



Characterizing emissions, chemistry, and health impacts of aged wildfire smoke in a western US city

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Abstract. We report hourly surface observations of PM_{2.5}, CO, NO_x, O₃, and 75 speciated VOCs in Missoula, Montana, during a strong smoke event in 2020. This study tests our current understanding of wildfire emissions, chemistry, and health effects as implemented in the GEOS-Chem chemical transport model. Three-or-more-day-old smoke transported from California and the Pacific Northwest increased CO, PM_{2.5}, and total measured VOCs by factors of 2–8, with hourly maxima of 800 ppb, 120 µg m⁻³, and 85 ppb, respectively. In contrast, NO_x levels were not elevated compared to the urban background. O₃ showed a non-monotonic response to wildfire smoke: MDA8 O₃ increased under light smoke but flattened or declined when PM_{2.5} exceeded ~30–40 µg m⁻³, a feature that GEOS-Chem failed to reproduce. A 2020-style wildfire season recurring annually would yield an excess lifetime cancer risk of 100-in-1 million or approximately 7 times the non-smoke baseline. The noncancer hazard index (HI) would reach 3.0, meaning substantially elevated acute risks during high-smoke periods. About 90% of cancer risks are from PM_{2.5}, whereas non-cancer risks are dominated by formaldehyde, benzene, acrolein, and acetaldehyde. GEOS-Chem captured major smoke intrusions but underestimated CO, PM_{2.5}, and VOCs by 30–90%. These model biases propagate to health metrics, with GEOS-Chem underestimating smoke-attributable cancer risk by ~40% and chronic HI by ~10 times. We attribute the model errors to underpredicted fire emissions and unrepresented VOC chemistry, which together led to an overestimation of OH and insufficient secondary production.

1 Introduction

Wildfires are the second largest source of volatile organic compounds (VOCs) in the western US (Hoesly et al., 2018; Jin et al., 2023). Some VOCs are designated hazardous air pollutants (HAPs) due to their carcinogenicity and potential for acute or chronic health effects (Naeher et al., 2007; Reid et al., 2016). Lengthening and intensified fire seasons in recent decades have posed direct air quality risks to rural populations and the inhabitants of ~50 million U.S. homes that already lie within the fire-prone wildland–urban interface (WUI), a figure expected to grow by one million every three years (Burke et al., 2021). Moreover, recent years have shown that western US and Canadian biomass burning (BB) smoke frequently impacts cities in



western North America and can reach population centers thousands of kilometers downwind on the East Coast (Yu et al., 2024). Despite the growing threat of more frequent and intense wildfires, the health effects of HAPs emitted from wildfires are not well documented, and residents often lack chemically resolved data on the pollutants to which they are exposed, because of limited ground-level measurements. Here, we combine hourly ground-based observations of VOCs and criteria
 35 pollutants with the GEOS-Chem chemical transport model (CTM) to characterize the composition, chemical evolution, and health implications of aged wildfire smoke.

Past observational efforts have provided valuable yet incomplete insights into the health risks associated with wildfire smoke exposure (Gould et al., 2024). For example, some studies have targeted near-source fireline PM (Navarro et al., 2021), CO
 40 (Semmens et al., 2021), or high-altitude plumes sampled by aircraft (O'Dell et al., 2020), and others have examined speciated VOCs and/or aerosol composition (Akagi et al., 2014; Fiddler et al., 2024; Joo et al., 2024; Liang et al., 2022). Speciated, near-surface data where the majority of human exposure to fresh and aged smoke actually occurs are still seriously under sampled. The lack of sufficient measurements also limits the evaluation of CTMs, which are widely used for smoke forecasting and health-risk assessments. Recent aircraft observations show that GEOS-Chem (a commonly used
 45 CTM) underestimates BB CO and key HAPs such as formaldehyde by a factor of three or more (Jin et al., 2023). However, analogous ground-based validation remains absent. Because near-surface chemistry differs fundamentally from lofted plumes (e.g., ambient temperature, nighttime oxidants, boundary layer mixing) (Decker et al., 2019; Pagonis et al., 2023), ground observations are urgently needed to improve model validation and exposure assessments.

50 The western U.S. fire season of 2020 produced some of the most extreme smoke levels on record in OR, WA, and CA (Albores et al., 2023; Reilly et al., 2022), with September standing out as the peak month (Abatzoglou et al., 2021; Mass et al., 2021). More than 4 million acres burned in California alone, releasing an estimated ~127 Tg CO₂-equivalent, nearly twice the state's cumulative 2003–2019 greenhouse-gas reductions (Jerrett et al., 2022). These BB events provide an ideal natural laboratory for quantifying the composition of aged smoke, and testing CTM capabilities for representing regional
 55 smoke and assessing the continental-scale impacts of wildfires.

In this study, we present hourly ground-based measurements of 75 VOCs (including 15 HAPs), PM_{2.5}, CO, NO_x, and O₃ collected during aged (> 3 days) regional BB events in September 2020 in Missoula, Montana—a representative
 60 northwestern U.S. city frequently impacted by regional wildfire smoke. Leveraging this comprehensive dataset, we (i) characterize the temporal evolution of VOCs and criteria pollutants (Sect. 4), (ii) quantify species-specific BB enhancements (Sect. 5), (iii) investigate O₃–PM_{2.5} relationships (Sect. 6), (iv) assess public health risks using regulatory exposure metrics for PM_{2.5} and HAPs (Sect. 7), and (v) evaluate GEOS-Chem simulations against observations to diagnose model biases in BB emissions and chemistry, and their implications for health-risk estimates (Sect. 8).



2 Methods

2.1 Missoula Science Observatory

Missoula, MT (46.8721°N, 113.9940°W) lies in a mountain valley in the northern Rocky Mountains with a population of approximately 120,000. Missoula has been studied in the past for its frequent impacts from both local (Montana and Idaho) and regional wildfires, including long-range smoke transport from the West Coast and the Pacific Northwest, such as California, Oregon, Washington, and British Columbia (Selimovic et al., 2019, 2020). Its frequent exposure to surface smoke makes it a representative site for evaluating human exposure and air quality under real-world wildfire conditions.

Long-term air quality monitoring at the Missoula Science Observatory (MSO) began in 2017 with measurements including four criteria pollutants ($\text{PM}_{2.5}$, CO, NO_2 , and O_3) and aerosol optical properties (Selimovic et al., 2019, 2020). In 2020, the monitoring scope expanded to include 75 individual VOCs, of which 15 are classified as HAPs by the US EPA. The University of Montana (UM) campus serves as the primary site for CO, NO, NO_2 , O_3 , and VOC measurements. The NO , NO_2 , and O_3 inlet was located 12.5 m above ground at the Charles H. Clapp Building; the CO and VOCs inlet was initially located 9 m above ground on the Chemistry Building, ~70 m away.

Measurements of $\text{PM}_{2.5}$ were obtained from the Montana Department of Environmental Quality site at Boyd Park, roughly 3 km southwest of the UM campus. Despite the spatial separation between measurement sites, previous studies have validated the temporal and spatial agreement in pollutant concentrations between the two sites for 2017–2019 summers (Selimovic et al., 2019, 2020); we confirm similar agreement among $\text{PM}_{2.5}$ measured at the Boyd Park and other tracers measured at the UM campus for this study ($R^2 > 0.9$), supporting the use of combined datasets in this analysis.

Hourly meteorological data were obtained from the MesoWest database and accessed via the Synoptic Data API (<https://synopticdata.com/>). Incoming shortwave radiation (W m^{-2}) was obtained from the Blue Mountain site (station ID: BLMM8; 46.73° N, 114.09° W), while air temperature (°C), relative humidity (percent), and precipitation accumulation (mm) were obtained from the Missoula Valley site (station ID: E0591; 46.86° N, 114.02° W). These stations were selected as the closest available meteorological observations to the MSO (10 km and 5 km away, respectively). We used broadband shortwave (SW) as a first-order proxy for actinic flux and photolysis as they were not measured; We note that the SW (200–4000 nm) can diverge from the near-UV range relevant to $J(\text{NO}_2)$ and $J(\text{O}^1\text{D})$ (~300–420 nm) under smoke. Planetary boundary layer (PBL) mixing height (m) was obtained from the High-Resolution Rapid Refresh (HRRR, 3km), an operational forecast and analysis system developed by NOAA with hourly updates at 3-km horizontal resolution (Dowell et al., 2022). We also obtained the mixing height data from the NASA Goddard Earth Observing System Forward Processing system (GEOS-FP, $0.25^\circ \times 0.3125^\circ$, hourly) as it serves as one of the inputs for GEOS-Chem.



Figure 1 illustrates the climatological distribution of daily $\text{PM}_{2.5}$ during the wildfire season (June–October) for 2010–2024 in Missoula. Despite large interannual variability, a clear seasonal progression is evident. Daily average $\text{PM}_{2.5}$ concentrations remain below $15 \mu\text{g m}^{-3}$ throughout June, increase steadily during July, and peak from mid-August to occasionally mid-September, coincident with increases in wildfire smoke episodes. During the month-long peak periods, the multi-year daily mean $\text{PM}_{2.5}$ concentrations exceed the WHO 24-h guideline ($15 \mu\text{g m}^{-3}$) on 18 days and the US EPA standard ($35 \mu\text{g m}^{-3}$) on 6 days based on the climatology. Peak hourly $\text{PM}_{2.5}$ even reached $\sim 470 \mu\text{g m}^{-3}$ in 2017. Reflecting this chronic smoke burden, Missoula ranks 14th among 223 U.S. metropolitan areas for 24-hour particle pollution and 29th for annual particle pollution (American Lung Association, 2024). In this study, we focus specifically on September 2020, when regional smoke transport led to one of the most prolonged and chemically distinct episodes of aged wildfire smoke observed at the surface (blue line in Fig. 1; Sect. 4).

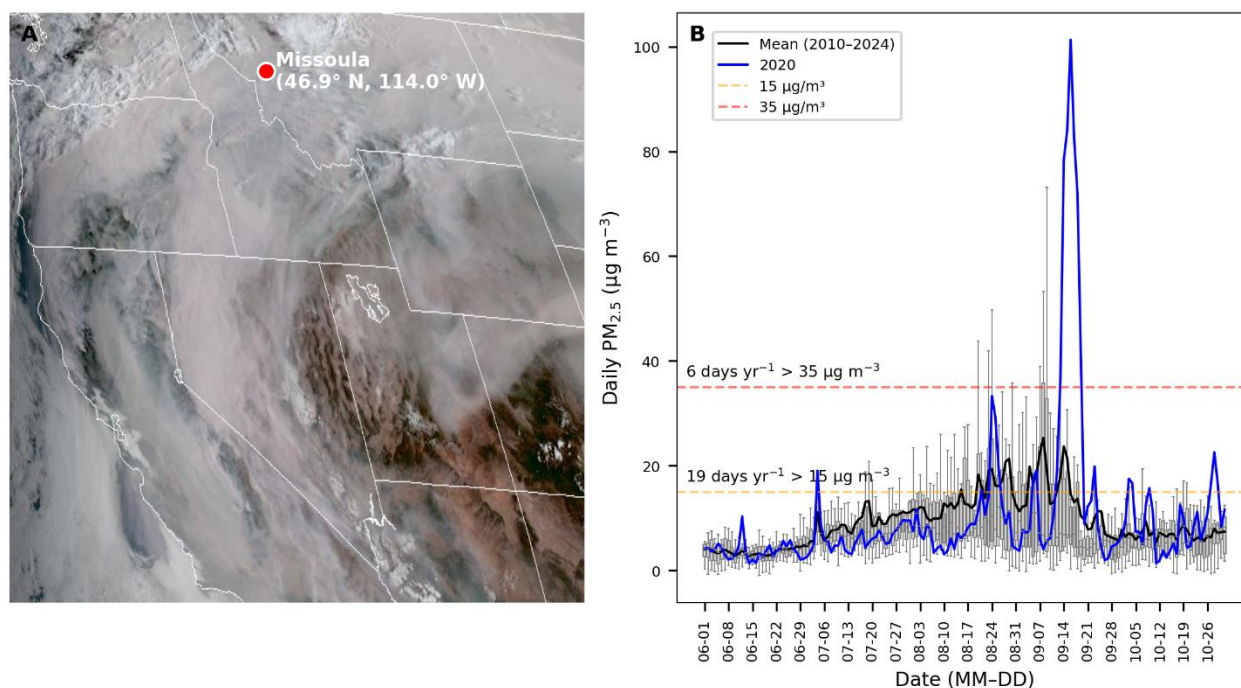


Figure 1. Wildfire smoke over the western US and climatological $\text{PM}_{2.5}$ in Missoula, Montana. (A) Cropped GOES-17 GeoColor satellite image (15 September 2020) over the Western United States, with the location of Missoula, Montana indicated by the red dot. (B) Daily-mean $\text{PM}_{2.5}$ concentrations at Missoula during the 2020 fire season (blue) compared with the 2010–2024 climatology (black line; grey box plots give the interquartile range and whisker range ($\pm 1.5 \times \text{IQR}$; vertical lines) for each calendar day). Dashed horizontal lines indicate 15 and $35 \mu\text{g m}^{-3}$, as the WHO 24-hour guideline and the US EPA 24-hour standard.

2.2 VOC measurements

Ambient mixing ratios of 75 VOCs were measured every 2 minutes using a custom-built Proton Transfer Reaction Time-of-Flight Mass Spectrometer (PTR-ToF-MS; PTR-ToF-4000, Ionicon Analytik GmbH, Innsbruck, Austria). Instrument



operation followed protocols developed in our previous campaigns (Cope et al., 2024; Permar et al., 2021; Selimovic et al., 2022), with drift tube conditions set to $E/N = 130$ Td, 3.00 mbar, 60 °C, and 800 V.

Air was continuously sampled from an inlet ~10 m above ground through ~10 m of heated (60 °C) 6.35 mm (1/4 inch) outer diameter (O.D.) PTFE tubing. The PTR-ToF-MS then subsampled via ~1 m of 1.59 mm (1/16 inch) O.D. PEEK tubing, also maintained at 60°C. The sampling inlet featured a 2- μ m PTFE filter to prevent particle intrusion. Filters were replaced every two weeks, or more frequently during high pollution periods. Instrument backgrounds were quantified every 2.5 hours by sampling VOC-free air generated via a heated platinum catalyst (375 °C; Sigma-Aldrich, 1 wt%).

Calibrations were conducted every other day using dynamically diluted standard gas mixtures for 26 of the 75 VOCs ($\pm 5\%$ at ~1 ppmv; Apel-Riemer Environmental, Inc.). The overall measurement uncertainty for these species is estimated to be less than 15%. Formic and acetic acids were calibrated using permeation tubes, with estimated uncertainties of ~30% (Permar et al., 2021). For 57 uncalibrated species, instrument sensitivities were estimated using theoretical methods (Sekimoto et al., 2017) refined by field comparisons (Permar et al., 2021), yielding overall uncertainties of ~50%. Sensitivity of all compounds used in this study ranges from 0.9 to 14 ncps ppbv⁻¹.

2.3 Measurements of CO, NO_x, O₃, and PM_{2.5}

CO was measured every ~3 minutes using a Reducing Compound Photometer (Peak Performer 1, PEAK Laboratories, Mountain View, CA, USA), which separates CO by gas chromatography and detects it via photometric absorption of mercury vapor produced from CO reduction of HgO. The instrument was calibrated weekly via CO standard in a compressed standard gas cylinder, with a detection limit of 0.3 ppb. The instrument shared the inlet with PTR-ToF-MS and sampled ambient air at 40 sccm through 5 m of 3.175 mm (1/8 inch) O.D. PTFE tubing.

NO₂ and NO were measured every 1 minute using a model 405 nm NO_x monitor (2B Technologies, Boulder, CO, USA), which directly quantifies NO₂ by measuring optical extinction at 405 nm, where NO₂ strongly absorbs. NO is measured by adding excess ozone into the optical cell, which quantitatively converts NO to NO₂. The instrument was calibrated by the manufacturer with a detection limit of 1 ppb for NO and NO₂. Ambient air was drawn at ~1.5 L min⁻¹ through ~1 m of 6.35 mm (1/4 inch) O.D. tubing and passed through a 47 mm, 5 μ m PTFE filter (Savillex), which was replaced biweekly or upon visible loading.

PM_{2.5} and O₃ were obtained from the hourly measurements at the Montana DEQ site at Boyd Park. PM_{2.5} measurements were conducted using a Met One BAM-1020 with VSCC inlet (Volumetric Size-Selective Cyclone), which determines particle mass by measuring the attenuation of beta radiation through the filter tape as particles accumulate. The instrument has a detection limit of 5 μ g m⁻³ for hourly measurements. O₃ was measured using an ultraviolet photometric analyzer (Thermo



Scientific Model 49i UV photometric O₃ analyzer), which quantifies absorption of UV radiation at 254 nm, and with a detection limit of 5 ppb.

2.4 GEOS-Chem chemical transport model

150 We use the nested-grid GEOS-Chem CTM over North America (version 13.3.0; 10-70 °N, and 140-60 °W) to interpret Missoula ground-based measurements. The model is driven by the NASA GEOS-FP meteorological data, with 0.25° × 0.3125° horizontal resolution (~25 km × 30 km; latitude × longitude) and 47 vertical layers extending up to 0.01 hPa (Kim et al., 2015; Wang et al., 2004). Boundary conditions were updated every 3 hours from a 4° × 5° global simulation. Transport and convection time steps were 5 minutes; emission and chemistry time steps were 10 minutes. Model spin-up included a 1-
 155 year global simulation followed by a 1-week nested simulation to remove the influence of initial conditions prior to September 1, 2020.

The chemical mechanism included detailed HO_x-NO_x-VOC-ozone-halogen-aerosol chemistry with fully coupled troposphere and stratosphere (Eastham et al., 2014; Mao et al., 2010; Park, 2004; Schmidt et al., 2016). Dry deposition used a resistance-in-series approach (Wesley and Wesely, 1989), and wet deposition included scavenging of soluble tracers in convective
 160 updrafts, as well as rainout and washout of soluble tracers (Liu et al., 2001). GEOS-Chem uses the non-local scheme for PBL mixing (Lin and McElroy, 2010). As a sensitivity test, we also conducted simulations using the “full mixing” scheme, assuming instantaneous vertical mixing of tracers evenly through the mixing depth, which did not affect the conclusions we derived.

Emissions were computed using the HEMCO module (Keller et al., 2014). These include biogenic VOC emissions from
 165 MEGANv2.1 (Guenther et al., 2012) as implemented in GEOS-Chem (Hu et al., 2015). Anthropogenic emissions are from the CEDS global emission inventory, overwritten with the 2011 EPA NEI inventory for the US (Hoesly et al., 2018). Daily BB emissions were taken from the Global Fire Assimilation System (GFAS) version 1.2, chosen for its relatively better VOC performance when compared to other commonly used BB inventories over the western US (Jin et al., 2023; van der Werf et al., 2017). Following previous work (Jin et al., 2023), we expanded the default GFAS VOC speciation by adding lumped
 170 ≥ C₃ aldehydes (RCHO), MEK, formic acid, and acetic acid by scaling CO BB emissions with corresponding emission ratios reported from temperate forest burns (1.01, 0.73, 9.5, and 8.61 ppb ppm⁻¹, respectively) (Permar et al., 2021). All fire emissions followed a climatological diurnal profile, which allocates ~85 % of daily fire emissions to the afternoon (local time) (Western Regional Air Partnership, 2005).

Biomass burning injection heights are prescribed with the daily 0.1° × 0.1° mean altitude of maximum injection (“mami”) product derived from a satellite-constrained plume rise model (Freitas et al., 2007; Latham, 1994; Rémy et al., 2017). For
 175 each GEOS-Chem grid cell, we computed emission-flux-weighted “mami” values to correct the grid-dependence inherent in the standard model (Jin et al., 2023). BB emissions were then distributed evenly from the surface up to this “mami”.



Overall, four nested simulations were conducted for September 2020: (i) a base run using default GFAS emissions and non-local PBL mixing scheme, (ii) a sensitivity run as the base but without fire emissions (noBB), (iii) a second sensitivity run as the base but with GFAS VOC and CO emissions tripled ($3 \times \text{BB}$), and (iv) a third sensitivity run using GFAS and the full instantaneous mixing within the PBL mixing height.

3 Identification of smoke-impacted events

Wildfire smoke at ground level is often detected using particle-based diagnostics, for example: (i) satellite-derived aerosol products such as the overhead information from the NOAA Hazard Mapping System (HMS) (Jaffe et al., 2022; O'Dell et al., 2021), (ii) column-integrated aerosol optical depth from AERONET, (iii) sustained elevations in surface $\text{PM}_{2.5}$ relative to the general urban background (Selimovic et al., 2019, 2020), and (iv) combinations of these metrics (Kaulfus et al., 2017; McClure and Jaffe, 2018). Some studies also incorporate gas-phase tracers—initially CO, hydrogen cyanide (HCN), acetonitrile (ACN), and more recently, furan, maleic anhydride (MA) (Coggon et al., 2019; de Gouw et al., 2003, 2006; Li et al., 2000). Among these VOCs, furan is a sensitive indicator of fresh BB smoke due to its short lifetime and smaller emission amount from anthropogenic sources. In addition, MA is an oxidation product of furan and its derivatives, with a longer lifetime; thus, it has been proposed as a tracer of aged smoke (Coggon et al., 2019).

Here, we evaluate some current BB-impacted identification metrics in the context of our MSO case study and examine whether our VOC measurements provide additional value in identifying BB events in the urban setting. We first derive the hourly median diurnal urban background for BB tracers and other species using HMS “no-fire” days for September 2020. Any enhancement (ΔX) > 1.5 times the background is considered BB-impacted, and a provisional smoke day is defined when at least six such hours occur. We also tested the $\text{PM}_{2.5}/\text{CO}$ criterion ($> 30 \mu\text{g m}^{-3} \text{ppm}^{-1}$) proposed for urban smoke identification (Jaffe et al., 2022). However, at MSO this threshold classified nearly all days as smoke-impacted, consistent with the limitation that $\text{PM}_{2.5}/\text{CO}$ becomes non-discriminating when background $\text{PM}_{2.5}$ and CO are low (Jaffe et al., 2022). We therefore do not use $\Delta\text{PM}_{2.5}/\Delta\text{CO}$ as a standalone classifier for this site.

Figure 2 summarizes the daily BB classification for September 2020 in Missoula. HMS alone identifies 20 smoke days (the greatest number among the diagnostics), but it can flag a day as smoke-impacted even when surface concentrations remain low if smoke is primarily aloft. For example, HMS triggers on 2 September and 30 September (daily average $\text{PM}_{2.5}$ is 8.5 and $5.5 \mu\text{g m}^{-3}$, respectively), which likely reflect elevated plumes that were not mixed down to the surface (Liu et al., 2024). By comparison, the $\Delta\text{PM}_{2.5}$ criterion identifies 16 days. Its diagnostic power depends strongly on aerosol loading at the surface.

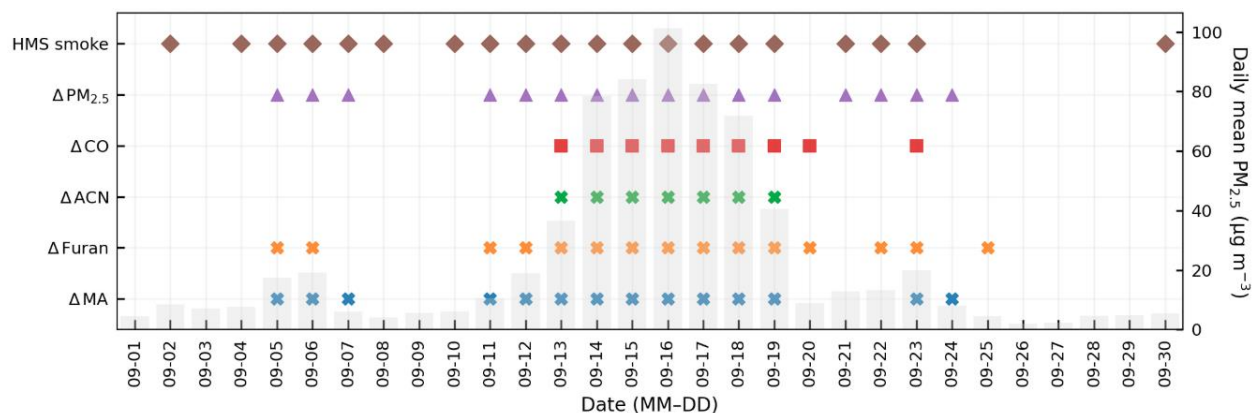


When daily mean $\text{PM}_{2.5}$ exceeded $35 \mu\text{g m}^{-3}$ ($n = 7$; 13–19 September), the $\Delta\text{PM}_{2.5}$ approach aligned with HMS and with all
210 gas-phase tracers considered here (i.e., CO, ACN, furan, and MA), indicating concentrated smoke that coupled large aerosol
mass with gaseous enhancements. A k-means clustering analysis ($k = 3$: smoke, no-smoke, uncertain) applied to pairwise
combinations of CO, $\text{PM}_{2.5}$, and each tracer corroborated this classification, reinforcing that at extreme aerosol loadings,
routine ground-level $\text{PM}_{2.5}$ alone can reliably identify smoke during summer in Missoula.

215 At intermediate daily mean $\text{PM}_{2.5}$ between 20 and $35 \mu\text{g m}^{-3}$ ($n = 5$; 5–6, 11–12, and 23 September), $\Delta\text{PM}_{2.5}$ still overlaps at
least two gas-phase tracers but not all, signaling moderately aged or dispersed smoke and therefore medium diagnostic
confidence. When the daily-mean $\text{PM}_{2.5}$ falls below $20 \mu\text{g m}^{-3}$ ($n = 4$; 7, 21–22, and 24), $\text{PM}_{2.5}$ overlaps with at most one gas
tracer, and HMS does not indicate smoke on 24 September, so these low-load cases warrant only low-to-medium confidence.
One additional case (21 September) meets the $\text{PM}_{2.5}$ threshold but lacks corroboration from the gas-phase tracers, suggesting
220 a potential false positive, although HMS indicates overhead smoke. Across September (a total of 30 days), the remaining 14
days do not meet the $\Delta\text{PM}_{2.5}$ criterion and are classified as non-smoke by this approach.

Gas-phase tracers extend the detectability of wildfire smoke beyond what is captured by HMS and $\text{PM}_{2.5}$ alone, particularly
during precipitation events. For example, on 20 September, CO and furan enhancements were observed in the absence of
225 corroborating HMS smoke or elevated $\text{PM}_{2.5}$. A rain event from the preceding day into the afternoon of 20 September
efficiently scavenged aerosol mass, resulting in a sharp decrease of $\sim 30 \mu\text{g m}^{-3}$ in $\text{PM}_{2.5}$, whereas gas-phase species were
less affected and their enhancements persisted in the boundary layer.

Because no single tracer is definitive, we adopt a balanced multi-indicator rule: an hour is classified as smoke-impacted
230 when at least three independent tracers exceed their thresholds, and a day is smoke-impacted if it contains six or more such
hours. Applied to September 2020, this criterion identified 14 smoke days (5–7, 11–20, and 22–23 September). For
subsequent sections, we describe the three smoke events as an overview, while we focus on the high-confidence BB period
of 13–19 September to analyze its enhancement ratios, thereby minimizing uncertainties from marginal or non-BB sources.



235 **Figure 2. Wildfire smoke identification in Missoula, Montana, during September 2020.** Rows show the six individual smoke metrics evaluated in this study. A filled marker indicates that HMS smoke was detected, or that the enhancement of a biomass burning (BB) tracer (ΔX) exceeded both $1.5\times$ its diurnal background and the metric-specific threshold for at least six hours on that calendar day. Gray bars (right axis) show the daily mean $PM_{2.5}$ concentration.

4 Overview of BB smoke in September 2020

240 Wildfires across the Pacific Northwest and northern California were exceptionally active in 2020 (Albores et al., 2023; Higuera and Abatzoglou, 2021; Neyra-Nazarrett et al., 2025; Reilly et al., 2022), and September delivered strong smoke signatures in Missoula. According to the U.S. EPA AirNow data, air quality index (AQI) values failed to reach the "Good" category on 19 out of 30 days. Specifically, 12 days were classified as "Moderate" and 7 days fell into "Unhealthy" or "Unhealthy for Sensitive Groups", due to wildfire emissions (US EPA AirNow, 2024). Applying the three-tracer
 245 classification of Section 3, smoke affected 16 days (or 384 hrs) of our measurements in September.

Figures 3 and S1 show three distinct multi-day smoke episodes affecting Missoula identified using the criteria described in Sect. 3, including 5–7 September (Event 1), 11–20 September (Event 2), and 22–24 September (Event 3). Because each episode involved multiple overlapping plumes from different fires with varying transport times, we refer to them as "events" rather than discrete "plumes". Analysis of GOES-17 and VIIRS satellite imagery, and synoptic meteorological conditions
 250 indicate that these events were predominantly driven by southwesterly transport of wildfire smoke from Oregon, Idaho, and California, with likely influence from local smaller fires.

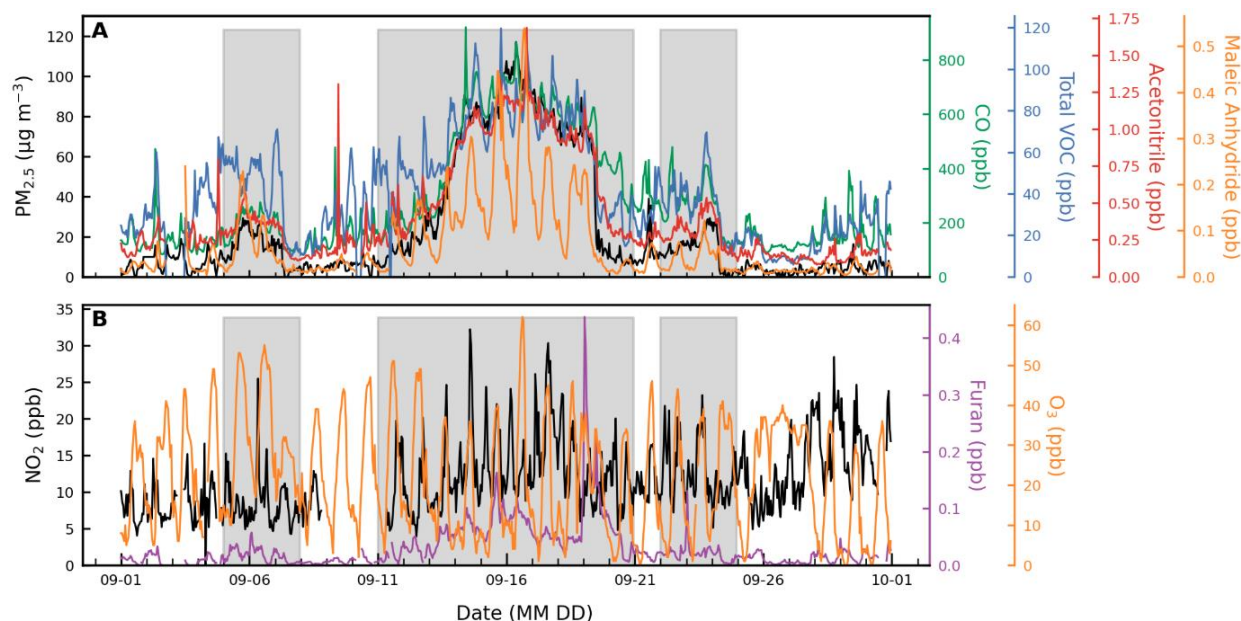
Event 1 (5–7 September) resulted from the California Creek Fire (United States Forest Service, 2025), which started on 4 September mixing with a localized, narrow plume moving east-northeast over the Bitterroot Mountains toward Missoula, as
 255 captured by GOES-17 imagery. Correspondingly, hourly $PM_{2.5}$ increased rapidly from <10 to $30 \mu g m^{-3}$ by 06:00 MDT on 6 September and CO from 100 to ~ 300 ppb by 09:00 MDT the same day, with brief spikes (≤ 50 ppt) of the fresh-smoke tracer

furan at 01:00 MDT. Pollutant concentrations returned to the urban background within 24 h after a post-frontal north-westerly wind flushed the valley.

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Event 2 (11–20 September) was the longest and strongest impact on Missoula from the Oregon–California “megafire” complex that ignited in August and erupted on 7 September (Abatzoglou et al., 2021). Intense smoke on 8 September was advected westward offshore, where it became entrained into a cut-off low over the NE Pacific (around 42°N, 135°W), and subsequently advected eastward inland, mixing with smoke freshly emitted by the megafire complex and fires in ID. By 11 September, dense smoke formed a clearly visible “smoke river” that stretched across Washington, Oregon, and Idaho into western Montana. Back-trajectory analyses indicate that air parcels reaching Missoula had aged for at least two days. Consistently, smoke-impacted hours during this event exhibited pollutant enhancements of four- to fifteen-fold for CO, PM_{2.5}, acetonitrile, and total measured VOCs (TVOCs), reaching peak values of 800 ppb CO, 120 µg m⁻³ PM_{2.5}, 2 ppb acetonitrile, and 85 ppb TVOCs in mid-September. Strong correlation ($r^2=0.9$; Fig. S2) among PM_{2.5} measured at the Boyd Park and other tracers measured at the UM campus indicates thorough smoke mixing. Brief furan spikes (≤ 0.7 ppb) on 12–13 and 18–19 September suggest additional influence from local fires. Overall, this event accounts for ~80 % of smoke hours in this month and is the focus of the enhancement ratio analysis (Sect. 5).

Event 3 (22–24 September) was a weaker resurgence of smoke from the same Oregon–California fires in Event 2. A preceding rain event on 20 September removed a substantial amount of aerosol mass, as indicated by the sharp drop in PM_{2.5} in Fig. 3, whereas most gas-phase species persisted. Subsequent south-westerly flow reintroduced the residual, aged smoke into the region. As a result, 24-h PM_{2.5} concentrations reached 40 µg m⁻³ and hourly CO peaked at 350 ppb before a front cleared the valley, which is especially evident for PM_{2.5} but still leaving fire influences for trace gases.



280 **Figure 3. Time series of selected VOCs and criteria pollutants in Missoula, Montana, during September 2020.** (A) Hourly measurements of $\text{PM}_{2.5}$ (black; left axis), carbon monoxide (CO; green, right axis), total measured VOCs (blue, right axis), acetonitrile (red, right axis), and maleic anhydride (orange, right axis). (B) Concurrent hourly NO_2 (black; left axis), furan (purple; right axis), and ozone (orange, right axis). Gray shading indicates smoke days based on the criteria in Sect. 3.

Figure 4 compares pollutant concentrations during smoke-impacted and background periods in September 2020. On average,
 285 MDA8 ozone increased by 3 ppb (7%) during smoke episodes, while NO_x showed no visible change. In contrast, CO, $\text{PM}_{2.5}$, and TVOCs were significantly elevated, with hourly concentrations increasing by a factor of 3–8 compared to urban background. Average hourly CO concentrations increased from 160 ± 80 ppb (mean $\pm$ SD of hourly data) to 420 ± 205 ppb on smoke days. $\text{PM}_{2.5}$ showed even larger enhancements, rising from 6.1 ± 4.5 $\mu\text{g m}^{-3}$ to 43 ± 34 $\mu\text{g m}^{-3}$. The background CO and $\text{PM}_{2.5}$ concentrations agree with typical western US urban levels (150–200 ppb and 5–10 $\mu\text{g m}^{-3}$) (Cope et al., 2024;
 290 Lill et al., 2022; Lopez-Coto et al., 2020; Pfister et al., 2011).

Measured hourly TVOCs tripled from 23 ± 15 ppb to 55 ± 24 ppb, with individual species enhancements summarized in Table S2. Ten VOCs increased by factors of ~4–7, five of which were furanoids or their derivatives. These species included furfural, methylfurfural, 2-furanmethanol, 2-furanone, 5-hydroxy-2-furfural/2-furoic acid, maleic anhydride, acrylonitrile,
 295 methyl benzoic acid, methyl methacrylate, and anisole. Forty-eight VOCs increased by a factor of 2–4, while 16 species increased by a factor of 1–2. Fifteen of the 75 measured VOCs are on the EPA's list of HAPs, and they collectively represented 51% of TVOCs by average molar mixing ratio. During smoke events, total HAPs (THAPs) increased threefold relative to background periods (28 ± 13 ppb vs. 11 ± 7 ppb), with individual species exhibiting enhancements ranging from 40% (dichlorobenzene) to nearly fivefold (acrylonitrile and maleic anhydride). Figure S3 shows that OH reactivity (OHR)
 300 from CO and TVOCs is doubled during smoke-impacted days compared to background (14.2 s $^{-1}$ vs. 7.1 s $^{-1}$). During



background periods, the OHR was dominated by the biogenic tracer isoprene (0.89 s^{-1} ; 12.5 % of total OHR), followed by
monoterpenes (9.6%), formaldehyde (8.3 %), CO (8.1 %), and other individual VOCs. During smoke-impacted days, CO
became the largest contributor (1.4 s^{-1} ; 10 %), and the relative importance of monoterpenes fell to 5.6 %, consistent with
previous urban smoke OHR analyses in Boise, ID (Permar et al., 2023). Other VOC groups show similar contributions to
OHR in both periods. Together, these enhancements underscore the strong influence of regional wildfire smoke on western
US air quality and motivate continued monitoring of smoke exposure in communities such as Missoula.

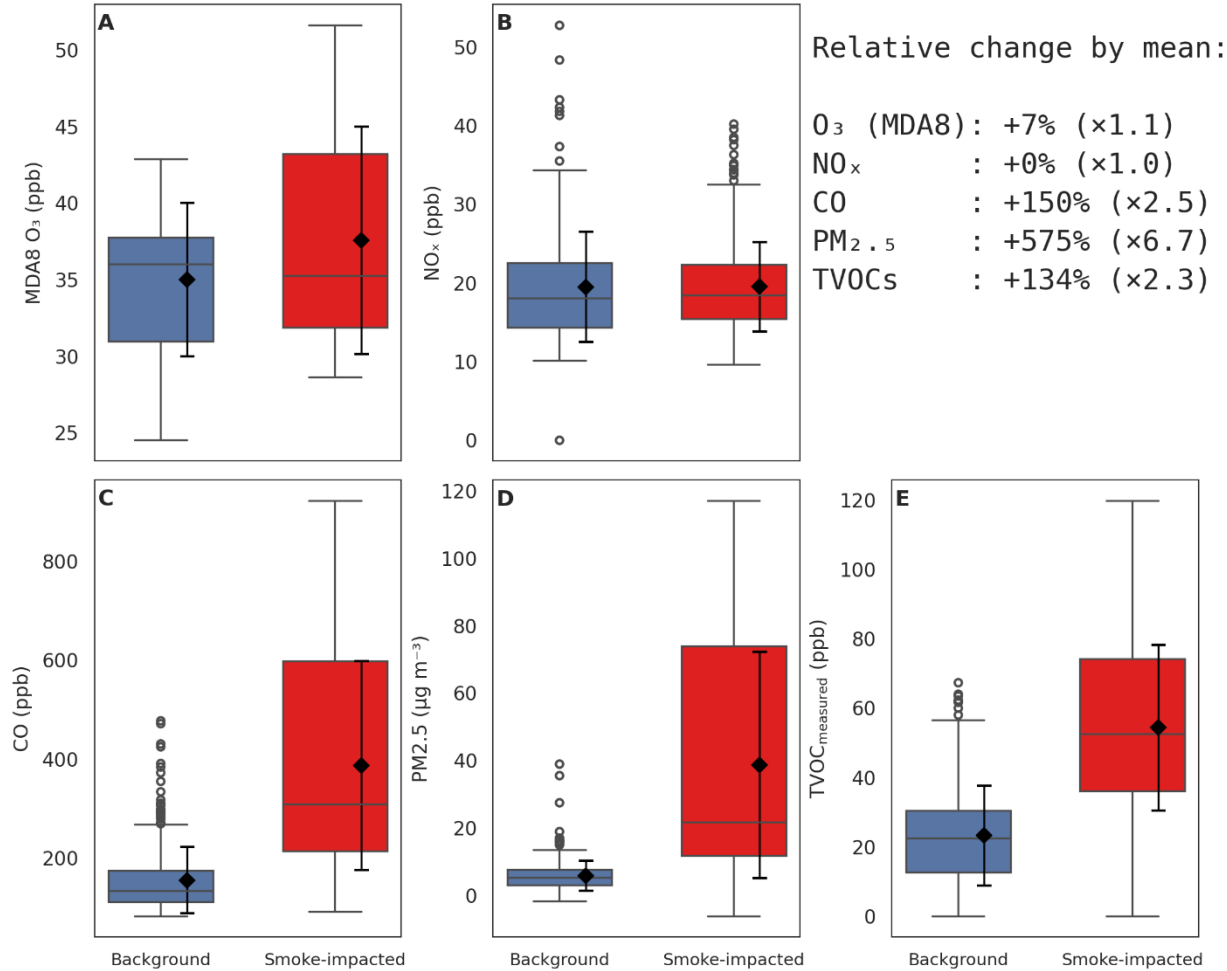


Figure 4. Enhanced surface-level pollutant concentrations during smoke-impacted days. Boxplots of hourly surface concentrations of (A) Maximum daily 8-hour average ozone (MDA8 O_3), (B) nitrogen oxides (NO_x), (C) carbon monoxide (CO), (D) fine particulate matter ($\text{PM}_{2.5}$), and (E) total measured volatile organic compounds (TVOCs) during smoke-impacted (red) and background (blue) periods observed in Missoula, Montana, during September 2020. Boxes represent interquartile ranges (25th–75th percentiles) with medians shown as center lines; whiskers extend to $1.5\times$ the interquartile range. Black diamonds and error bars indicate mean $\pm$ standard deviation for each group. The right-hand summary block lists mean enhancement factors (smoke-impacted $\div$ background) for each pollutant.



5 Smoke enhancement ratios (EnRs) and emission ratios (ERs)

315 We here combine enhancement ratios (EnRs) with a photochemical clock to assess smoke aging for all measured VOCs. Figure 5 illustrates the $\ln(\text{EnR})$ -age framework used to retrieve emission ratios (ERs) and effective OH rate constants (k_{OH}) for representative primary aromatics, where the regression intercept and slope correspond to ER and k_{OH} , respectively (method in Sect. S1).

320 From the photochemical clock analysis of aged smoke reaching Missoula, we infer initial ERs of 2.05 ± 0.02 ppb ppm⁻¹ for benzene, 1.42 ± 0.02 ppb ppm⁻¹ for toluene, and 1.20 ± 0.05 ppb ppm⁻¹ for C₈ aromatics. We also infer ERs of 0.41 ± 0.03 ppb ppm⁻¹ for C₉ aromatics and 0.18 ± 0.01 ppb ppm⁻¹ for C₁₀ aromatics. For C₃–C₅ alkenes (propene, butenes, and pentenes/methylbutenes) and 1,3-cyclopentadiene, inferred ERs are 1.19 ± 0.04 , 1.45 ± 0.05 , 0.17 ± 0.01 , and 0.10 ± 0.01 ppb ppm⁻¹, respectively.

325 The inferred benzene and toluene ERs agree with near-source ranges reported from the WE-CAN and FIREX-AQ campaigns within ~20% (benzene: 1.8–2.3; toluene: 1.2–1.5 ppb ppm⁻¹) (Gkatzelis et al., 2024; Permar et al., 2021). In contrast, inferred ERs for C₈–C₁₀ aromatics are ~2–3× higher than published near-source wildfire ERs. These higher apparent aromatic ERs likely reflect mixing with urban emissions during transport; consistent with this interpretation, city-based
 330 measurements report elevated aromatics relative to CO during wildfire-smoke periods (Cope et al., 2024). Comparable literature ERs are not available for the small alkenes and 1,3-cyclopentadiene and thus not discussed further here.

The relative decay rates also broadly followed their OH rate constants (k_{OH}). For example, benzene exhibited only modest decay over one week of photochemical aging, whereas toluene and C₈ aromatics with higher k_{OH} decayed more rapidly. The
 335 k_{OH} values inferred from the photochemical clock were $(9.8 \pm 1.6) \times 10^{-13}$ and $(5.6 \pm 0.2) \times 10^{-12}$ cm³ molecule⁻¹ s⁻¹ for benzene and toluene, respectively, agreeing with literature within ~10–20%, although this agreement is partly expected because photochemical age is derived from the toluene-to-benzene clock. For C₈ aromatics, the inferred k_{OH} (7.9×10^{-12} cm³ molecule⁻¹ s⁻¹) lies between those of ethylbenzene and the xylene isomers, consistent with expectations for a lumped mixture.

340 In contrast, effective k_{OH} inferred for several more reactive hydrocarbons (e.g., C₉–C₁₀ aromatics and small alkenes) are lower by factors of 3 or more than literature. This low bias is consistent with ground-based sampling of aged wildfire smoke and likely reflects preferential loss of highly reactive VOCs during the first few hours after emission, when OH concentrations are highest; thus, the plume sampled at MSO represents the surviving fraction of the original emissions.
 345 Additional contributions may arise from dilution and mixing with urban air, as well as uncertainties associated with background correction and species lumping.

We find that EnRs of primary VOCs decrease significantly with photochemical age ($p \ll 0.01$), consistent with OH-driven oxidation. In contrast, oxygenated VOCs (OVOCs) such as methanol, formaldehyde, and acetaldehyde exhibit weaker and often statistically insignificant $\ln(\text{EnR})$ –age trends ($p \approx 0.02$ – 0.21), and in several cases the relationships flatten with age, consistent with secondary production partially offsetting chemical loss during plume evolution. As a result, the photochemical clock framework is well suited for constraining ERs (and effective k_{OH}) for primary VOCs, but it cannot be directly used to derive ERs for OVOCs because net production violates the assumption of purely first-order decay (Sect. S1). Overall, photochemical clocks provide useful constraints on ERs for many primary VOCs in aged smoke but can overestimate BB contributions for species with strong non-fire sources.

Additionally, we report the event-integrated EnRs of $\text{PM}_{2.5}$ and VOCs relative to CO. The $\text{PM}_{2.5}$ EnR in several-day-aged smoke was $0.11 \pm 0.01 \text{ g g}^{-1}$ at Missoula, consistent with previous surface measurements (0.11 – 0.13 g g^{-1}) but about half of high-elevation/aircraft values (Pagonis et al., 2023; Selimovic et al., 2019, 2020). We derived event-integrated EnRs for 27 VOCs, excluding species weakly correlated with CO ($R^2 < 0.4$); corresponding species-level EnRs and literature comparisons are summarized in Table S3 and Fig. S4.

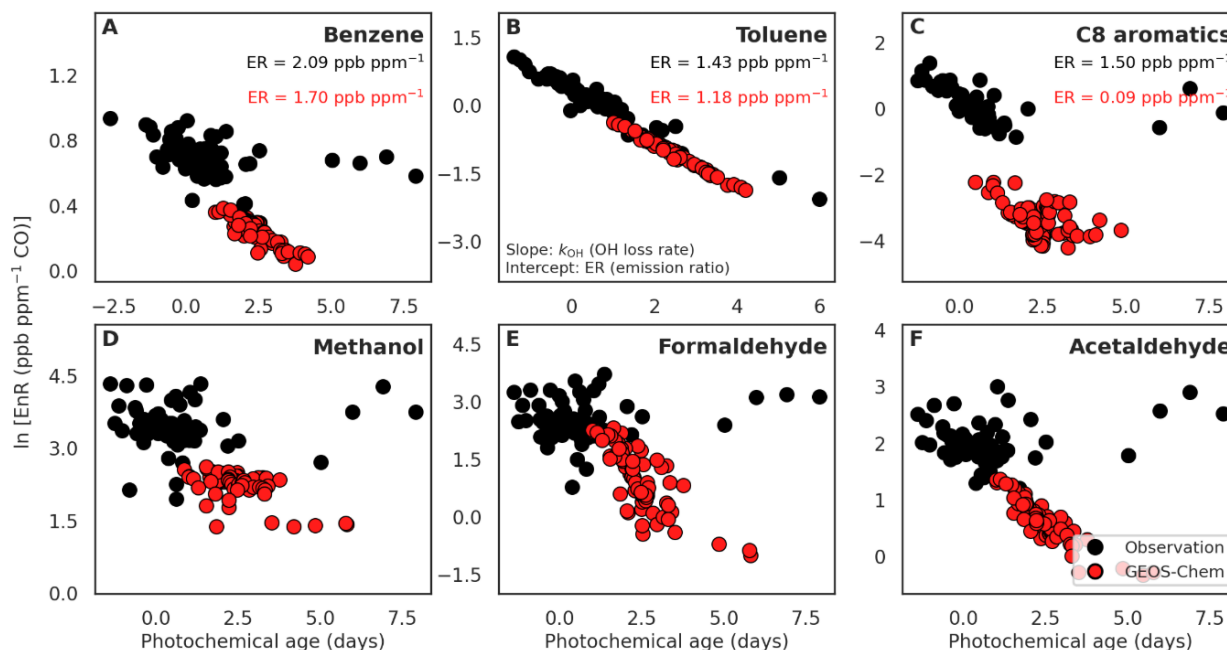


Figure 5. Observed and modeled enhancement ratios (EnR) of key volatile organic compounds (VOCs) versus photochemical age. Panels show $\ln(\text{EnR})$ versus photochemical age for (A) benzene, (B) toluene, (C) C₈ aromatics, (D) methanol, (E) formaldehyde, (F) acetaldehyde, where EnR is defined as $\Delta\text{VOC}/\Delta\text{CO}$ (ppb ppm^{−1} CO). Observations are shown as black circles and results from GEOS-Chem driven by GFAS fire emissions are shown as red circles. Points represent 3 h binned data.

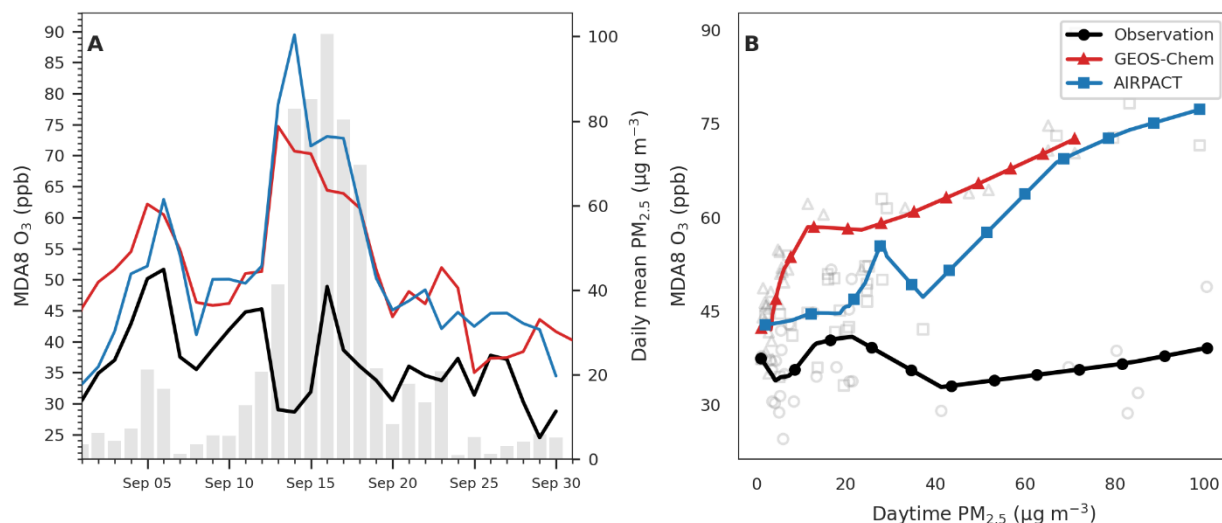


6 Non-monotonic O₃–PM_{2.5} relationship

Figure 6 shows the relationship between maximum daily 8-hour average (MDA8) O₃ and daytime mean PM_{2.5} at Missoula during September 2020. During the strongest smoke episodes, observed MDA8 O₃ was suppressed by up to ~15 ppb (e.g., 6–
 370 7 September; Fig. 6A). These decreases coincide with ~50% reductions in downwelling shortwave radiation (SW) (Fig. S5A), a first-order proxy for actinic flux and photolysis $J(\text{NO}_2)$ and $J(\text{O}^1\text{D})$, indicating attenuated photolysis reduces radical production and suppresses in-situ ozone formation. NO_x levels were not substantially different from the urban background. In addition, heavy smoke was associated with lower observed daytime maximum temperature and a shallower planetary boundary layer height (PBLH) (Fig. S5B–S5C), which may further reduce surface ozone by slowing reaction rates and
 375 weakening entrainment of O₃-rich air aloft.

Across BB days, the relationship between MDA8 O₃ and daytime mean PM_{2.5} is non-monotonic (Fig. 6B). A locally weighted scatterplot smoothing (LOWESS) curve suggests that MDA8 O₃ increases at low PM_{2.5} (< ~20 µg m⁻³), decreases at moderate PM_{2.5} (~20–60 µg m⁻³), and may increase again at higher PM_{2.5} (>60 µg m⁻³) despite only four daily points exist.
 380 The non-monotonic relationship likely reflects a transition from precursor-driven O₃ production under light smoke (< ~20 µg m⁻³) to aerosol-dominated suppression of net O₃ production at moderate aerosol loadings (>~20–60 µg m⁻³).

The non-monotonic behavior is not unique to Missoula but is also evident across other northwestern sites during the same September 2020 period. At Cheeka Peak (WA) and Eugene (OR), where they were near the BB source during that month
 385 and daily PM_{2.5} reached up to 250 and 450 µg m⁻³ respectively, O₃ similarly increases with PM_{2.5} under low smoke but levels off or declines at higher aerosol loadings (Fig. S6). Such nonlinear O₃–PM_{2.5} relationships have been reported across western U.S. smoke observations (Buysse et al., 2019; McClure and Jaffe, 2018), reflecting similar controlling factors that affect the O₃-smoke interaction.



390 **Figure 6. Temporal evolution and $PM_{2.5}$ dependence of maximum daily 8-hour average ozone during September 2020.** (A) Time
 series of daily maximum 8-hour average ozone (MDA8 O_3 , ppb) from surface observations (black), GEOS-Chem driven by GFAS fire
 emissions (red), and the AIRPACT forecast system (blue). Shaded bars (right axis) show observed daily mean $PM_{2.5}$ ($\mu g m^{-3}$), providing
 context for smoke influence. (B) Relationship between MDA8 O_3 and daytime $PM_{2.5}$ for observations (circles), GEOS-Chem (triangles),
 395 and AIRPACT (squares). Open gray symbols show individual daily values for observation and model simulations. Solid curves denote
 locally weighted regression (LOWESS) fits applied consistently across datasets (smoothing fraction = 0.5; robust iteration = 3).
 AIRPACT outputs are sampled in space and time at the monitoring locations and were obtained from the Washington State University
 AIRPACT data portal (<https://airpact.wsu.edu/>).

7 Chronic and acute health risks

Figure 7 summarizes the upper-limit chronic inhalation risks from wildfire smoke, assuming that a 2020-style wildfire
 400 season were to recur annually over the next century. Over lifetime exposure (70 yr), the total excess cancer risk attributable
 to wildfire smoke is 100 cases per million people. Roughly 90 % of this smoke-driven risk is attributed to $PM_{2.5}$, with the
 remaining 10 % arising from HAPs combined. Among HAPs the major contributors are formaldehyde (42 % of HAP cancer
 risk), benzene (34 %), acetaldehyde (8 %), acrylonitrile (9 %), and naphthalene (6 %). These relative contributions agree
 with recent aircraft observations of lofted plumes and confirm that $PM_{2.5}$ dominates cancer risk both aloft and at ground level
 405 (Pye et al., 2024).

For context, the climatological annual-mean non-smoke background $PM_{2.5}$ is $\sim 6 \mu g m^{-3}$ in Missoula. If the BB-impacted
 days instead experienced typical non-smoke conditions, the excess cancer risk would be ~ 15 cases per million people. Thus,
 the 2020-style wildfire season increases the $PM_{2.5}$ attributable cancer risk by $\sim 7\times$ relative to baseline. Even using a 10-fold
 410 lower $PM_{2.5}$ unit-risk estimate ($4.8 \times 10^{-5} \mu g^{-1} m^3$), $PM_{2.5}$ still accounts for $\sim 50\%$ of the total cancer risk, and the total risk
 remains ~ 20 cases per million people (i.e., $\sim 30\%$ higher the baseline). Thus, even when the community's exposure to



wildfire smoke is annualized and using a lower-bound $PM_{2.5}$ unit-risk estimate, aged wildfire smoke as the level experienced in September 2020 in Missoula still shows a quantifiable carcinogenic burden.

415 Non-cancer chronic risk is expressed as a hazard index (HI), with the dominant effects in this study associated with respiratory and cardiopulmonary systems. The calculated $HI = 3$ indicates appreciable potential for adverse effects. HAPs account for ~90 % of this index, driven by acrolein (60 % of HAP non-cancer risk), formaldehyde (24 %), acetaldehyde (3 %), and all other species (< 1 % each). $PM_{2.5}$ contributes to the remaining ~10 %. The result contrasts with aircraft-based assessments, in which $PM_{2.5}$ dominated non-cancer risk (O'Dell et al., 2020; Pye et al., 2024). The lower relative
420 contribution of $PM_{2.5}$ observed here likely reflects the evaporation of semi-volatile particulate mass as plumes descend from aloft to the surface (Pagonis et al., 2023; Selimovic et al., 2019, 2020). If $PM_{2.5}$ were assumed 10-times more toxic ($RfC = 0.5 \mu g m^{-3}$, comparable to acrolein), it would instead contribute ~70 % of the HI.

No acute reference exposure levels (RELs) are available for $PM_{2.5}$, so the acute HI analysis was restricted to HAPs. During
425 September 2020, 719 of 720 one-hour windows exhibited $HI < 1$, implying negligible acute non-cancer concern. A single exceedance (19 September, $HI = 1.05$) was apportioned to acrolein (52 %), formaldehyde (32 %), benzene (12 %), and other HAPs (< 5 %). Expanding to eight-hour windows (Fig. S7), 49 % of periods remained below the $HI = 1$ threshold, whereas 51 % exceeded it, aligned with smoke incursions. Of the exceedances, 30 % fell within $HI = 1-2$ and 21 % reached up to $HI = 5$. The eight-hour hazards were driven by formaldehyde (38 %), acrolein (31 %), and benzene (29 %), with all other HAPs
430 contributing < 2 %.

The recurrence of the same three drivers across acute and chronic metrics highlights two practical needs. First, smoke-impacted communities would benefit from continuous, high-time-resolution monitoring of formaldehyde, acrolein, and benzene to track rapidly evolving exposures and guide advisories. Second, because particle-only strategies do not address
435 these gases, indoor interventions should pair particle filtration (HEPA or MERV 13+ for $PM_{2.5}$) with sorbent media (e.g., activated carbon) when using indoor air cleaners during smoke events (Maximoff et al., 2022; May et al., 2021).

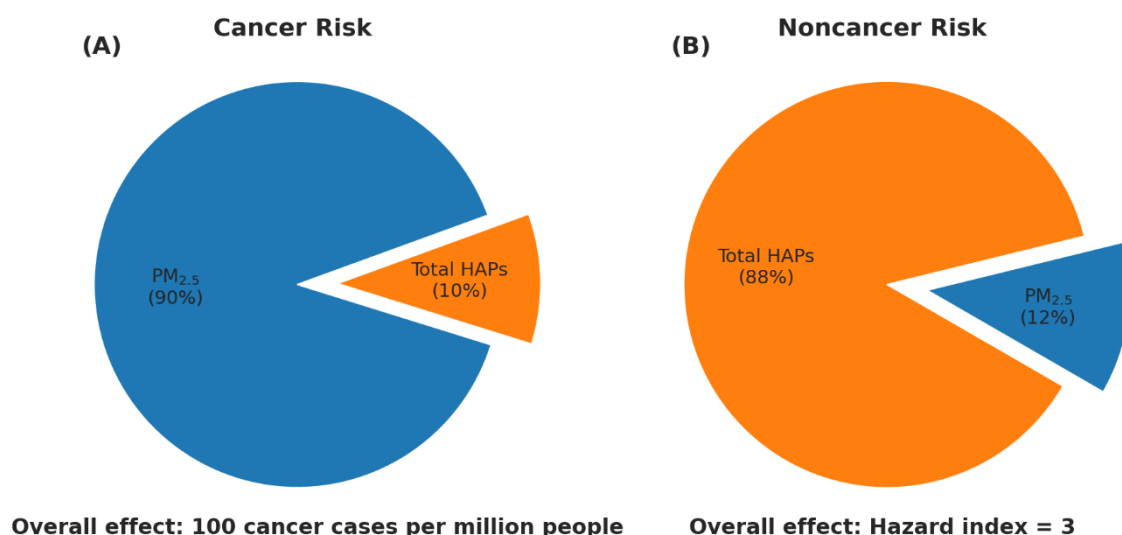


Figure 7. Chronic health-risk from fine particulate matter (PM_{2.5}) and hazardous air pollutants (HAPs). (A) Estimated excess cancer risk (cases per million people) attributed to ambient PM_{2.5} versus all combined HAPs. (B) Noncancer hazard index (HI) attributed to ambient PM_{2.5} versus combined HAPs. In each panel, slices are exploded for clarity, and labels show the percentage of total risk contributed by each component. The bottom annotation reports the overall risk magnitude: total cancer cases per million for panel A and HI for panel B.

8 Model evaluation

8.1 Model uncertainties in fire emissions and OH exposure

Figure 8 compares hourly observations of CO, PM_{2.5}, and four representative VOCs with model simulations; additional VOC comparisons are shown in Fig. S8. The default GEOS-Chem simulation captures smoke-impacted enhancements of CO, PM_{2.5}, and VOCs during the first two events (5–7 September and 11–20 September), with high correlations ($R^2 = 0.8$ – 0.9 for hourly measurements). The agreement indicates that the model generally captures the timing, transport, and location of fire smoke. However, the model fails to reproduce the observed enhancements during Event 3 (22–24 September), likely reflecting overly efficient modeled wet scavenging during the 21 September rainout and/or unresolved smoke transport pathways discussed in Sect. 4. Even during Events 1–2, GEOS-Chem underestimates the magnitude of fire impacts, with low biases of 30–40% for CO and PM_{2.5}, 60–80% for primary aromatics, and 50–90% for most OVOCs. Model performance during background periods is notably better, suggesting BB-related emissions and/or chemistry drive the majority of these underestimations under smoky conditions. MEK is the only exception, showing an average high bias of ~20%, reflecting overestimated anthropogenic or biogenic emissions, as pointed out in early studies (Chen et al., 2019; Jin et al., 2023).

GEOS-Chem systematically underestimates event-integrated VOC EnRs by 35% for benzene and 60–80% for other VOCs (Fig. S9), despite ERs being reproduced within ~20–40% in Fig. 5 and other work (Jin et al., 2023). This ER–EnR decoupling



points to chemistry-related biases: the model overpredicts plume photochemical age (Fig. 5) and OH exposure (by ~100%;
 Fig. S10), which over-oxidizes VOCs relative to CO and depresses EnRs. For OVOCs, the negative bias of EnRs likely also
 reflects missing secondary production, particularly for methanol, formaldehyde, and acetone, which exhibit multi-day
 growth in aged smoke in Fig. 5 and previous work (Alvarado et al., 2020; Bates et al., 2021; Holzinger et al., 2005; Jin et al.,
 2023). By contrast, GEOS-Chem overestimates the event-integrated $PM_{2.5}$ EnR by ~33%, implying excessive $PM_{2.5}$
 produced (or retained) per unit CO, consistent with the reported biases in the simplified GEOS-Chem SOA scheme used here
 (Oak et al., 2022; Pai et al., 2019).

Motivated by aircraft constraints that standard inventories underpredict BB emissions by ~3× (Jin et al., 2023), we conducted
 a 3×BB sensitivity test, which reduces modeled OH by ~2× and lowers the OH-exposure bias from ~100% to ~30% during
 Missoula smoke episodes. The remaining modeled OH bias likely reflects missing OH reactivity from unrepresented
 compounds such as furanoids (Permar et al., 2023). Correspondingly, gas-phase NMBs improve (CO: -40% to +20%;
 benzene: -70% to -10%; toluene: -80% to -50%). Several OVOCs remain substantially underestimated (e.g., methanol
 -80% to -50%; formaldehyde -70% to -40%; acetaldehyde -75% to -35%; acetic acid -80% to -45%; acetone -60% to
 -40%), consistent with missing secondary production as the cause of the persistent OVOC low bias. Meanwhile, $PM_{2.5}$ shifts
 from a -30% bias to a +80% bias, indicating that a uniform BB scaling cannot reconcile gases and aerosol. It also points to
 limitations in the default GEOS-Chem “simple SOA” fire treatment, where the SOA precursor tracer (SOAP) is
 parameterized proportional to BB CO; scaling fire CO therefore scales SOAP and can over-amplify OA and $PM_{2.5}$ during
 smoke events.

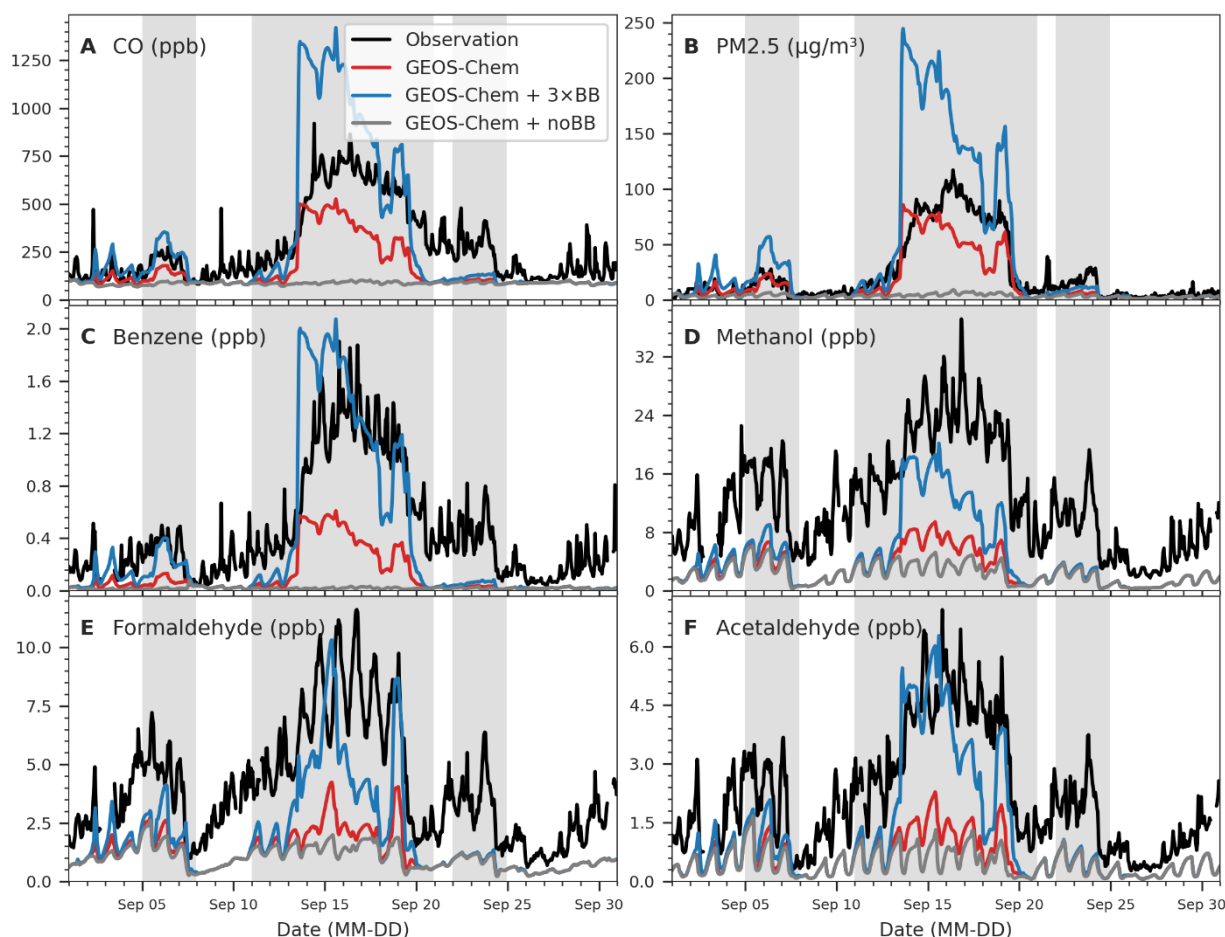


Figure 8. Time series of observed and modeled concentrations for key pollutants during September 2020 in Missoula, Montana. Observations (black lines) are compared with the base GEOS-Chem simulation (red). Also shown are two model sensitivity tests: one with tripled biomass burning emissions (GEOS-Chem + 3×BB; blue) and one with biomass burning emission turned off (GEOS-Chem + noBB; gray). Panels show hourly averaged concentrations of (A) carbon monoxide (CO), (B) fine particulate matter (PM_{2.5}), (C) benzene, (D) methanol, (E) formaldehyde, and (F) acetaldehyde. Model outputs are sampled at the observational location and time. Gray shading indicates smoke-impacted days based on the criteria in Sect. 3. The modeled PM_{2.5} is calculated offline from the SpeciesConc diagnostic as the sum of inorganic ions ($1.10 \times [\text{NH}_4^+ + \text{NO}_3^- + \text{SO}_4^{2-}]$), black carbon (BCPI + BCPO), organic matter ($(\text{OCPO} + 1.05 \times \text{OCPI}) \times \text{OM:OC}$), fine dust ($\text{DST1} + 0.30 \times \text{DST2}$), sea salt ($1.86 \times \text{SALA}$), and secondary organic aerosol ($1.05 \times (\text{SOAS} + \text{SOAP} + \text{SOAIE} + \text{SOAGX})$).

8.2 Model uncertainties in ozone formation under high PM_{2.5}

GEOS-Chem overestimates MDA8 O₃ in September of 2020 by ~15 ppb on average and fails to capture the observed day-to-day variability (Fig. S11), regardless of the BB emission scenario (noBB, base, or 3×BB). The persistent positive bias in the noBB simulation suggests an overestimate of background MDA8 O₃. Biases further increase on smoke days, reaching up to ~70 ppb at Missoula and scaling with observed daytime PM_{2.5} in the base model ($r \approx 0.7$) (Fig. S12). Similar smoke-amplified positive ozone biases have been reported in other CTMs (e.g., CMAQ-based simulations; Fig. 6A) and in prior



GEOS-Chem work (Zhang et al., 2014), suggesting that regional-to-global CTMs may systematically overestimate background O₃ and/or net O₃ production under BB influence.

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Figure 6B shows that GEOS-Chem does not reproduce the observed non-monotonic O₃–PM_{2.5} relationship. At Missoula, the model captures the initial O₃ increase when PM_{2.5} is below ~30–40 µg m⁻³, but it predicts increasing O₃ at higher PM_{2.5}, whereas observations flatten or decline beyond this threshold. A similar failure is evident in AIRPACT, a publicly available regional air quality forecasting system (Chen et al., 2008; Vaughan et al., 2004). Across Missoula and other western U.S. sites (e.g., Eugene, OR; Yreka, CA; Cheeka Peak, WA; Fig. S6), observations show strong O₃ suppression under heavy smoke, while AIRPACT predicts a monotonic increase of O₃ with PM_{2.5}. Together, these consistent biases across two independent CTM frameworks point to a shared model limitation in representing the nonlinear ozone response under intense BB smoke.

Part of the positive model bias may reflect errors in meteorological processes under smoke conditions. First, GEOS-Chem captures the relative reduction in broadband shortwave radiation on smoke days, suggesting reduced photolysis (i.e., $J(\text{O}^1\text{D})$ and $J(\text{NO}_2)$), but it still overestimates the absolute downwelling shortwave flux by ~25% during smoke-impacted periods (versus ~20% on non-BB periods) (Fig. S5). These “over-sunny” conditions likely result in excessive radical production, although we lack observed J values for a direct photolysis evaluation. Second, model temperature is biased high by +1.13 °C on smoke days (implying ~20% faster PAN thermal loss at ~289 K) and biased low by –1.5 °C during non-BB periods, which could favor enhanced NO_x recycling and sustained in situ O₃ formation. Third, PBLH tracks the temporal variability of assimilated products (e.g., HRRR, 3 km) but is higher by ~500 m (33%) during smoke and slightly lower (–20 m, –1%) during non-BB periods. The elevated PBLH can increase entrainment of O₃-rich air from aloft and strengthen the influence of transported (upwind) background O₃. In addition, heterogeneous NO₃/N₂O₅ loss in BB plumes may be too weak (Decker et al., 2019, 2021), which could prolong NO_x lifetime and contribute to overproduction of O₃ under smoke (Shen et al., 2025). We cannot attribute the discrepancy to a single process, but collectively these factors provide a plausible explanation for the spurious modeled MDA8 O₃ increases during heavy smoke and the growth of the O₃ error with PM_{2.5}.

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8.3 Model uncertainties in health-risk estimates

GEOS-Chem captures the order of magnitude of the smoke-attributable excess lifetime cancer risk but still underestimates it by ~40% (63 vs 100 per million), primarily due to its low bias in PM_{2.5}. Because cancer risk is dominated by PM_{2.5} whereas HI is driven by a small number of high-potency HAPs, model errors in HAP composition have a much larger impact on HI than on cancer risk. Accordingly, the base simulation substantially underestimates chronic non-cancer risk (HI ≈ 0.3, ~10× lower than observed). For acute risk, GEOS-Chem predicts no 1-h HI exceedances (max 1-h HI = 0.2) and only limited 8-h exceedances (15 hours with HI > 1; max 8-h HI = 1.2), far fewer and weaker than observed (476 hours; max 8-h HI = 6.3).

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In the 3×BB sensitivity run, the modeled cancer risk increases to 155 per million (exceeding the observation-based estimate), highlighting the uncertainty in modeled PM_{2.5} under smoke conditions. Acute non-cancer risk increases in the 3×BB run (135 hours with 8-h HI > 1; max 8-h HI = 3.4), but the model still predicts no 1-h exceedances (max 1-h HI = 0.5). This low bias in HI is driven by incomplete BB VOC representation, including underestimated BB VOC burdens (by ~3×), missing secondary sources, and missing high-potency HAPs (e.g., acrolein, which accounts for >50% of the observed HI). Overall, CTMs such as GEOS-Chem can provide a first-order estimate of PM_{2.5}-driven cancer burden, but the predicted HI is much less reliable without improved HAP speciation, secondary production, and inclusion of high-potency compounds.

9 Conclusion

Missoula experienced persistent and chemically complex wildfire smoke during September 2020, reflecting the widespread influence of wildfires across California and the Pacific Northwest. We report hourly surface observations of PM_{2.5}, CO, NO_x, O₃, and 75 speciated VOCs in Missoula, Montana (46.9 °N, 114.0 °W), during the wildfire-smoke-impacted month of September 2020. Of the 75 measured VOCs, 15 were classified as HAPs by the EPA. Leveraging comprehensive measurements, we quantified EnRs of PM_{2.5} and VOCs relative to CO in smoke aged for several days, and characterized the temporal evolution of criteria pollutants and VOCs under smoke-impacted and no-/low-smoke conditions. We performed nested GEOS-Chem simulations and compared them with observations to constrain BB emissions and photochemistry in the model. Finally, we assessed public health risks based on regulatory exposure metrics for PM_{2.5} and HAPs.

We find that combining gas-phase tracers with conventional particle-based diagnostics improves the identification of surface-level wildfire smoke in urban environments. While traditional aerosol-based smoke diagnostics (HMS, AERONET, and PM_{2.5}) remain useful for detecting dense, optically thick plumes, they often misclassify events when plumes are aloft, optically thin, or when rainfall removes aerosols. In contrast, VOC tracers, especially furan and maleic anhydride (MA), provide complementary value: furan identifies fresh smoke, while MA captures more aged plumes. We thus develop a balanced multi-indicator rule that requires concurrent enhancements in at least three independent tracers, thereby capturing smoke episodes with higher confidence. Three major smoke events driven by long-range transport resulted in multi-day elevations of CO and total VOC abundances by factors of 2–3, while ozone increased only modestly and NO_x showed little to no change. In addition, PM_{2.5} and total HAPs (which comprised ~half of the measured TVOCs by molar fraction) increased by factors of ~3–8 during smoke periods. Concurrently, the total OH reactivity doubled (14.2 vs 7.1 s⁻¹), shifting the dominant OHR contributor from biogenic tracers in background conditions to CO and BB VOCs during smoke. These results highlight the strong influence of regional wildfire smoke on air quality in the western U.S.



Photochemical-clock analysis constrains ERs for a wide range of primary VOCs in aged BB smoke, yielding values consistent with those reported for western U.S. wildfires. The observed decay patterns of primary VOCs broadly follow their known OH rate constants (k_{OH}). Some underestimation of k_{OH} (by factors of ≈ 3) for reactive VOCs likely arises from rapid depletion, dilution, or background contamination. OVOCs such as methanol, formaldehyde, and acetaldehyde also decrease with age, but their shallow slopes indicate substantial secondary production offsetting primary decay.

Intense smoke episodes in Missoula suppressed surface ozone, with MDA8 O_3 declining by up to ~ 15 ppb (6–7 September), coincident with 50 % reductions in solar radiation and lower planetary boundary layer heights (PBLH), confirming that reduced photolysis and weaker mixing limit in-situ ozone formation. Across smoke-impacted days, the MDA8 O_3 – $PM_{2.5}$ relationship was non-monotonic: ozone increased under light smoke ($PM_{2.5} < \sim 20 \mu g m^{-3}$) but tended to flatten or decrease at higher smoke loadings ($> \sim 20 \mu g m^{-3}$).

Chronic and acute health assessments indicate that prolonged exposure to 2020-level wildfire smoke in Missoula poses quantifiable health risks. If a season of similar intensity were to recur annually, the smoke-attributable excess lifetime cancer risk would be ~ 100 cases per million people ($\approx 7\times$ the non-smoke baseline of ~ 15 per million for the same days), with $PM_{2.5}$ accounting for ~ 90 % of the cancer burden. The chronic non-cancer hazard index (HI) is ~ 3 , with $\sim 90\%$ attributable to HAPs (driven primarily by acrolein and formaldehyde) and the remaining $\sim 10\%$ to $PM_{2.5}$. For acute non-cancer risk, 1-hour HI exceedances are rare, but 8-hour averaging reveals frequent smoke-aligned exceedances ($\sim 51\%$ of periods with $HI > 1$). Across both chronic and acute metrics, the repeated importance of the same few HAP drivers (notably acrolein, formaldehyde, and benzene) indicates that mitigating wildfire-related health impacts should extend beyond particle-only strategies to include gas-phase toxics.

The GEOS-Chem simulation reproduces the timing and transport of major smoke plumes ($R^2 = 0.8$ – 0.9) but underestimates the magnitude of smoke enhancements, with model low biases of ~ 30 – 40% for CO and $PM_{2.5}$ and ~ 60 – 90% for most VOCs. The model overestimates OH exposure by roughly 100 %, leading to excessive chemical loss and artificially low EnRs for reactive VOCs. Persistent low EnRs for oxygenated VOCs further suggest missing secondary production during multi-day aging, whereas the +33% bias in $PM_{2.5}$ EnR points to overly efficient secondary aerosol formation and/or retention in the GEOS-Chem simplified “simple SOA” treatment. Tripling BB emissions ($3\times$ BB) largely improves the modelled CO and primary VOCs (e.g., the model CO bias is changed from -40% to $+20\%$; benzene from -70% to -10%) and reduces the model bias of OH exposure to ~ 30 %, but the modeled $PM_{2.5}$ is worse (from -30% to $+80\%$), demonstrating that uniform emission scaling cannot reconcile errors in both gases and aerosol. Together, these results highlight that reducing model biases in smoky environments requires not only better fire emission magnitudes but also improved representation of missing OH reactivity (e.g., furanoids), secondary OVOC formation, and smoke SOA parameterizations.



GEOS-Chem exhibits a persistent positive bias in September MDA8 O₃ in Missoula (+15 ppb on average) and fails to reproduce observed day-to-day variability, regardless of the BB emissions scenario (base, 3×BB, or noBB). This is in part due to the modeled O₃ being too high for urban general background, and the model bias further amplifies on smoke days, reaching +70 ppb and increasing with observed daytime PM_{2.5} ($r \approx 0.7$). More importantly, the model fails to reproduce the observed non-monotonic O₃–PM_{2.5} response, where observations flatten or decline beyond ~30–40 µg m⁻³. Instead, GEOS-Chem continues to increase O₃ with PM_{2.5}. The same monotonic behavior is also present in the independent CMAQ-based AIRPACT forecast system across Missoula and other western U.S. sites, pointing to a shared CTM limitation in representing nonlinear ozone responses under intense BB smoke. These model biases are likely related to overestimated radiation, warm temperature biases, PBLH/entrainment errors, and potentially too-weak heterogeneous NO₃/N₂O₅ loss in CTMs, although isolating the dominant drivers will require additional observational constraints.

GEOS-Chem underpredicts the smoke-attributable lifetime excess cancer risk by 40% (63 vs 100 cases per million), due to the underestimated PM_{2.5}. The model's capability to predict non-cancer HI is even worse (~0.3 in GEOS-Chem vs 3 in observations). The model also underrepresents acute hazards, predicting limited 8-h exceedances (15 h with 8-h HI > 1; max 8-h HI = 1.2), far fewer and weaker than observed (476 h; max 8-h HI = 6). Overall, these results indicate that GEOS-Chem can provide a defensible screening-level estimate of PM_{2.5}-dominated cancer burden, whereas HI estimates remain unreliable without improved BB VOC speciation, secondary production pathways, and inclusion of high-potency toxics such as acrolein.

Code and data availability

The data and analysis code used in this study will be made publicly available upon final publication. During the review process, the full analysis code and processed datasets have been made available to reviewers through the Copernicus review system with access restricted to reviewers only. The archived materials allow full reproduction of all figures and results presented in this paper. Raw air-quality observations were obtained from the Montana Department of Environmental Quality and are subject to the agency's data-use policies. Meteorological and ancillary datasets were obtained from publicly available sources cited in the manuscript.

Author contributions

LJ: investigation; conceptualization; methodology; simulations; formal analysis; visualization; writing (original draft); data curation. LT: investigation; data curation. DK: investigation; data curation. KN: investigation; data curation. VS: investigation; writing (review and editing). BY: supervision; conceptualization; writing (review and editing). LH:



supervision; conceptualization; writing (review and editing); funding acquisition. All authors reviewed and approved the final manuscript.

Disclaimer

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The review statement will be added by Copernicus Publications listing the handling editor as well as all contributing referees according to their status anonymous or identified.



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