelation (e.g., two or more fires burning at same time and one destroys structures at a monitor in the middle). For example, if we use state-level shuffling of structures burned, the attributed smoke would be the same even if we swap the destroyed structures between the fires. To address this, we explicitly sample from observations within state, but ensure that the samples are from different years, thereby explicitly breaking temporal autocorrelation. We run this simulation 1,000 times for each species and compare our true coefficient to the distribution of the simulated coefficients. We calculate the percentiles of the observed regression coefficient estimates (**Supplementary Table 6**).

Estimating wildfire attributable species concentrations across the US. We estimate species concentration attributable to wildfire smoke in grid cells that have monitoring stations using the location of each monitor (latitude and longitude) via:

$$\begin{aligned} spec_{idmt} = {} & \beta_s PM^{smoke}_{idmt} + \beta_{ns} PM^{non-smoke}_{idmt} + \beta_s PM^{smoke}_{idmt} * \beta_y lat_{idmt} + \\ & \beta_s PM^{smoke}_{idmt} * \beta_x long_{idmt} + \beta_s PM^{smoke}_{idmt} * \beta_x long_{idmt} * \beta_y lat_{idmt} + +\alpha_{im} + \delta_t + \varepsilon_{idmt} \end{aligned}$$

where the outcome is the concentration of a given species at monitor i located at longitude x and latitude y on day d in month m and year t. The coefficients of interest are the β_s and the interacted terms, which allow us to flexibly predict the concentration of a given chemical species in grid cells without monitors. To estimate population-level exposure to chemical concentrations attributable to wildfire smoke $PM_{2.5}$, we predict daily concentrations of each chemical species in 10x10km grid-cells that match those from the smoke $PM_{2.5}$ dataset.

Using our estimated coefficients, we predict across the US:

$$spec_{gmt} = PM^{smoke}_{gmt}(\beta_s + \beta_s * X_g + \beta_s * Y_g + \beta_s * X_g * Y_g) \tag{4}$$

where the outcome is the predicted attributable concentration of a given species in grid cell *g* located at longitude *x* and latitude *y* on day *d* in month *m* and year *y*.

To see how the wildfire-attributable concentration of three health-relevant chemicals, As, Ni, and Pb, have changed spatially over our sample period, we calculate the average wildfire attributable concentration in each grid cell across the first five years (2006-2010) as well as the last five years (2016-2020) of our sample and plot this in **Figure 5A**.

Assessing the human health risk from chronic exposures to historical wildfire attributable concentrations. Following a similar approach to Dickinson et al. (2022)[40] and building on guidance from the US EPA and California's Office of Environmental Health Hazard Assessment[67], we assess the potential cancer risks of chronic smoke exposure by exploring different wildfire exposure scenarios. We consider one standard chronic exposure scenario used to assess cancer risk[68]. The "lifetime" exposure scenario is defined by the projection of chronic and repeated exposure over the average human's life expectancy, which the EPA's designates as 70 years[40,68]. We do not assess acute exposure risk to chemicals in wildfire smoke in this study.

We focus on three chemicals that are measured in our sample and have no known safe level of exposure: As, Ni, and Pb. As and Ni are known carcinogens, while lead has been associated with a number of harmful health outcomes that will likely occur before a person develops cancer; still, Pb is considered a carcinogen[67]. To conduct a risk assessment of exposure to these three chemicals, we first calculate an exposure concentration (EC) using a standard equation developed by the US EPA[68]:

$$EC = \frac{\sum(CA) \times ET \times EF \times ED}{AT} \tag{5}$$

where CA is the chemical concentration in air in μg/m^3 (here the CA is the summed wildfire attributable concentration over the five year period of interest); ET is the exposure time in hours per day (24 hr/day); EF is the exposure frequency in days per year (here the EF is one since each day has a unique value); ED is the exposure duration in years (70 for lifetime); and AT is the averaging time in hours (or 24 hr/day × 365 days/yr × 70 years).

Previous work has set a specific window of exposure to wildfire and assumed that exposure concentrations (ECs) remain similar for the entire window[40]. We modify these assumptions in two key ways: (1) we use our predicted daily wildfire attributable concentrations of As, Ni, and Pb in each 10x10km grid cell over a five year period. As a result, we allow species' concentrations to vary over the exposure window rather than assuming a constant concentration over that window, which is standard practice often due to limited air quality concentration sampling data; (2) we choose a much longer exposure window (5 years versus Dickinson et al. (2022) which uses 30 days) to evaluate the risk from exposure to wildfire attributable concentrations. This longer exposure window has the advantage of being more representative of what a population may have been exposed to over that window and allows us to compare how exposure on average in the first five years (2006-2010) of our sample compared to the last five years (2016-2020) has changed population-level cancer risk.

We use our EC to calculate the cancer risk associated with each chronic exposure scenario for As, Ni, and Pb. We apply the standard equation used by the US EPA and other risk assessment agencies[69–71]:

$$CancerRisk = IUR * EC \tag{6}$$

where IUR (ug/m^3) is the inhalation unit risk, which represents the incremental risk posed by a specific concentration unit in air. We use IUR values from the California's Office of Environmental Health Hazard Assessment (OEHHA), which are widely used in risk assessment practice because they are considered more protective than the current federal standards. For Pb, the IUR is 1.2E-5 m^3/ug, As IUR is 3.3E-3 m^3/ug, and Ni is 2.6E-4 m^3/ug)[67,71].

The calculated outcome of cancer risk represents the number of excess cases above the baseline cancer rate from exposure to wildfire attributable concentrations of individual carcinogenic species. When calculated at the population-scale (rather than the individual), this is known as the excess cancer burden. In practice, if cancer risk is calculated to be 0.0006, this can be interpreted as for every 10,000 people similarly exposed, we would expect 6 excess cases of cancer. Multiplying this estimate of risk by the population will provide the excess cancer burden (or number of cancer cases) from exposure to a specific chemical[67,71,72].

We calculate the population and regional cancer burden by multiplying the cancer risk in each grid cell by the population in that grid cell (gathered from Landscan data[73]. We re-weight the gridded population data using US state-wide census data as Landscan overestimates the population of the US by roughly 40,000,000 people and the US census data is not available at a grid-cell scale. We create weights by dividing the population in each year from the US census data by the Landscan data, and then multiply the weight by the population in each grid cell so that total population from Landscan will match the total population in the US census data[74]. Once we multiply cancer risk by population (we assume the entire population in a grid cell is exposed to wildfire smoke), we get a number that represents the excess cancer burden due to exposure. In other words, this is the expected number of excess cases above background in that grid cell for that exposure level and population. We then we sum across all grid cells in a region as well as across the US to produce a measure of cancer burden from exposure to each wildfire attributable carcinogenic concentration **Figure 5B**.

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Figures

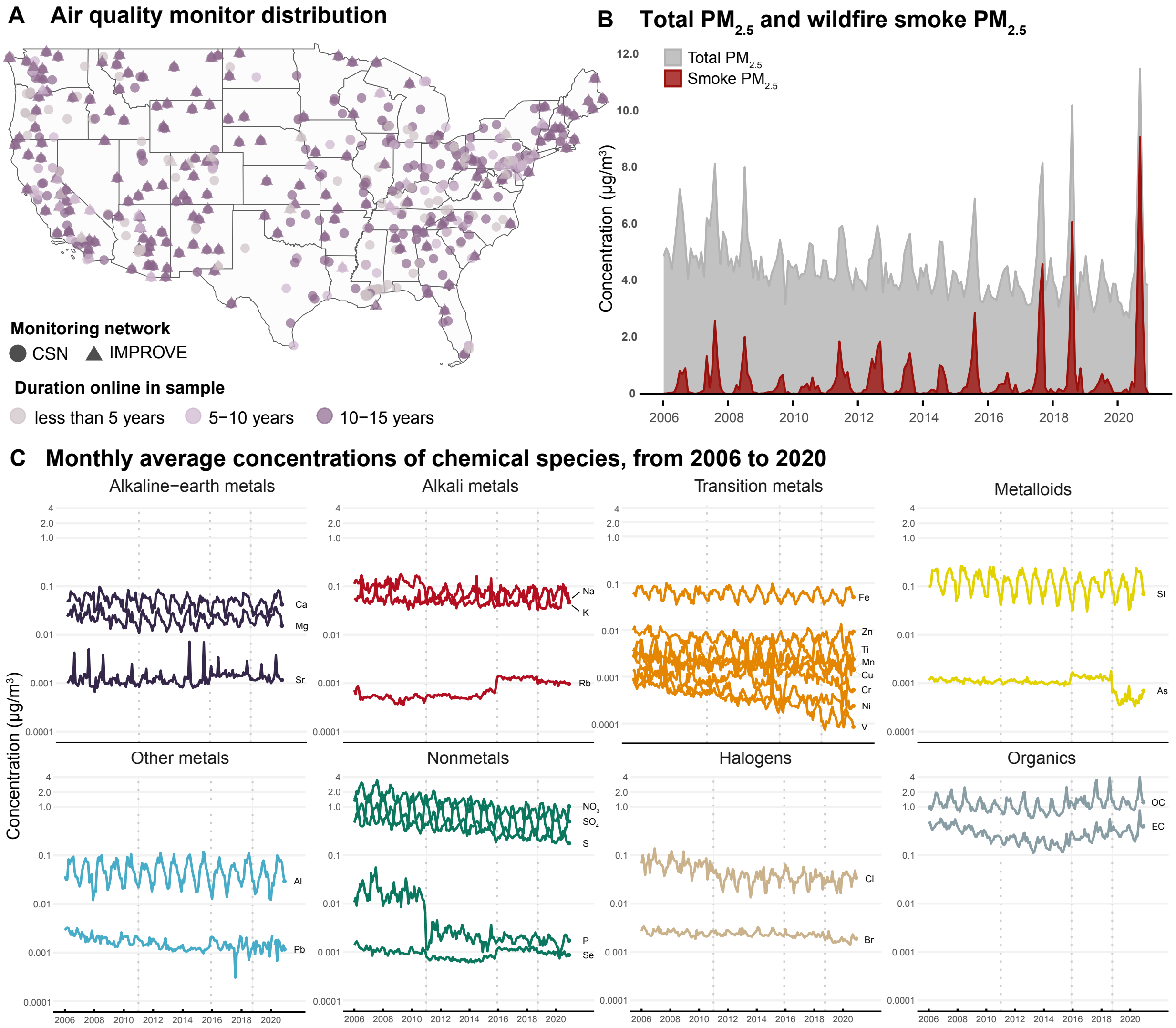


Figure 1: **Temporal trends of total PM$_{2.5}$, wildfire smoke PM$_{2.5}$, and chemical species in the United States.** (**A**) Chemical Speciation Network (CSN) and IMPROVE air quality monitor locations across the United States, colored by duration each monitor is online over our sample period (2006 - 2020). Most monitors sample air quality for the full 15 years. (**B**) Monthly average concentrations (μg/m^3) of total PM$_{2.5}$ and wildfire smoke PM$_{2.5}$ from 2006 to 2020. (**C**) Monthly average concentrations (μg/m^3) of individual chemical species measured by CSN and IMPROVE monitors from 2006 to 2020. Dates at which monitoring networks had documented changes in their monitor methods or other operations are indicated in dashed lines (some changes may not be represented). There is clear seasonality in all of the chemical species.

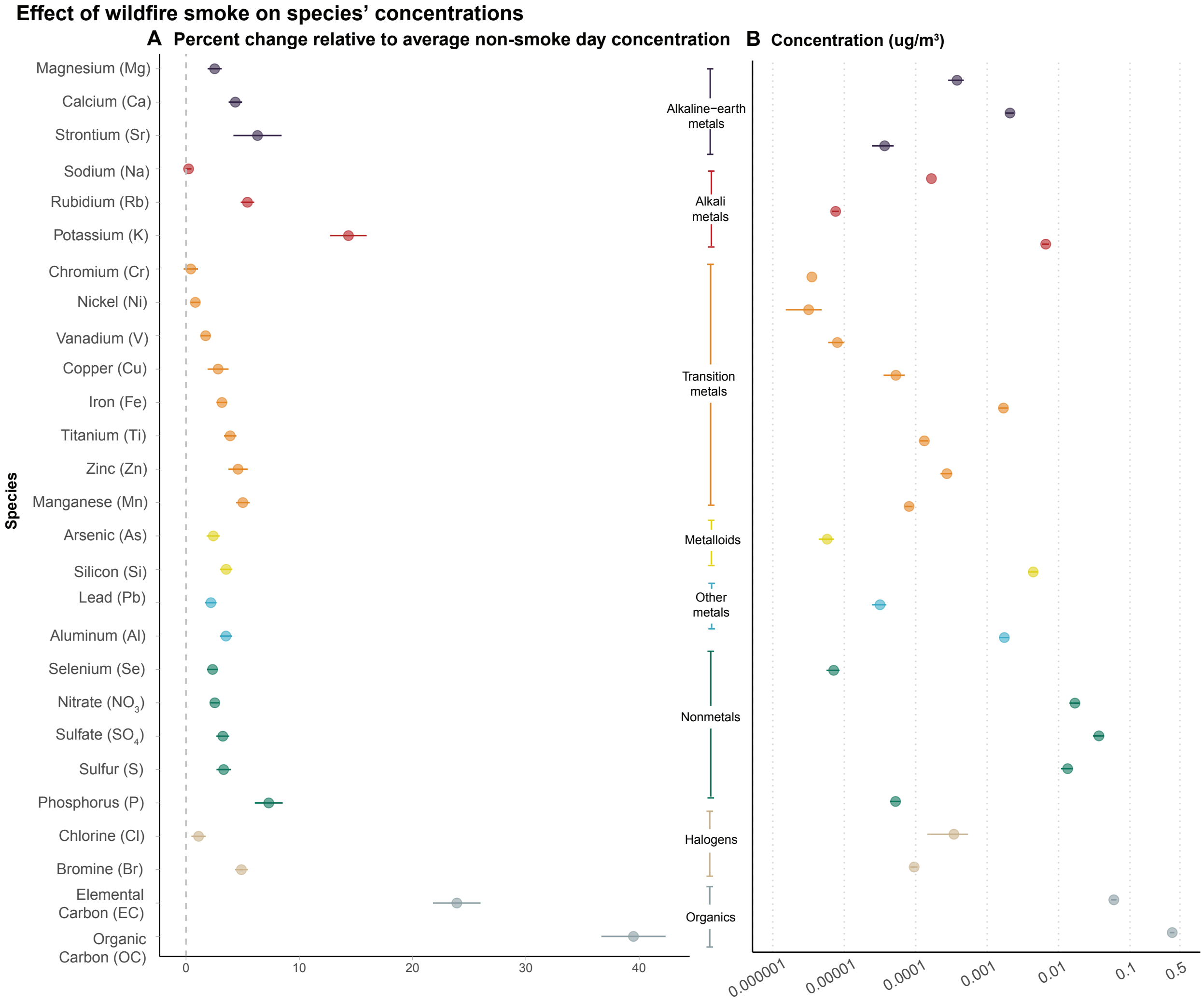


Figure 2: **Effect of wildfire smoke $PM_{2.5}$ on chemical species' concentrations measured at monitoring stations across the contiguous US.** Species are categorized and colored by their chemical properties. (**A**) The percent change in a chemical species concentration relative to its non-smoke day sample average (i.e., relative to a day when there is no smoke in ambient air) given a one $\mu g/m^3$ increase in wildfire smoke $PM_{2.5}$. The largest increases relative to baseline are in species associated with biomass burning, including OC, EC, and K (**B**) The concentration change due to an additional $\mu g/m^3$ of wildfire smoke $PM_{2.5}$ (log scale).

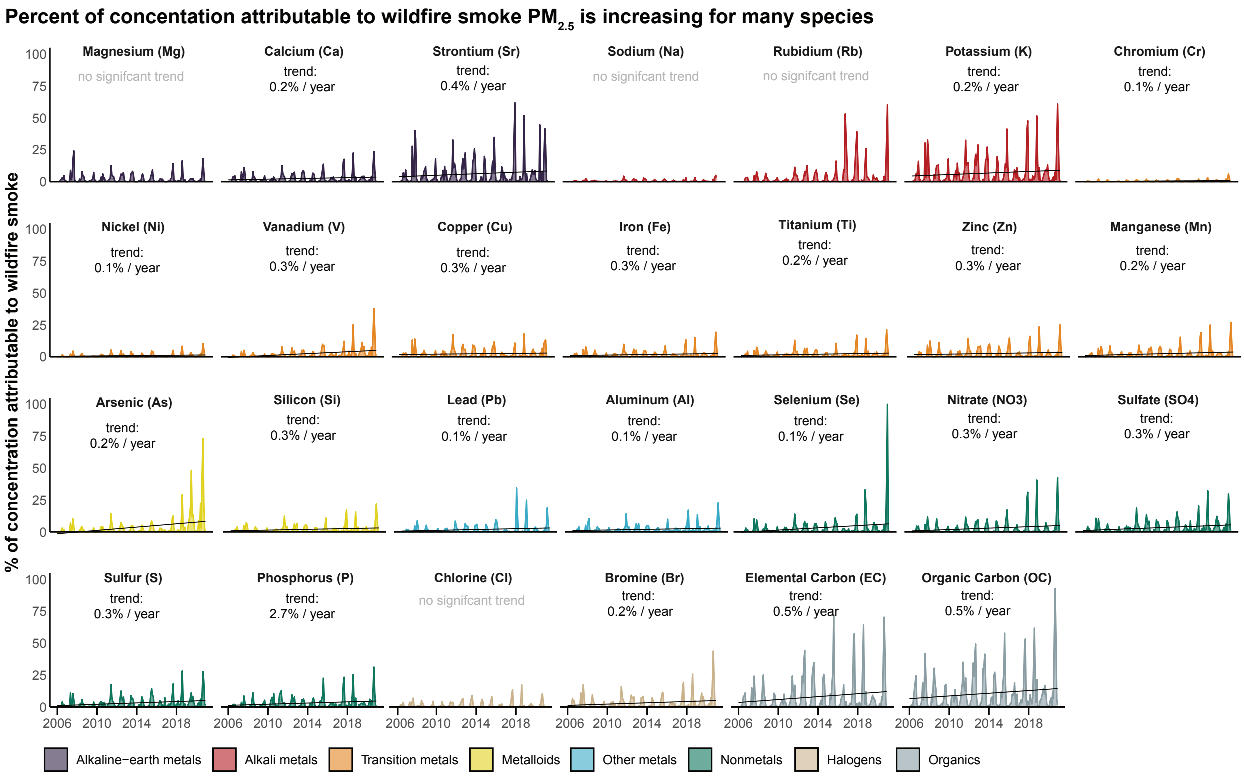


Figure 3: **The percentage of each species' concentration attributable to wildfire smoke PM$_{2.5}$ is increasing for many species**. Predicted concentration of each species due to wildfire smoke over time as proportion of total observed species concentration. Monitor- and species-specific estimates are averaged to the monitor-month and then averaged across monitors to generate the time series shown in each panel. Annual rate of increase for each species due to wildfire smoke is estimated using linear regression, with percentage annual increase annotated in each panel. When wildfire is not driving an increasing fraction of a chemical species, there is no trend line or rate indicated, such as for Mg, Na, Cl.

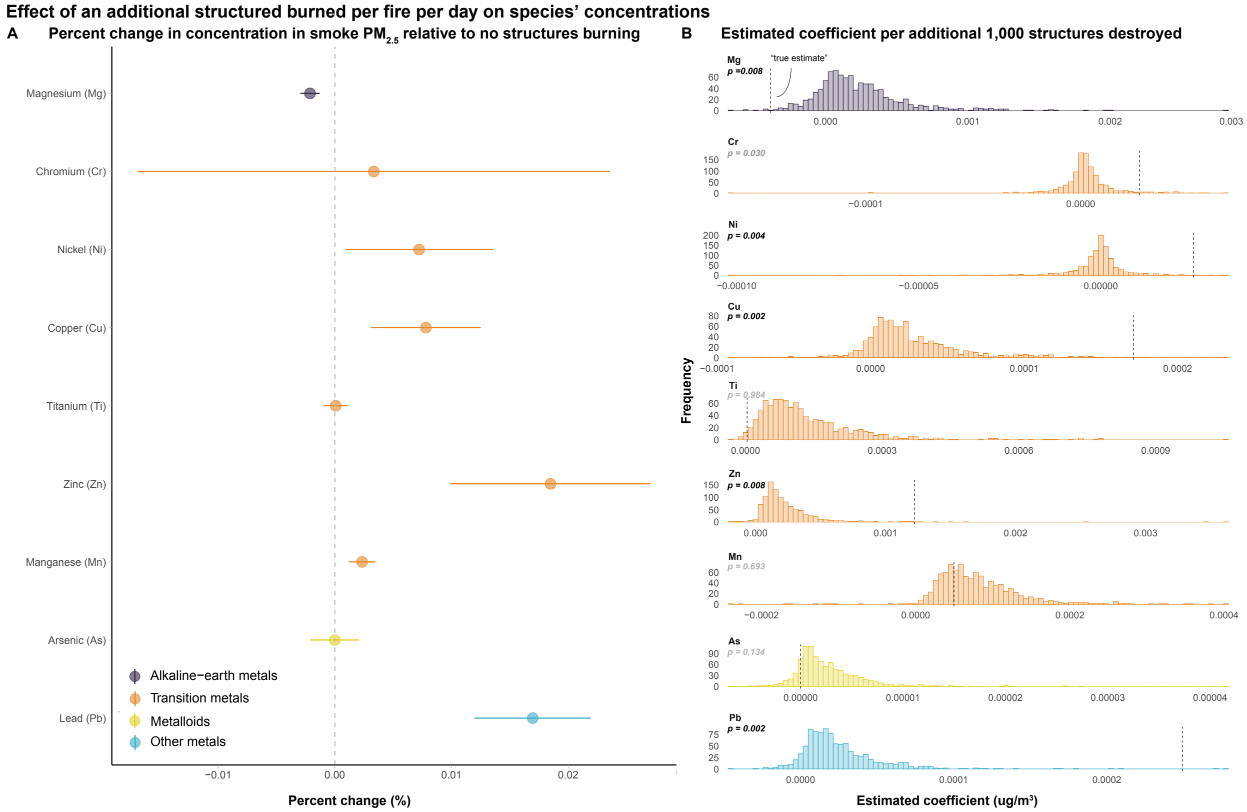


Figure 4: **Smoke $PM_{2.5}$ originating from wildfires that destroyed structures has higher concentrations of Ni, Cu, Zn, Mn, and Pb than smoke from wildfires that did not burn structures.** **A** Estimated effect of one additional structure burned in source fire on species concentration at affected monitor, for specific species previously associated with infrastructure burning. **B** Randomization inference test in which we estimate placebo "treatments" by randomly re-assigning structures burned across fires within each state and then re-estimating the effect of structures burned on species concentration (1,000 replications). Distribution of placebo treatment effects centered at zero, and the estimated treatment effect under true assignment (vertical dashed line) is unlikely to happen by chance (Ni, Cu, Zn, Pb, and Mg are significant at the 1% level).

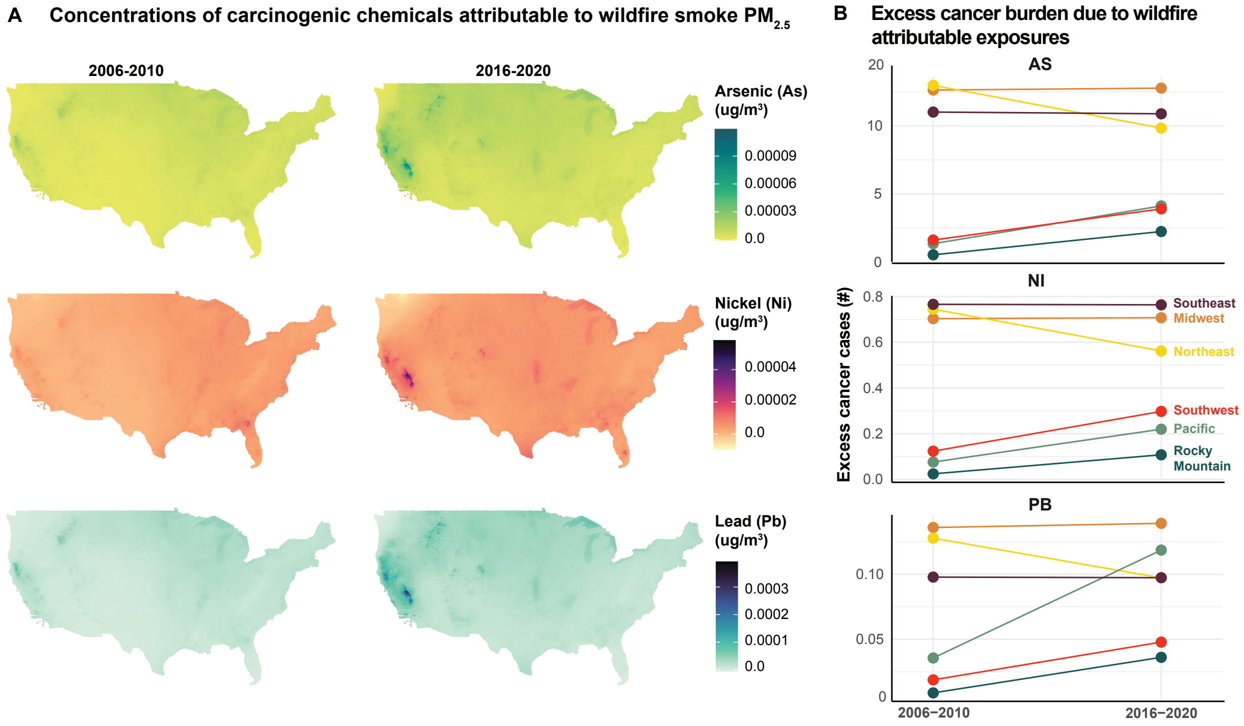
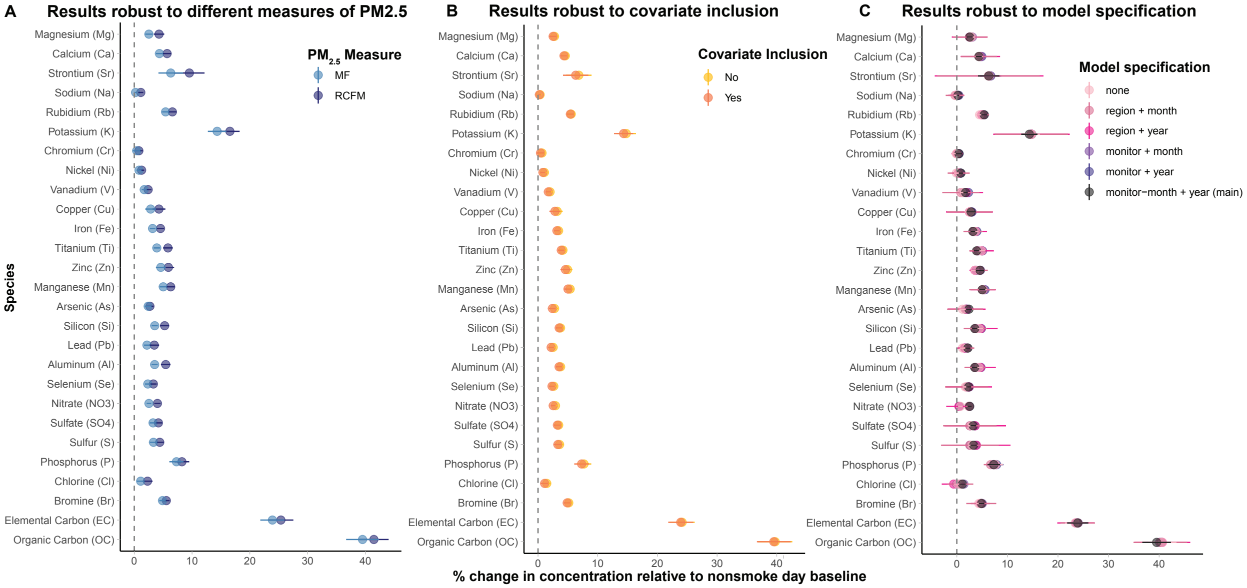


Figure 5: **Health risks from exposure due to wildfire attributable concentrations of carcinogenic species in smoke PM$_{2.5}$.** **A** Average concentrations of carcinogenic species (As, Ni, and Pb) attributable to wildfire smoke PM$_{2.5}$ over two five-year periods (early versus late in our sample) across the US, with noticeable increases in the West and Rocky Mountain Regions. **B** Estimated excess cancer burden due to lifetime exposure to wildfire attributable concentrations observed early versus late in our sample by region. The cancer burden represents the excess number of cancer cases that would be expected above the background rate due to ambient exposure to specific chemicals. Regional differences show the direction and magnitude of difference in estimated cancer risk between exposure scenarios for early and late periods.

Supplementary Information

Supplementary Table 1. List of chemical species in our sample measured at IMPROVE and CSN monitoring sites from 2006 - 2020.

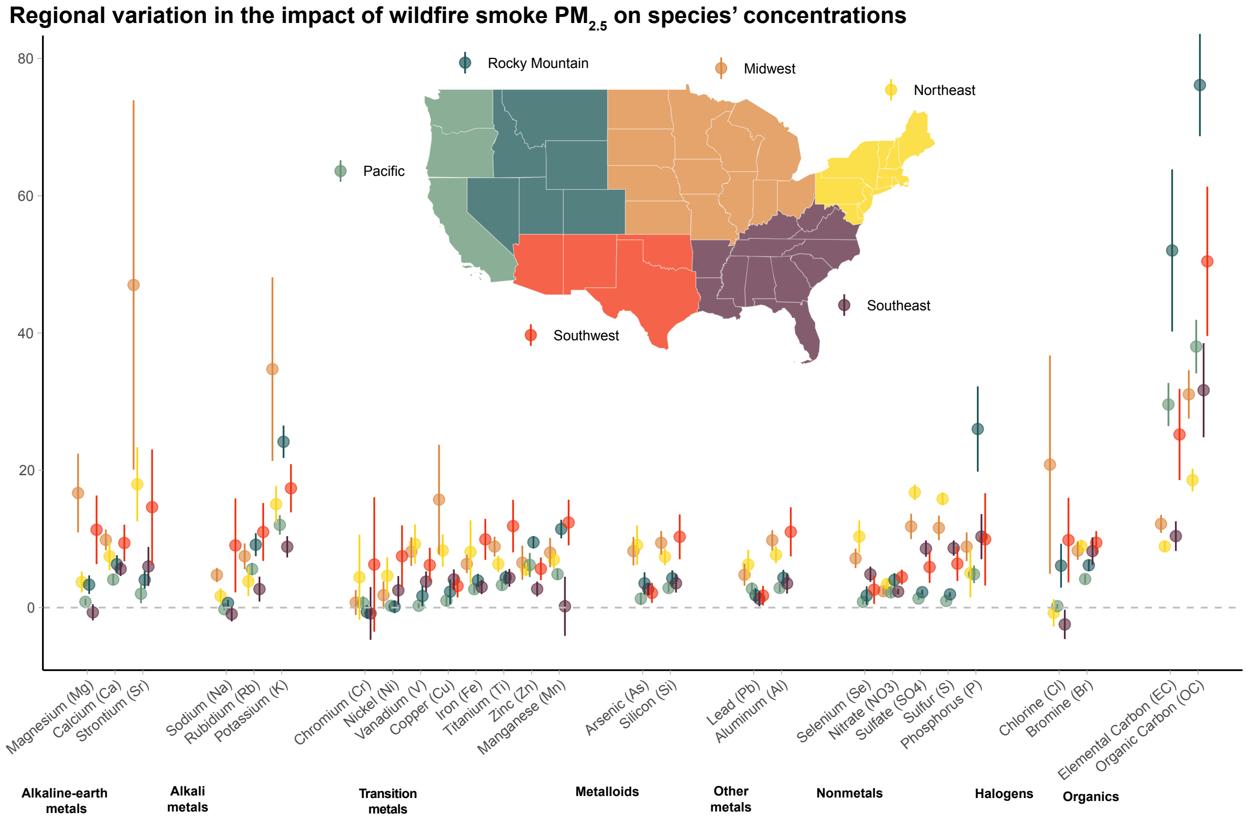
Species type	**Species**
Alkaline-earth metals	Magnesium (Mg)
	Calcium (Ca)
	Strontium (Sr)
Alkali metals	Sodium (Na)
	Rubidium (Rb)
	Potassium (K)
Transition metals	Chromium (Cr)
	Nickel (Ni)
	Vanadium (V)
	Copper (Cu)
	Iron (Fe)
	Zinc (Zn)
	Manganese (Mn)
Metalloids	Arsenic (As)
	Silicon (Si)
Other metals	Lead (Pb)
	Aluminum (Al)
	Titanium (Ti)
Nonmetals	Selenium (Se)
	Nitrate (NO3)
	Sulfate (SO4)
	Sulfur (S)
	Phosphorus (P)
Halogens	Chlorine (Cl)
	Bromine (Br)
Organics	Elemental Carbon (EC)
	Organic Carbon (OC)



Supplementary Figure 1. Results are robust to various sensitivity analyses. A Robustness checks for using different measures of Total $PM_{2.5}$. Using our main model specification, we estimate coefficients for each species using reconstructed fine mass (RCFM) and compare this to our results using measured fine mass (MF) in the main text. We find there our results are quantitatively and qualitatively similar. **B** Robustness checks for including non-smoke $PM_{2.5}$ as a covariate in our model. We find that there are no significant differences in our estimates when directly controlling for non-smoke $PM_{2.5}$. **C** Robustness checks for model specification. We find that there are no significant differences in our estimates when trying alternative model specifications.

Supplementary Table 2. Main model results. Raw estimates correspond to Figure 2B, which is plotted in concentration (μg/m^3) levels. Normalized estimates are our main model coefficients divided by the baseline non-smoke day concentration and multiplied by 100 to provide a measure of the percent change in each species concentration due to smoke $PM_{2.5}$, and correspond to Figure 2A.

Species	Estimate (raw)	Std. Error (clustered)	t-stat	p-value	Baseline non-smoke conc (μg/m^3)	Estimate (normalized)	2.5% CI	97.5% CI
Al	0.0017	0.0001	12.8858	0.0000	0.0491	3.5563	3.0144	4.0982
As	0.0000	0.0000	8.0861	0.0000	0.0002	2.3897	1.8094	2.9699
Br	0.0001	0.0000	17.1362	0.0000	0.0020	4.8568	4.3003	5.4133
Ca	0.0021	0.0001	14.3860	0.0000	0.0481	4.3366	3.7447	4.9285
Cl	0.0003	0.0001	3.4290	0.0006	0.0311	1.1194	0.4784	1.7604
Cr	0.0000	0.0000	1.2843	0.1995	0.0009	0.4079	-0.2157	1.0314
Cu	0.0001	0.0000	5.9792	0.0000	0.0019	2.8076	1.8856	3.7296
EC	0.0595	0.0027	21.6658	0.0000	0.2509	23.6965	21.5488	25.8442
Fe	0.0017	0.0001	13.0489	0.0000	0.0537	3.1617	2.6860	3.6375
K	0.0066	0.0004	17.3209	0.0000	0.0462	14.3469	12.7206	15.9732
Mg	0.0004	0.0000	7.9256	0.0000	0.0149	2.5444	1.9140	3.1748
Mn	0.0001	0.0000	16.2403	0.0000	0.0016	4.9821	4.3798	5.5844
Na	0.0002	0.0001	1.6966	0.0902	0.0726	0.2188	-0.0344	0.4720
Ni	0.0000	0.0000	3.7044	0.0002	0.0004	0.8091	0.3803	1.2380
NO_3	0.0170	0.0014	12.2009	0.0000	0.6726	2.5324	2.1248	2.9400
OC	0.3903	0.0148	26.3438	0.0000	0.9895	39.4472	36.5054	42.3890
P	0.0001	0.0000	11.3754	0.0000	0.0007	7.1348	5.9033	8.3664
Pb	0.0000	0.0000	8.5022	0.0000	0.0015	2.1866	1.6817	2.6916
Rb	0.0000	0.0000	17.6088	0.0000	0.0001	5.3946	4.7931	5.9962
S	0.0132	0.0013	10.2264	0.0000	0.4013	3.2959	2.6631	3.9287
Se	0.0000	0.0000	9.3185	0.0000	0.0003	2.3113	1.8243	2.7983
Si	0.0044	0.0003	12.9684	0.0000	0.1238	3.5671	3.0270	4.1072
SO_4	0.0363	0.0033	11.0922	0.0000	1.1283	3.2184	2.6487	3.7881
Sr	0.0000	0.0000	5.7872	0.0000	0.0006	6.3287	4.1815	8.4759
Ti	0.0001	0.0000	13.9269	0.0000	0.0034	3.9198	3.3672	4.4725
V	0.0000	0.0000	7.5670	0.0000	0.0005	1.7224	1.2755	2.1694
Zn	0.0003	0.0000	10.5446	0.0000	0.0060	4.5619	3.7125	5.4114



Supplementary Figure 2: Regional comparison of selected species concentration in wildfire smoke measured at CSN and IMPROVE sites in the contiguous US. For a one unit increase in smoke PM 2.5, the corresponding percent change ($\mu g/m^3$) relative to the regional sample average for a given chemical species on non-smoke days.

Supplementary Table 3. Regional estimates of the impact of wildfire smoke on species concentrations

Species	Estimate (μg/m^3)	SE	p-value (μg/m^3)	Baseline conc. (%)	Norm est (%)	Norm CI25 (%)	Norm CI975
MIDWEST							
Al	0.0031	0.0002	0.0000	0.0311	9.8791	8.4335	11.3248
As	0.0000	0.0000	0.0000	0.0003	8.2685	6.1965	10.3404
Br	0.0002	0.0000	0.0000	0.0023	8.3283	6.9744	9.6822
Ca	0.0056	0.0004	0.0000	0.0560	9.9399	8.4076	11.4722
Cl	0.0025	0.0010	0.0112	0.0120	20.9204	4.8238	37.0170
Cr	0.0000	0.0000	0.4322	0.0011	0.7305	-1.1032	2.5641
Cu	0.0003	0.0001	0.0001	0.0020	15.8906	7.8587	23.9224
EC	0.0339	0.0019	0.0000	0.2838	11.9385	10.5904	13.2867
Fe	0.0039	0.0004	0.0000	0.0622	6.3348	5.0006	7.6690
K	0.0174	0.0034	0.0000	0.0492	35.3996	21.6950	49.1041
Mg	0.0018	0.0003	0.0000	0.0103	17.1306	11.2179	23.0433
Mn	0.0002	0.0000	0.0000	0.0021	8.0250	5.8572	10.1927
Na	0.0017	0.0002	0.0000	0.0355	4.7668	3.7325	5.8011
Ni	0.0000	0.0000	0.0846	0.0004	1.7780	-0.2457	3.8017
NO_3	0.0319	0.0036	0.0000	1.3267	2.4051	1.8662	2.9440
OC	0.3250	0.0191	0.0000	1.0728	30.2965	26.7577	33.8353
P	0.0000	0.0000	0.0000	0.0005	8.5165	6.4916	10.5415
Pb	0.0001	0.0000	0.0000	0.0022	4.8072	3.1755	6.4390
Rb	0.0000	0.0000	0.0000	0.0002	7.3953	5.4835	9.3071
S	0.0627	0.0048	0.0000	0.5365	11.6928	9.9314	13.4542
Se	0.0000	0.0000	0.0000	0.0005	7.1741	5.7518	8.5964
Si	0.0084	0.0008	0.0000	0.0883	9.5021	7.7384	11.2659
SO_4	0.1800	0.0138	0.0000	1.5176	11.8599	10.0559	13.6638
Sr	0.0002	0.0001	0.0007	0.0005	48.2687	20.6644	75.8730
Ti	0.0002	0.0000	0.0000	0.0022	8.8641	7.4726	10.2556
V	0.0000	0.0000	0.0000	0.0004	8.1921	6.0849	10.2993
Zn	0.0007	0.0001	0.0000	0.0105	6.5610	4.0452	9.0767

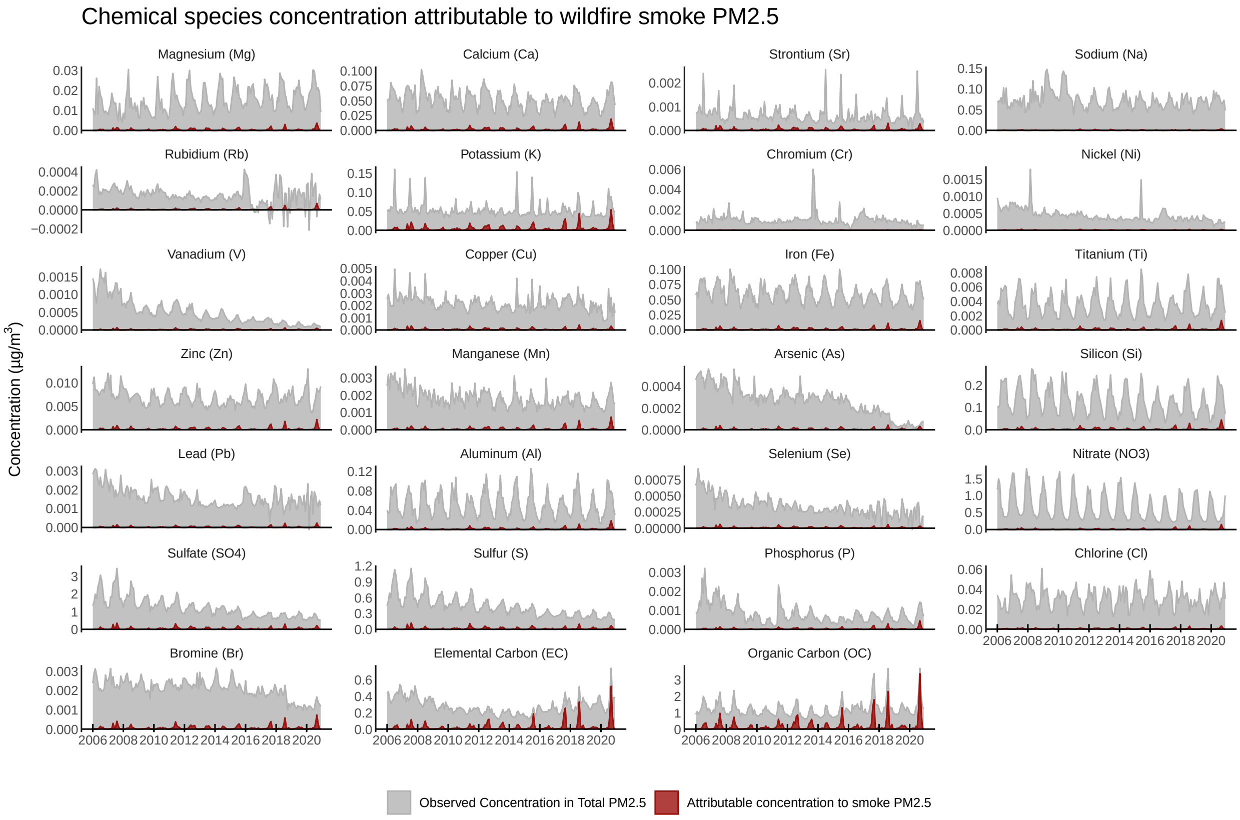
Species	**Estimate**	**SE**	**p-value**	**Baseline conc.**	**Norm est**	**Norm CI25**	**Norm CI975**
	(μg/m^3)		(μg/m^3)	(%)	(%)	(%)	
NORTHEAST							
Al	0.0015	0.0001	0.0000	0.0194	7.8136	6.5548	9.0724
As	0.0000	0.0000	0.0000	0.0004	9.1260	6.2381	12.0138
Br	0.0002	0.0000	0.0000	0.0022	8.9673	8.0504	9.8842
Ca	0.0019	0.0003	0.0000	0.0250	7.4338	5.3532	9.5145
Cl	-0.0003	0.0004	0.4481	0.0421	-0.7605	-2.7420	1.2209
Cr	0.0001	0.0000	0.1610	0.0013	4.3893	-1.7773	10.5558
Cu	0.0002	0.0000	0.0000	0.0023	8.2895	5.9400	10.6390
EC	0.0317	0.0013	0.0000	0.3613	8.7783	8.0685	9.4881
Fe	0.0036	0.0010	0.0008	0.0444	8.0912	3.4549	12.7276
K	0.0067	0.0006	0.0000	0.0443	15.1059	12.4258	17.7861
Mg	0.0004	0.0001	0.0000	0.0115	3.6899	2.1692	5.2106
Mn	0.0001	0.0000	0.0000	0.0012	6.9762	5.6314	8.3211
Na	0.0014	0.0005	0.0050	0.0866	1.6590	0.5139	2.8041
Ni	0.0000	0.0000	0.0010	0.0009	4.6014	1.9074	7.2955
NO_3	0.0258	0.0029	0.0000	0.7612	3.3956	2.6496	4.1415
OC	0.2200	0.0103	0.0000	1.1882	18.5181	16.8002	20.2360
P	0.0000	0.0000	0.0105	0.0009	4.6075	1.1049	8.1102
Pb	0.0001	0.0000	0.0000	0.0020	6.3110	4.1456	8.4765
Rb	0.0000	0.0000	0.0006	0.0001	3.8450	1.6979	5.9921
S	0.0878	0.0026	0.0000	0.5522	15.9051	14.9552	16.8551
Se	0.0000	0.0000	0.0000	0.0005	10.4067	8.0097	12.8038
Si	0.0038	0.0003	0.0000	0.0513	7.4335	6.1965	8.6705
SO_4	0.2630	0.0090	0.0000	1.5694	16.7598	15.6247	17.8948
Sr	0.0001	0.0000	0.0000	0.0004	18.0403	12.5859	23.4946
Ti	0.0001	0.0000	0.0000	0.0019	6.3618	5.1564	7.5672
V	0.0001	0.0000	0.0000	0.0007	9.2215	6.3536	12.0894
Zn	0.0005	0.0000	0.0000	0.0084	5.5566	4.5502	6.5630

Species	Estimate (μg/m^3)	SE	p-value (μg/m^3)	Baseline conc. (%)	Norm est (%)	Norm CI25 (%)	Norm CI975
PACIFIC							
Al	0.0012	0.0001	0.0000	0.0400	2.9092	2.2639	3.5546
As	0.0000	0.0000	0.0056	0.0002	1.2717	0.3798	2.1636
Br	0.0001	0.0000	0.0000	0.0017	4.1507	3.4855	4.8159
Ca	0.0013	0.0001	0.0000	0.0316	4.1016	3.2549	4.9483
Cl	0.0001	0.0001	0.1898	0.0677	0.2075	-0.1040	0.5191
Cr	0.0000	0.0000	0.1712	0.0005	0.6649	-0.2913	1.6211
Cu	0.0000	0.0000	0.0215	0.0020	1.0159	0.1524	1.8794
EC	0.0642	0.0036	0.0000	0.2191	29.2888	26.0448	32.5329
Fe	0.0012	0.0001	0.0000	0.0455	2.6978	2.1005	3.2950
K	0.0052	0.0003	0.0000	0.0431	11.9852	10.5594	13.4110
Mg	0.0002	0.0000	0.0004	0.0191	0.8490	0.3901	1.3079
Mn	0.0001	0.0000	0.0000	0.0014	4.8449	4.0643	5.6256
Na	-0.0003	0.0001	0.0014	0.1189	-0.2342	-0.3755	-0.0929
Ni	0.0000	0.0000	0.2874	0.0004	0.2604	-0.2219	0.7427
NO_3	0.0162	0.0025	0.0000	0.7395	2.1897	1.5307	2.8486
OC	0.3746	0.0202	0.0000	0.9886	37.8937	33.8351	41.9524
P	0.0001	0.0000	0.0000	0.0011	4.7655	3.4913	6.0398
Pb	0.0000	0.0000	0.0000	0.0010	2.7287	1.8070	3.6504
Rb	0.0000	0.0000	0.0000	0.0001	5.5915	4.6852	6.4978
S	0.0022	0.0002	0.0000	0.2209	0.9748	0.7811	1.1686
Se	0.0000	0.0000	0.0000	0.0002	0.8331	0.4436	1.2226
Si	0.0029	0.0003	0.0000	0.0997	2.9025	2.2439	3.5610
SO_4	0.0080	0.0006	0.0000	0.6046	1.3219	1.1415	1.5024
Sr	0.0000	0.0000	0.0048	0.0006	2.0117	0.6236	3.3999
Ti	0.0001	0.0000	0.0000	0.0032	3.3196	2.6248	4.0144
V	0.0000	0.0000	0.1302	0.0005	0.2831	-0.0847	0.6509
Zn	0.0003	0.0000	0.0000	0.0042	6.1736	4.4220	7.9253

Species	Estimate (μg/m^3)	SE	p-value (μg/m^3)	Baseline conc. (%)	Norm est (%)	Norm CI25 (%)	Norm CI975
ROCKY MOUNTAIN							
Al	0.0020	0.0003	0.0000	0.0468	4.3105	3.1458	5.4751
As	0.0000	0.0000	0.0000	0.0001	3.4709	1.8523	5.0896
Br	0.0001	0.0000	0.0000	0.0012	6.1225	5.2596	6.9855
Ca	0.0025	0.0003	0.0000	0.0406	6.2350	4.9377	7.5322
Cl	0.0003	0.0001	0.0003	0.0052	6.0883	2.8772	9.2994
Cr	-0.0000	0.0000	0.2112	0.0004	-0.6322	-1.6294	0.3650
Cu	0.0000	0.0000	0.0137	0.0006	2.2834	0.4790	4.0878
EC	0.0583	0.0067	0.0000	0.1139	51.2289	39.5167	62.9411
Fe	0.0014	0.0002	0.0000	0.0371	3.8952	2.9128	4.8777
K	0.0070	0.0003	0.0000	0.0293	24.0908	21.7285	26.4530
Mg	0.0004	0.0001	0.0000	0.0113	3.3053	1.9524	4.6583
Mn	0.0001	0.0000	0.0000	0.0009	11.3706	10.0200	12.7212
Na	0.0001	0.0000	0.0038	0.0192	0.6804	0.2248	1.1360
Ni	0.0000	0.0000	0.7787	0.0001	0.1327	-0.8026	1.0681
NO_3	0.0119	0.0015	0.0000	0.2966	4.0100	3.0119	5.0082
OC	0.4439	0.0220	0.0000	0.5871	75.6151	68.1609	83.0693
P	0.0000	0.0000	0.0000	0.0002	24.9478	18.9425	30.9530
Pb	0.0000	0.0000	0.0015	0.0005	1.7672	0.6959	2.8385
Rb	0.0000	0.0000	0.0000	0.0001	9.1060	7.4208	10.7912
S	0.0028	0.0002	0.0000	0.1473	1.9319	1.6423	2.2216
Se	0.0000	0.0000	0.0158	0.0001	1.6413	0.3152	2.9673
Si	0.0053	0.0007	0.0000	0.1255	4.2554	3.0882	5.4226
SO_4	0.0091	0.0007	0.0000	0.4020	2.2521	1.9256	2.5787
Sr	0.0000	0.0000	0.0000	0.0004	4.0207	2.7143	5.3271
Ti	0.0001	0.0000	0.0000	0.0028	4.3568	3.3326	5.3811
V	0.0000	0.0000	0.0308	0.0001	1.6720	0.1579	3.1862
Zn	0.0002	0.0000	0.0000	0.0020	9.4327	8.5337	10.3316

Species	Estimate (μg/m^3)	SE	p-value (μg/m^3)	Baseline conc. (%)	Norm est (%)	Norm CI25 (%)	Norm CI975
SOUTHEAST							
Al	0.0019	0.0004	0.0000	0.0498	3.7429	2.2037	5.2822
As	0.0000	0.0000	0.0000	0.0004	2.7697	1.8163	3.7230
Br	0.0002	0.0000	0.0000	0.0025	8.0737	6.0058	10.1417
Ca	0.0021	0.0002	0.0000	0.0366	5.8089	4.7670	6.8508
Cl	-0.0009	0.0004	0.0237	0.0329	-2.6555	-4.9506	-0.3604
Cr	-0.0000	0.0000	0.6323	0.0016	-0.9254	-4.7405	2.8898
Cu	0.0001	0.0000	0.0000	0.0025	4.1221	2.6286	5.6156
EC	0.0386	0.0043	0.0000	0.3789	10.1805	7.9462	12.4148
Fe	0.0017	0.0003	0.0000	0.0552	3.0867	2.0300	4.1433
K	0.0049	0.0004	0.0000	0.0549	9.0026	7.3939	10.6112
Mg	-0.0001	0.0001	0.2925	0.0137	-0.6343	-1.8221	0.5534
Mn	0.0000	0.0001	0.9420	0.0023	0.1595	-4.1638	4.4829
Na	-0.0010	0.0005	0.0544	0.0951	-1.0300	-2.0798	0.0198
Ni	0.0000	0.0000	0.0198	0.0004	2.5830	0.4164	4.7497
NO_3	0.0121	0.0011	0.0000	0.5127	2.3573	1.9394	2.7752
OC	0.4794	0.0552	0.0000	1.5227	31.4836	24.2496	38.7176
P	0.0001	0.0000	0.0000	0.0006	10.0159	6.6120	13.4198
Pb	0.0000	0.0000	0.0202	0.0020	1.2845	0.2032	2.3659
Rb	0.0000	0.0000	0.0053	0.0002	2.6829	0.8122	4.5536
S	0.0611	0.0040	0.0000	0.7012	8.7198	7.5967	9.8430
Se	0.0000	0.0000	0.0000	0.0005	4.8679	3.7037	6.0320
Si	0.0043	0.0008	0.0000	0.1165	3.7139	2.3137	5.1141
SO_4	0.1716	0.0125	0.0000	1.9731	8.6992	7.4459	9.9525
Sr	0.0000	0.0000	0.0001	0.0005	6.1818	3.2081	9.1555
Ti	0.0002	0.0000	0.0000	0.0036	4.5132	3.1702	5.8563
V	0.0000	0.0000	0.0000	0.0006	4.0071	2.4073	5.6069
Zn	0.0002	0.0000	0.0000	0.0077	2.7151	1.6073	3.8228

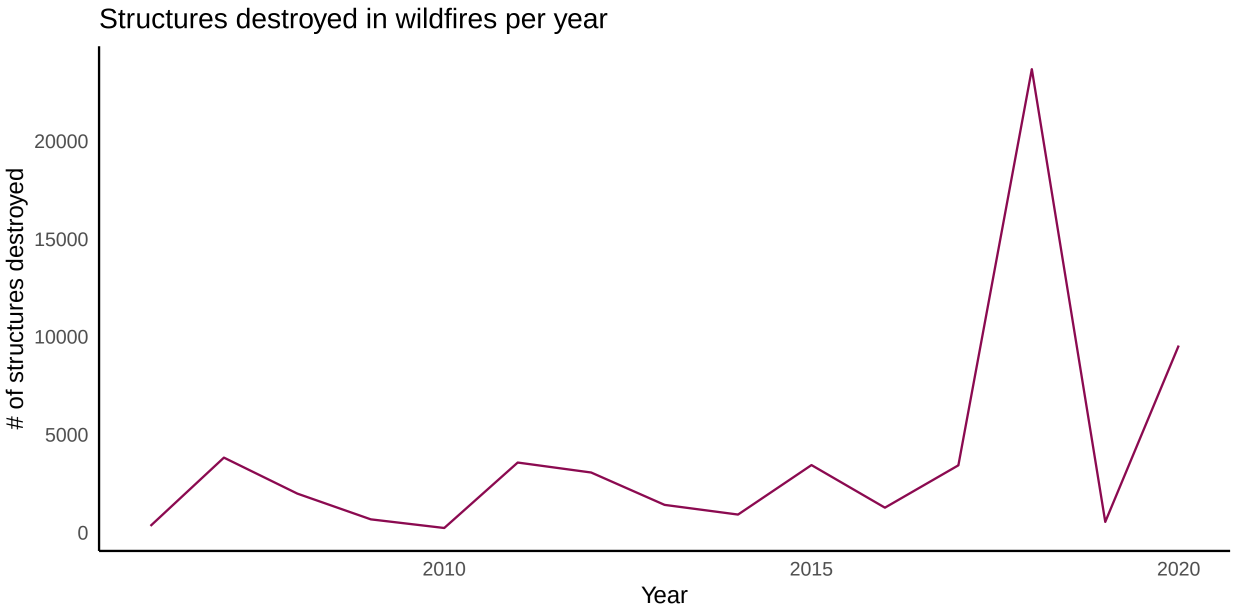
Species	**Estimate**	**SE**	**p-value**	**Baseline conc.**	**Norm est**	**Norm CI25**	**Norm CI975**
	(μg/m^3)		(μg/m^3)	(%)	(%)	(%)	
SOUTHWEST							
Al	0.0123	0.0020	0.0000	0.1109	11.1128	7.5098	14.7158
As	0.0000	0.0000	0.0048	0.0002	2.1270	0.6642	3.5899
Br	0.0002	0.0000	0.0000	0.0020	9.4107	7.7182	11.1032
Ca	0.0096	0.0014	0.0000	0.1024	9.4036	6.7272	12.0801
Cl	0.0019	0.0006	0.0021	0.0196	9.7994	3.6575	15.9413
Cr	0.0000	0.0000	0.2063	0.0003	6.2211	-3.4869	15.9291
Cu	0.0001	0.0000	0.0003	0.0020	3.1405	1.4766	4.8045
EC	0.0516	0.0069	0.0000	0.2055	25.0889	18.4435	31.7344
Fe	0.0080	0.0012	0.0000	0.0804	9.9637	6.9654	12.9620
K	0.0101	0.0010	0.0000	0.0582	17.4137	13.8801	20.9474
Mg	0.0027	0.0006	0.0000	0.0234	11.3764	6.3452	16.4076
Mn	0.0002	0.0000	0.0000	0.0020	12.4369	9.0916	15.7823
Na	0.0071	0.0027	0.0104	0.0782	9.0828	2.1915	15.9740
Ni	0.0000	0.0000	0.0014	0.0002	7.4687	2.9692	11.9683
NO_3	0.0148	0.0018	0.0000	0.3325	4.4508	3.3574	5.5443
OC	0.3974	0.0432	0.0000	0.7896	50.3371	39.4353	61.2390
P	0.0001	0.0000	0.0042	0.0010	9.8600	3.1921	16.5278
Pb	0.0000	0.0000	0.0189	0.0011	1.7285	0.2921	3.1649
Rb	0.0000	0.0000	0.0000	0.0002	11.1188	6.8484	15.3893
S	0.0205	0.0040	0.0000	0.3175	6.4552	3.9224	8.9880
Se	0.0000	0.0000	0.0138	0.0002	2.6388	0.5521	4.7255
Si	0.0279	0.0045	0.0000	0.2689	10.3920	7.1039	13.6801
SO_4	0.0528	0.0104	0.0000	0.8921	5.9154	3.6103	8.2205
Sr	0.0002	0.0000	0.0009	0.0011	14.6368	6.2049	23.0687
Ti	0.0008	0.0001	0.0000	0.0067	11.9170	8.0799	15.7540
V	0.0000	0.0000	0.0000	0.0005	6.2412	3.6890	8.7935
Zn	0.0002	0.0000	0.0000	0.0037	5.6142	3.9725	7.2560



Supplementary Figure 3: The concentration of each species attributable to wildfire smoke PM$_{2.5}$. Observed average species concentration across ground monitors is shown in grey, and attributed portion of the concentration due to wildfire smoke is shown in red. Concentrations are averaged to the monitor, year, and month and then averaged across all monitors for a given year and month to depict the monthly time series over our sample. For some chemical species, such as As, EC and OC, the attributable fraction due to wildfire smoke PM$_{2.5}$ is increasing over time.

Supplementary Table 4. Average annual concentration attributable to wildfire in 2006 and 2020. The multiple is calculated by dividing the average annual wildfire attributable concentration for each species in 2020 by that in 2006.

Species	2006 conc. (μg/m^3)	2020 conc. (μg/m^3)	Increase multiple
Al	0.0006158	0.0033442	5.4
As	0.0000031	0.0000062	2.0
Br	0.0000347	0.0001262	3.6
Ca	0.0008030	0.0034977	4.4
Cl	0.0001659	0.0005888	3.5
Cr	0.0000011	0.0000058	5.2
Cu	0.0000305	0.0000621	2.0
EC	0.0126064	0.0774973	6.1
Fe	0.0006225	0.0028002	4.45
K	0.0022521	0.0089506	4.0
Mg	0.0001763	0.0007078	4.0
Mn	0.0000249	0.0001240	5.0
Na	0.0001574	0.0008456	5.4
Ni	0.0000018	0.0000045	2.6
NO_3	0.0049829	0.0222207	4.5
OC	0.0993863	0.5194907	5.2
P	0.0000134	0.0000724	5.4
Pb	0.0000126	0.0000384	3.1
Rb	0.0000021	0.0000111	5.3
S	0.0085559	0.0152454	1.8
Se	0.0000044	0.0000074	1.7
Si	0.0015560	0.0081340	5.2
SO_4	0.0249822	0.0451040	1.8
Sr	0.0000213	0.0000558	2.6
Ti	0.0000435	0.0002384	5.5
V	0.0000045	0.0000109	2.4
Zn	0.0000901	0.0003476	3.9



Supplementary Figure 4. Number of burned structures per year in our sample.

Supplementary Table 5. Estimates for the effect of an additional burned structure on species concentration in smoke.

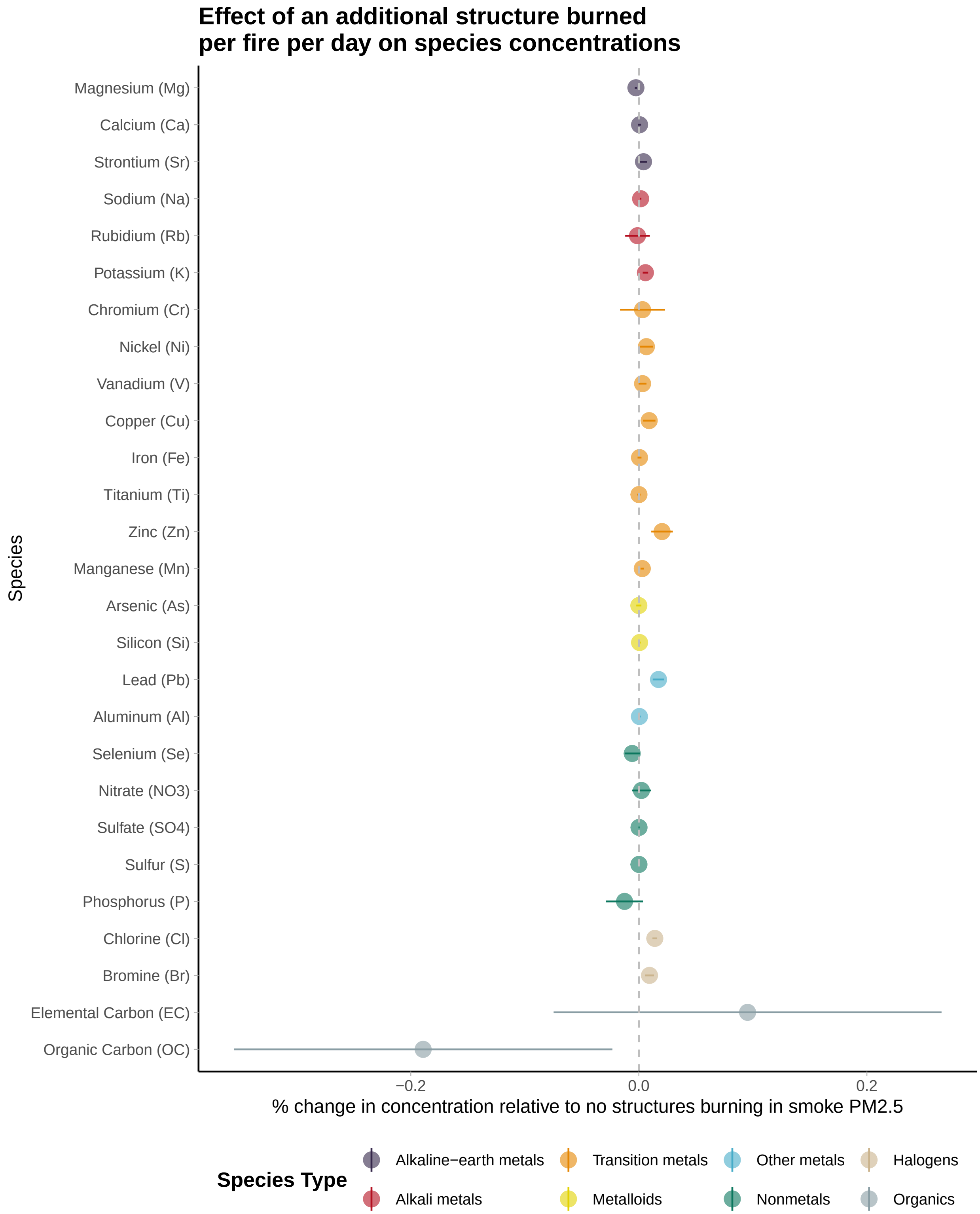
Species	**Baseline conc.** (μg/m^3)	**Estimate** (μg/m^3)	**CI25** (μg/m^3)	**CI975** (μg/m^3)	**Est** (%)	**CI25** (%)	**CI975** (%)
As	2.57E-04	-2.71E-11	-5.42E-09	5.37E-09	-1.05E-05	-2.11E-03	2.09E-03
Cr	8.19E-04	2.73E-08	-1.39E-07	1.93E-07	3.33E-03	-1.69E-02	2.36E-02
Cu	2.19E-03	1.71E-07	6.80E-08	2.74E-07	7.80E-03	3.10E-03	1.25E-02
Mg	1.82E-02	-3.87E-07	-5.38E-07	-2.37E-07	-2.13E-03	-2.96E-03	-1.30E-03
Mn	2.13E-03	4.96E-08	2.54E-08	7.38E-08	2.33E-03	1.19E-03	3.47E-03
Ni	3.52E-04	2.54E-08	3.11E-09	4.78E-08	7.22E-03	8.82E-04	1.36E-02
Pb	1.47E-03	2.49E-07	1.76E-07	3.22E-07	1.70E-02	1.20E-02	2.19E-02
Ti	4.74E-03	3.16E-09	-4.61E-08	5.24E-08	6.66E-05	-9.71E-04	1.10E-03
Zn	6.61E-03	1.22E-06	6.54E-07	1.79E-06	1.85E-02	9.90E-03	2.71E-02

Supplementary Table 6. Randomization inference results. Sharp null hypothesis of no effect of a burned structure on species' concentrations (H_0: Average effect = 0). Comparison of our estimated coefficient (observed estimate) for each species against the distribution of permuted treatment assignment, with the calculated p-value for each comparison.

Species	**Observed Estimate** (μg/m^3)	**p-value**
Pb	2.49E-07	2.00E-03
Cu	1.71E-07	2.00E-03
Ni	2.54E-08	4.00E-03
Zn	1.22E-06	8.00E-03
Mg	-3.87E-07	8.00E-03
Cr	2.73E-08	3.00E-02
As	-2.71E-11	1.34E-01
Mn	4.96E-08	6.93E-01
Ti	3.16E-09	9.84E-01

Supplementary Table 7. Excess cancer burden due to wildfire attributable exposures by region. Expected excess cancer cases due to a lifetime exposure based on the concentrations in two different five-year periods: 2006 - 2010 and 2016 - 2020. Total cases for a given time period and chemical are shown in in the last two columns.

Region	**2006-2010 cases**	**2016-2020 cases**	**Increase multiple**	**Total cases (early period)**	**Total cases (late period)**
As					
Midwest	12.6	12.8	1.0	40.1	43.7
Northeast	13.0	9.8	0.8		
Pacific	1.3	4.1	3.1		
Rocky Mountain	0.5	2.2	4.3		
Southeast	11.0	10.9	1.0		
Southwest	1.6	3.9	2.4		
Ni					
Midwest	0.7	0.7	1.0	2.4	2.7
Northeast	0.7	0.6	0.8		
Pacific	0.1	0.2	2.9		
Rocky Mountain	0.0	0.1	4.4		
Southeast	0.8	0.8	1.0		
Southwest	0.1	0.3	2.4		
Pb					
Midwest	0.1	0.1	1.0	0.4	0.5
Northeast	0.1	0.1	0.8		
Pacific	0.0	0.1	3.3		
Rocky Mountain	0.0	0.0	4.1		
Southeast	0.1	0.1	1.0		
Southwest	0.0	0.0	2.5		



Supplementary Figure 5. Effect of an additional burned structure on all chemical species concentrations in our sample.