

## Article

# Impacts on Urban VOCs and PM<sub>2.5</sub> during a Wildfire Episode

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**Abstract:** This study focuses on the impact of wildfire smoke emissions on regional, urban air quality during a wildfire event. We measured volatile organic compounds (VOCs) and fine particulate matter (PM<sub>2.5</sub>) in the San Francisco Bay Area to assess air quality during a wildfire event and compared them to those in a later non-wildfire period. VOCs were collected using thermal desorption tubes and quantified using thermal desorption-gas chromatography/mass spectrometry (TD-GC/MS). Elevated concentrations of VOCs such as 1,3,5-trimethylbenzene ( $0.33 \pm 0.01 \mu\text{g}/\text{m}^3$ ), benzene ( $1.03 \pm 0.02 \mu\text{g}/\text{m}^3$ ), toluene ( $2.15 \pm 0.04 \mu\text{g}/\text{m}^3$ ), ethylbenzene ( $0.60 \pm 0.02 \mu\text{g}/\text{m}^3$ ), and m, p-xylene ( $0.77 \pm 0.07 \mu\text{g}/\text{m}^3$ ) were observed in the wildfire event. Compared with that in the non-wildfire season, the toluene concentration during the wildfire period was more than three times the non-wildfire concentration. Similarly, the benzene concentration during the wildfire was almost four times higher, and that of p, m-xylene was three times higher. The higher wildfire levels were statistically significant for sec-butylbenzene, 1,2,4-trimethylbenzene, n-propylbenzene, o-xylene, styrene, 1,3,5-trimethylbenzene, benzene, toluene, ethylbenzene, and p,m-xylene ( $p < 0.00001$ ). These higher VOC levels compared with those for the non-fire period may potentially pose a public health concern. Open face passive sampler (OFPS)-collected PM was analyzed using scanning electron microscopy/energy dispersive X-ray spectroscopy (SEM/EDS) and revealed organic carbon tar balls. The highest daily average beta-attenuation-monitored PM<sub>2.5</sub> during the fire period was  $200 \mu\text{g}/\text{m}^3$  and the highest hourly average was  $270 \mu\text{g}/\text{m}^3$ . Monitoring gas phase species in addition to PM concentrations is useful during wildfire season to inform public health guidance.



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**Keywords:** wildfire; climate change; urban air pollution; air quality; VOCs; tar ball; c/o ratio; combustion aerosol; BTEX

## 1. Introduction

Interest in wildfire smoke and its health effects has sharply increased in recent years [1,2]. Estimates have suggested that anthropogenic climate change caused the cumulative geographic land area impacted by forest fires to double between 1970 and 2016 [3]. The state of California has seen impacts of a warmer, drier climate in the form of yearly fire seasons. The worst single incident of the 2018 fire season was the Camp Fire in Butte County, California [4], which burned 620.53 km<sup>2</sup> and resulted in 5 firefighters being injured, 89 civilian deaths, 12 civilians being injured, 18,804 structures being destroyed, and 564 structures being damaged [5]. The increased frequency and size of such wildfires represents a notable source of airborne particulate matter (PM) and volatile organic compounds (VOCs) [6,7]. The United States Environmental Protection Agency (US EPA) has named wildfires as one of the important climate change indicators [8]. Wildfire smoke currently accounts for 40% of the total PM emitted in the US [9].

The World Health Organization (WHO) estimated that at least 250,000 deaths will occur every year due to wildfires and other aspects of climate change [10]. The US EPA and the United States Center for Disease Control and Prevention (US CDC) have indicated that the smoke from wildfires can cause respiratory tract irritation and more serious disorders,

including reduced lung function [11], bronchitis, an exacerbation of asthma and heart failure, and premature death [12,13]. Children, pregnant women, and the elderly are especially vulnerable to smoke exposure [14,15]. When a wildfire impacts residential or industrial areas, burning building materials such as painted furniture, polymer flooring, electronic products, and vehicles can release large amounts of harmful VOCs, carbon monoxide (CO), polycyclic aromatic hydrocarbons (PAHs) [16,17], nitrogen oxides (NO<sub>x</sub>), and PM, all of which could have acute and/or chronic human health implications [9,15,18,19].

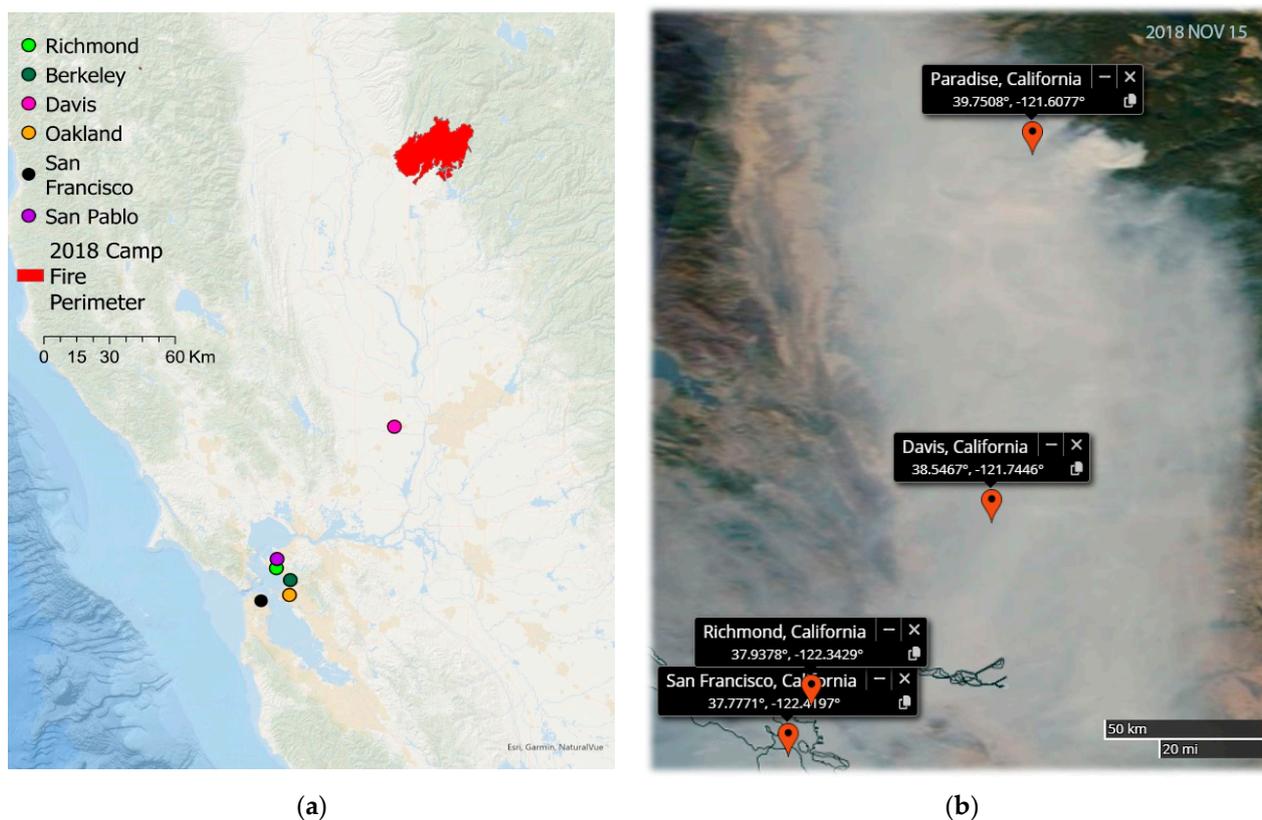
Fine PM<sub>2.5</sub> in general has great potential to impact human health due to its ability to reach the thoracic and alveolar regions of the respiratory system [20,21]. The Global Burden of Disease Comparative Risk Assessment conducted by WHO reports that in 2005, PM<sub>2.5</sub> was globally responsible for 3% of cardiopulmonary disease mortality. These figures amount to 0.8 million premature deaths and 6.4 million years of life lost (YLL) worldwide [22].

The black carbon (BC) or soot component of wildfire PM<sub>2.5</sub> is an important contributor to climate change [23,24]. Studies have shown that the increased soot and organic carbon emitted from wildfires could lead to further climate change [25]. Previous studies have reported measurements of BC and UV-absorbing organic carbon PM (UVPM) in the San Francisco Bay Area for different scenarios [26–28]. To control wildfire smoke exposure more effectively and better protect public health, a further characterization of wildfire smoke is necessary [6,12,27].

Wildfires can also generate toxic VOCs and PAHs from burning plants, furniture, plastics, and building materials in the fire area [29–32]. The VOCs released from wildfires are associated with serious health effects [33–35], and they can participate in complex photochemical reactions and create harmful ultrafine aerosols [36,37]. Many VOCs are mutagenic and/or carcinogenic themselves, presenting a long-term risk of cancer to those exposed. Acute exposure to high concentrations of VOCs causes sensory irritation, especially to the eyes, skin, and respiratory system.

Depending on fire size, location, and weather conditions, PM and VOCs may potentially be transported across long distances from the burning area to more densely populated areas. The 2018 Camp Fire led to severe air quality deterioration throughout the region. Residents of the San Francisco Bay Area 240 km away experienced strong odors, high PM concentrations, and poor visibility until the fire was fully contained [4]. After the initial transport of smoke to the Bay Area, low winds and a stable inversion resulted in a stagnant system that trapped smoke there for weeks [38]. Figure 1a shows the Camp Fire location and the air monitoring site locations in this study. Figure 1b is a NASA satellite image from 15 November 2018 that shows smoke during the Camp Fire period extending south down along Sacramento Valley and then west to the Bay Area.

The objective of the current study is to characterize a range of particle and gas-phase species before, during, and after this fire event to better understand the impacts of the wildfire smoke emissions on the regional air quality in an urban area distant from the fires.



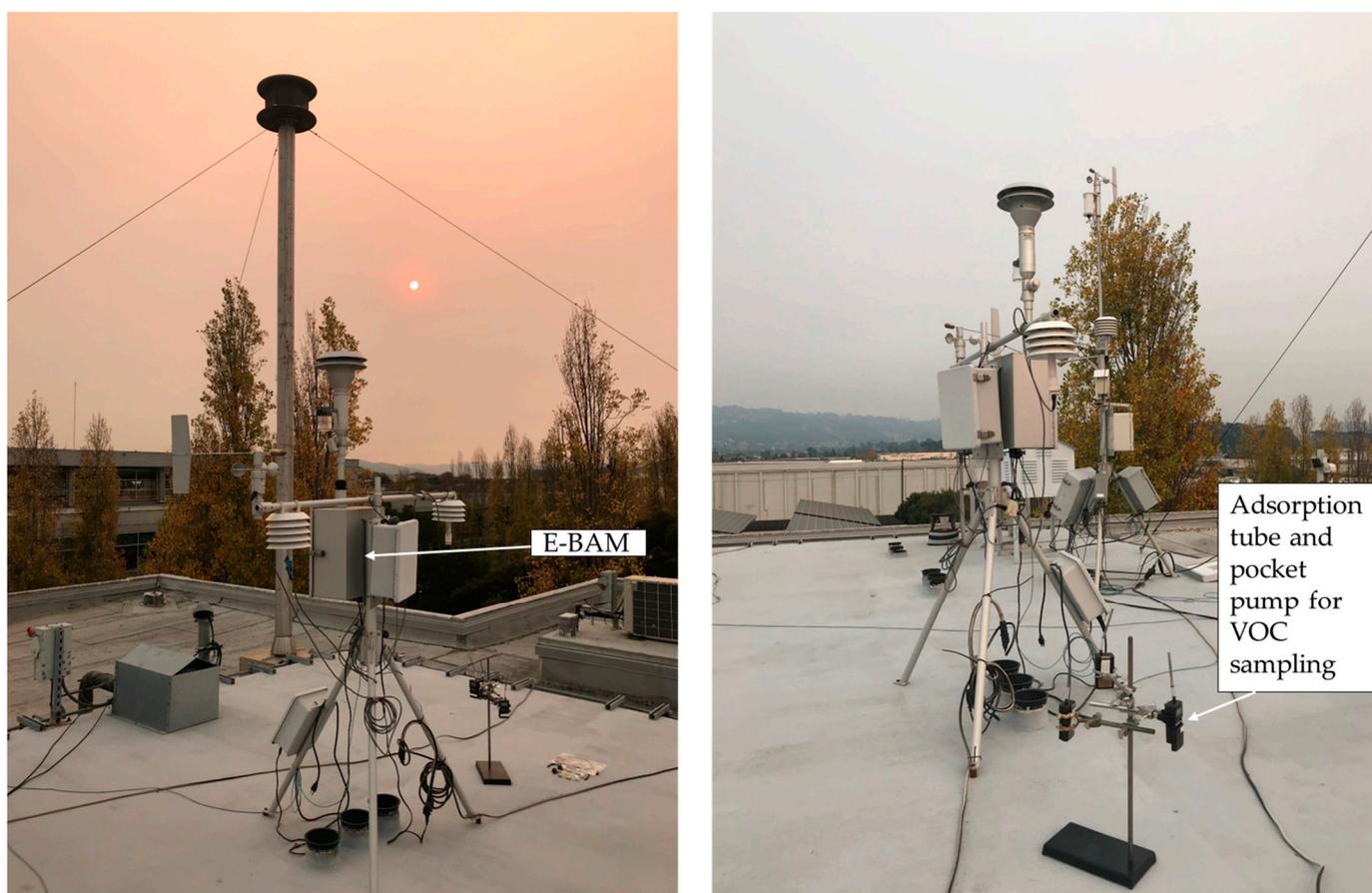
**Figure 1.** (a) 2018 Camp Fire perimeter and locations of samples acquired in Richmond, Berkeley, Oakland, and San Pablo. The map was generated using ArcGIS Pro (ESRI, Redlands, CA, USA) and wildfire perimeter data [39]. (b) NASA satellite image for Camp Fire smoke [40].

## 2. Materials and Methodology

### 2.1. VOC Assessment

#### 2.1.1. TD-GC/MS Analysis

VOCs were collected onto stainless steel Tenax Sorbent Tubes measuring 3<sup>1</sup>/<sub>2</sub>-inch long x 1<sup>1</sup>/<sub>4</sub>-inch o.d. (C1-AAXX-5003, Markes International Inc., Llantrisant, UK) using small, battery-powered pumps (Model 210-1002C, SKC Inc., Eighty Four, PA USA) at 100 mL/min for 2 h (total sampled air volume = 12 L per tube). All the sampling tubes, including field blanks, were cleaned before air sampling using a heated TC-20 tube conditioning system (Markes Inc., Llantrisant, UK) with high-purity nitrogen flow using the manufacturer's recommended temperature and time. Any contaminants or interferences were required to be ≤0.2 parts-per-billion-volume before use. Air samples were collected on the rooftop (12 m above ground) of the California Department of Public Health (CDPH) laboratory in Richmond, California from 2:45–4:50 PM on 20 November 2018 during the wildfire period (8–25 November 2018) (Figure 2). Non-wildfire samples were taken on 12 December 2018, which was 17 days after the wildfire episode ended. For each sampling period, three pumps with tubes were co-located to calculate standard deviations. The sampling pumps were calibrated using mini-BUCK Calibrator M-5 (A.P. Buck Inc., Orlando, FL, USA) before and after sampling. Flow rates (103.4 ± 3.1, 106.3 ± 6, 106.1 ± 3.1, mL/min for the three sampling pumps) were averaged for calculations. Field blank samples were also collected by taking sealed tubes to the sampling site but not pumping any air through them. After sample collection, the Tenax tubes were tightly sealed and stored in a closed, emission-free container at 4 °C in a refrigerator until analysis, which was conducted within one week of sampling.



**Figure 2.** The E-Bam set up for PM<sub>2.5</sub> (left), and the adsorption tube set up for VOCs (right).

VOCs were analyzed using a thermal desorption-gas chromatography/mass spectrometry (TD-GC/MS) method adopted from EPA TO-17 [41]. The tubes were directly analyzed using TD-100 (Markes International Inc., Llantrisant, UK) coupled with a GC 7890/MS 5977 (Agilent Technologies Inc., Santa Clara, CA, USA) for VOC analysis. Briefly, VOCs trapped in the Tenax tubes were transferred to TD-100 at 300 °C for 10 min under high-purity helium (5N grade; no inlet split flow). The compounds were re-concentrated on a cold/sorbent trap (U-T 15 ATA-2S, cold trap—Air Toxic Analyzer from Markes International Inc.) at 23 °C. The cold trap was rapidly heated to 300 °C over 3 min while passing a helium flow for VOC transfer to an uncoated and deactivated fused-silica capillary transfer line (the internal and outer diameters of 0.25 and 0.35 mm, respectively) heated at 150 °C. The helium gas (with a 10 mL/min split flow) delivered the volatiles to the GC/MS system for analysis. The GC was equipped with an Agilent J&W VF-5ms capillary column (length: 30 m; I.D.: 0.25 mm; Film thickness: 1.0 μm). Helium was the carrier gas at a flow of 1.2 mL/min (constant flow mode). The GC oven temperature program started at 32 °C (holding time 5 min), then increased to 150 °C at 7 °C/min and to 280 °C at 30 °C/min, and held for 5 min. The GC/MS transfer line was heated to 230 °C. The MS source and quadrupole temperatures were 230 °C and 150 °C, respectively. The detector was in full-scan mode between 30 and 450 amu with electron impact ionization at 70 eV. The 76 VOC compounds listed in EPA Method 8260B [42] were selected and quantified by comparing them with standard chemicals. The calibration curve for each chemical compound was created by spiking 7 different levels with 10, 20, 50, 100, 200, 300, and 500 ng of pure standards onto the clean tubes. Three gas-phase chemical compounds, bromofluorobenzene, chlorobenzene-d<sub>5</sub>, 1,4-difluorobenzene, were automatically injected into each adsorbent tube through the TD-100 as internal standards. The method detection limit (MDL) for this assay was 0.42–4.68 ng/tube (corresponding to 0.04–0.39 μg/m<sup>3</sup>) for the 75 analytes, except

that for 1,1,2-Trichlorotrifluoroethane (CFC-113) was 10.22 ng/tube, or 0.85  $\mu\text{g}/\text{m}^3$ . The limit of quantitation was 10 ng/tube (or 0.83  $\mu\text{g}/\text{m}^3$ ) for all analytes. Table 1 gives the method detection limit for the major chemicals detected in this study.

**Table 1.** Method detection limits (MDL) for major gas-phase chemicals.

Compound	Name	Retention Time (RT), min	MDL, $\mu\text{g}/\text{m}^3$
1	carbon disulfide	3.948	0.39
2	methylacrylonitrile	5.726	0.08
3	benzene	8.113	0.10
4	toluene	11.975	0.12
5	ethylbenzene	15.017	0.08
6	m-xylene	15.28	0.05
7	p-xylene	15.28	0.18
8	styrene	15.951	0.07
9	o-xylene	15.986	0.11
10	cis-1,4-dichloro-2-butene	16.921	0.08
11	benzene, propyl-	17.698	0.17
12	benzene, 1,3,5-trimethyl-	18.097	0.08
13	1,2,4-trimethylbenzene	18.804	0.08
14	sec-butylbenzene	19.203	0.05

### 2.1.2. VOC Data Analysis and Processing

Target VOC compound identifications and amounts were determined using MSD ChemStation Data Analysis software (Agilent Technologies Inc., Santa Clara, CA, USA) based on the retention time (RT), mass spectrum, and selected ions (one quantifier ion and two additional qualifier ions for each compound). VOC concentrations were reported as the mean value of the triplicate air sample measurement results, along with their standard deviations. The quantified VOC results during the wildfire event were compared with those after the Camp Fire period. An integrated NIST 11 mass spectral database in MassHunter qualitative analysis from Agilent was used for the identification of other unknown peaks. The tentatively identified chemical compounds with a NIST mass library match score  $\geq 80$  are reported without further confirmation using chemical standards.

## 2.2. PM Assessment

### 2.2.1. PM<sub>2.5</sub> Continuous Measurement during the Fire Season

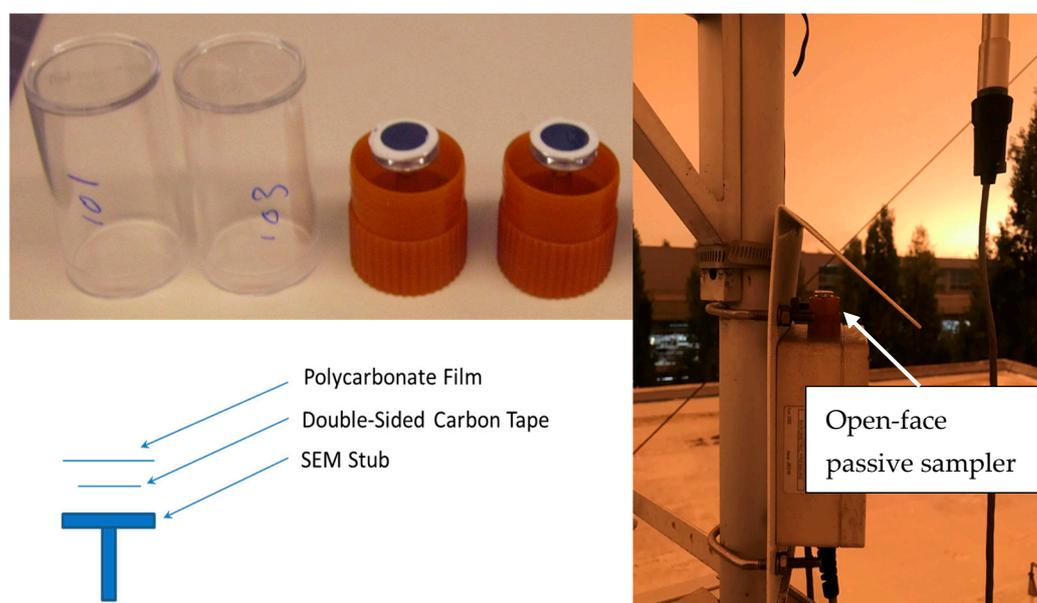
PM<sub>2.5</sub> levels ( $\mu\text{g}/\text{m}^3$ ) were measured during the Camp Fire between 10 and 30 November 2018 using a beta-attenuation monitor (E-BAM, Met One Instruments Inc., Grants Pass, OR, USA) on the CDPH rooftop, as shown in Figure 2. Met-One E-BAM is a non-regulatory method that is useful for fast deployment and for measuring temporal trends. The unit was calibrated and serviced by the manufacturer before the fire episode. The flow rate was also calibrated onsite using a lab-calibrated rotary flow meter. The measured time resolution was 10 min, with the data exported as hourly averages.

The measured November 2018 wildfire data were compared to measurements made at the same sampling site during November 2015, when there were no fires. To compare our E-BAM PM<sub>2.5</sub> values to other regional data, PM<sub>2.5</sub> data were obtained from Bay Area Air Quality Management District (BAAQMD). Data from two beta-attenuation (BAM-1020, Met One Instruments Inc. a federal equivalent method (FEM) instrument) monitoring stations (San Pablo (5.6 km north of Richmond) and Berkeley (9.5 km southeast)) were downloaded and compared with the CDPH E-BAM values.

Black carbon data were also obtained from BAAQMD, which were measured at a community site in Richmond, CA, using an aethalometer (Magee Scientific AE33, Berkeley, CA, USA).

### 2.2.2. Passive PM Sampling and SEM/EDS Particle Analysis

During the wildfire episode, open-face passive samplers (OFPS) [43] were placed out on the CDPH roof (Figure 3), and in a residential backyard in Albany, CA (7.2 km south of the Richmond campus, 2 m in height). Samplers with copper (Ted Pella, prod # 16072) and carbon substrates (Ted Pella, Product # 16084 and GE polycarbonate 0.01-micron filter on the top) were deployed side by side at the same time; copper tape was used for the optimal detection of the carbon content in the aerosols during the wildfire season. The samplers were analyzed using scanning electron microscopy/energy dispersive X-ray spectroscopy (SEM/EDS) for morphology and elemental chemistry. A Tescan MIRA3 field emission scanning electron microscope (FESEM) in high-vacuum mode with a BSE detector was used for imaging, and elemental chemistry was analyzed with a Bruker Quantax energy-dispersive x-ray spectroscope.



**Figure 3.** Open-face passive sampler with polycarbonate or copper substrate.

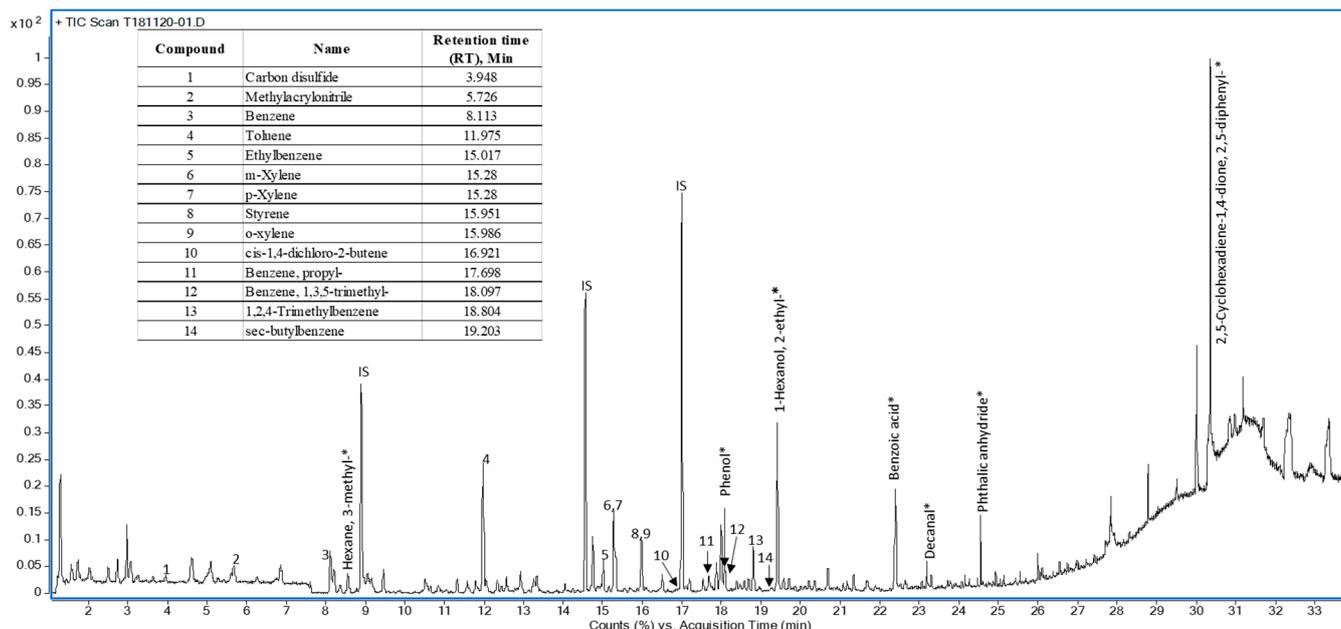
## 3. Results and Discussion

### 3.1. VOCs Detected during the Wildfire Period

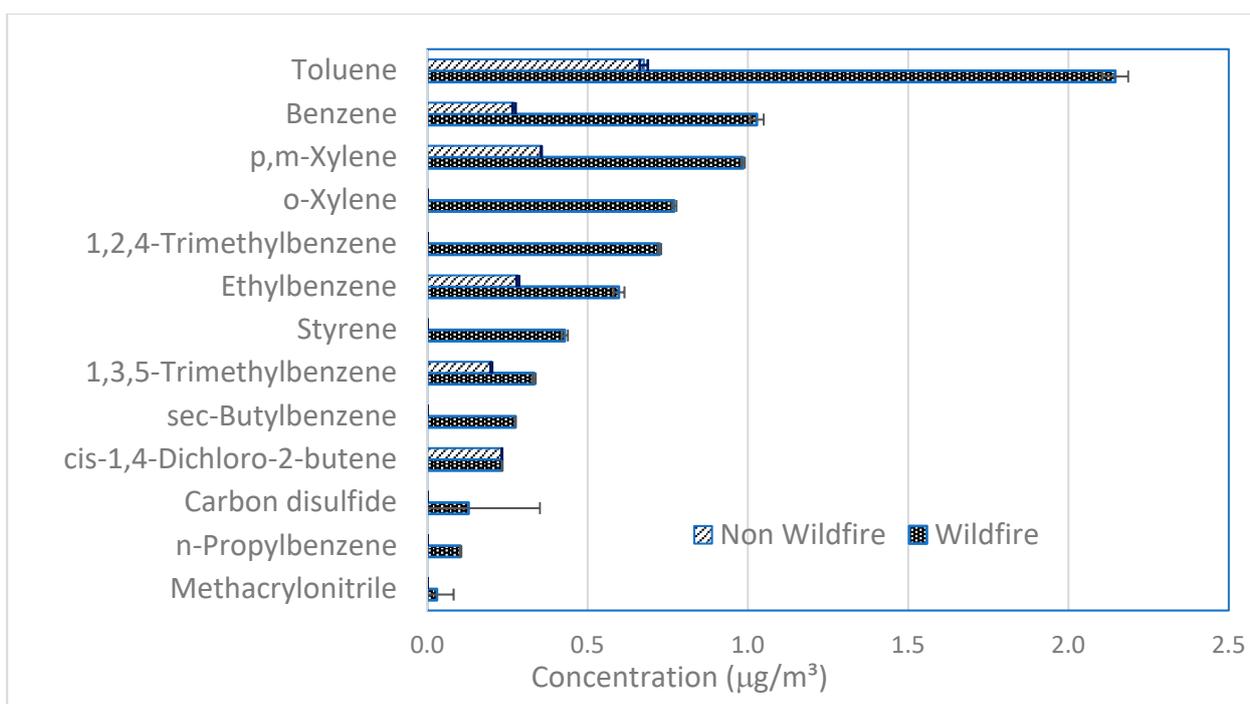
Figure 4 is an example of a GC/MS chromatogram from an air sample collected during the wildfire event showing the retention times for the major VOCs detected in our 76-target compound list. Compounds tentatively identified with a NIST library match score > 80 also can be seen in the figure and are marked by asterisks. However, many other visible peaks were not identified.

Figure 5 shows a comparison of the major quantified VOCs for samples collected on 20 November and 12 December. VOCs for which the concentration dropped to a lower concentration on 12 December were attributed to wildfires. Some VOCs that had similar concentrations on 12 December, such as cis-1,4-dichloro-2-butene, were likely created by a non-wildfire regional source such as local factory emissions, motor vehicle exhaust, solvent use, painting, or cooking [44]. Other VOCs such as sec-butylbenzene, 1,2,4-trimethylbenzene, n-propylbenzene, o-xylene, styrene, and methacrylonitrile were detected during the wildfire period, but not in the non-fire period. Additional VOCs, such as 1,3,5-trimethylbenzene, benzene, toluene, ethylbenzene, and p,m-xylene (BTEX) were detected in the non-fire period, but their concentrations were much higher during

the wildfire period. Notably, the toluene concentration during the wildfire period was more than three times the non-wildfire concentration. Similarly, the benzene concentration during the wildfire was almost four times higher, and that of p,m-xylene was three times higher. These higher wildfire levels were statistically significant for sec-butylbenzene, 1,2,4-trimethylbenzene, n-propylbenzene, o-xylene, styrene, and, 1,3,5-trimethylbenzene, benzene, toluene, ethylbenzene, and p,m-xylene ( $p < 0.00001$ ). Wildfire levels were not statistically higher for carbon disulfide, methacrylonitrile, or cis-1,4-dichloro-2-butene ( $p > 0.05$ ).



**Figure 4.** An example GC/MS chromatogram of the air sample collected during the wildfire event. (\*: not confirmed with standard chemical; NIST library match score > 80. IS: added internal standard.)



**Figure 5.** Mean concentrations of major detected VOCs compared between wildfire and non-wildfire periods. (Some standard deviation error bars are too small to be seen).

Dickinson et al. [31] reported increased concentrations of BTEX in ground-level wildfire smoke, with that of benzene ranging from 0.04 to 25 ppbv (0.13 to 79.87  $\mu\text{g}/\text{m}^3$ ). They calculated the ratio of benzene/toluene (B/T) to distinguish wildfire emissions from other emission sources ranging from 0.57 to 3 in different wildfire scenarios. In this work, B/T during the wildfire period was 0.56 ( $<1$ ). Various factors such as fire fuels, air sample aging, and weather conditions could affect this value.

Simms et al. [45] also measured BTEX during the 2018 Camp Fire in Davis, California, which is about 100 km northeast of Richmond. Their study reported 19 detected VOCs, while our study reported 13 quantified VOCs that did not include VOCs not in our method list or under our detection limits. The most abundant VOCs measured by Simms et al. were benzene and toluene, with average concentrations of 4.7 and 15.1  $\mu\text{g}/\text{m}^3$ , respectively, which are around 4.3 (4.7/1.0) and 6.9 (15.1/2.2) times the present work's benzene ( $1.03 \pm 0.02 \mu\text{g}/\text{m}^3$ ) and toluene ( $2.15 \pm 0.04 \mu\text{g}/\text{m}^3$ ) levels, respectively. In general, our major detected VOC compounds are similar to those of Simms et al., but have lower concentrations. This is not surprising since Davis is much closer to the fire center than Richmond, and toluene reacts with species like OH more quickly than benzene does. Besides the dilution effect over greater distances, the location difference and the different oxidation conditions may have also resulted in some differences in the final species. The oxidation of aromatics such as benzene, styrene, and toluene can form other oxygenated compounds, such as phenols, cresols, and benzaldehyde [31,32,45]. In this study, we detected phenol (Figure 4), but did not quantify it because it was not on our target compound list. Other possible explanations for the differences between these studies include differing sampling methods and temporal variability; for example, Simms et al. [45]'s toluene measurements varied from 4.1  $\mu\text{g}/\text{m}^3$  to 40.6  $\mu\text{g}/\text{m}^3$  on different sampling days.

During the same 2018 Camp Fire episode, Ivancic M. et al. [46] studied the aging of organic carbon (OC) and investigated how the characteristics of carbonaceous aerosols evolved over the course of the fire event at Berkeley (9.5 km southeast to our VOC monitoring site). They observed a higher fraction of secondary carbonaceous aerosols in the later phase of the fire, where smoldering and spot fires were more typical, a phase that coincides with our VOC sample collection period. This suggests the possible formation of oxygenated aromatics such as phenols, cresols, and benzaldehydes from their primary parent VOCs, benzene, toluene, and styrene, directly emitted from the wildfire [47]. However, due to the lack of VOC measurements closer to the fire, and the fact that most of the oxygenated aromatics were not quantified in this investigation, it is difficult to distinguish the originally emitted compounds from those formed via chemical evolution during the aging of the fire plume.

Compared to OEEHA's 8 h reference exposure levels [48], most of the VOCs measured in San Francisco Bay Area were relatively low. For example, the measured benzene concentration ( $1.03 \pm 0.02 \mu\text{g}/\text{m}^3$ ) is 3 times lower, that of 1,3,5-trimethylbenzene ( $0.33 \pm 0.01 \mu\text{g}/\text{m}^3$ ) is about 26 times lower, and that of toluene ( $2.15 \pm 0.04 \mu\text{g}/\text{m}^3$ ) is 400 times lower. Nevertheless, communicating any major increase in VOC concentrations could be of potential benefit to the public, especially for regions closer to a fire that could have much higher VOC concentrations. For example, the investigators in Davis closer to the Camp Fire [45] reported measured benzene ( $4.7 \mu\text{g}/\text{m}^3$ ) levels that were higher than OEEHA's 8 h exposure limit ( $3 \mu\text{g}/\text{m}^3$ ). In addition, because this wildfire episode was approximately three weeks long, people may have experienced much longer exposure times than 8 h, which could also have led to potentially different health concerns.

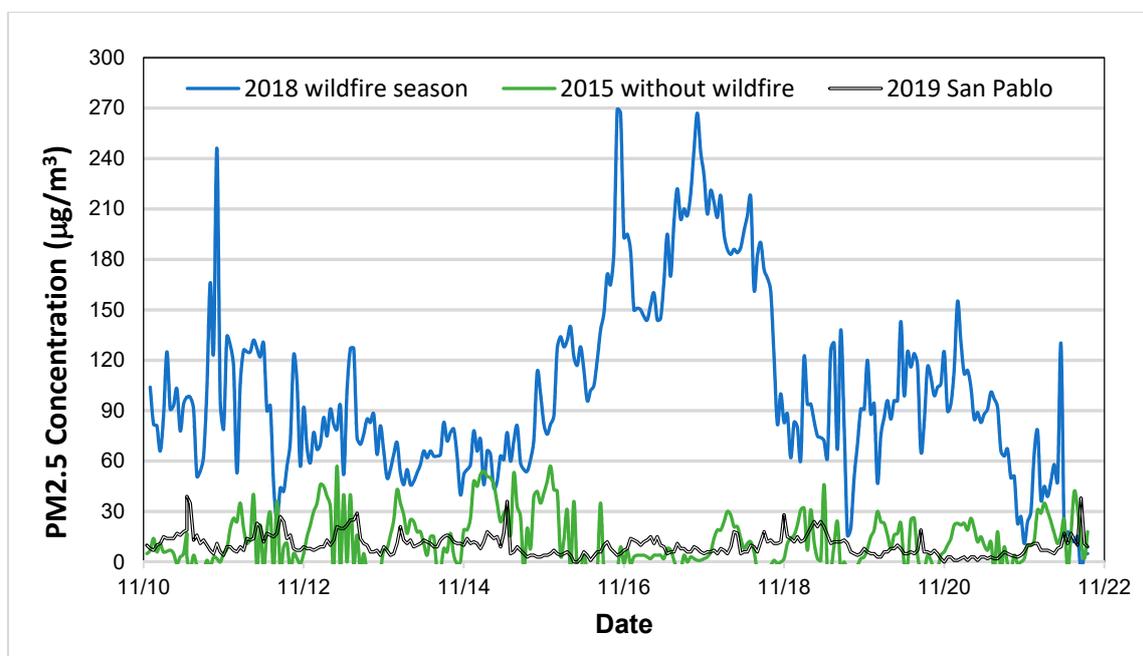
The greater abundance of VOCs in the wildfire samples compared with those in non-wildfire samples suggests potential gas-phase health risks associated with wildfires and the burning of natural and man-made materials. Particularly, methacrylonitrile is highly toxic and poisonous upon inhalation and can cause tearing and skin reactions externally [49]. Benzene is known to cause dizziness and confusion in the short term and can further lead to autoimmune disorders or cancer in the long term [30,31]. Finally, toluene, which was measured in the largest concentration during the wildfire, can cause dizziness and

headaches as well as permanent incoordination upon exposure [50]. BTEXs are well known hazardous air pollutants predominately through emissions from motor vehicles, aircrafts, manufacture processing, and tobacco smoking. Our results show that wildfires are another potentially important emission source of these chemicals. BTEXs can readily move through air, contaminated groundwater, and soils, thus affecting air quality and water resources. The US EPA maintains guidelines for the emission of these chemicals in drinking water and air [33]. Although this work represents a limited number of air samples for VOC analysis, our results suggest that future work should measure VOCs in air at multiple locations throughout a wildfire-impacted region. Note that the method used in this study was limited by the 76 compounds in our target list. Different wildfire fuel types can generate other VOCs [7,51], which were not detectable with this method. Future work will expand our methods and detection lists for the better characterization of wildfire-generated VOCs.

There are several limitations to this study. We only measured very limited numbers of gas-phase VOCs. Some important VOCs that are known to be emitted by wildfires, including methanol, acrolein, formaldehyde, acetaldehyde, acetic acid, hydrogen cyanide, phenols, and PAHs, were not quantified. In addition, although observations of VOCs emitted by wildfires are likely to be both spatially and temporally dependent due to chemical transformations, secondary oxidation products of parent VOCs were not distinguishable in this study. More gas-phase monitoring during wildfire events is needed to better understand the impact of wildfires on local and regional air quality.

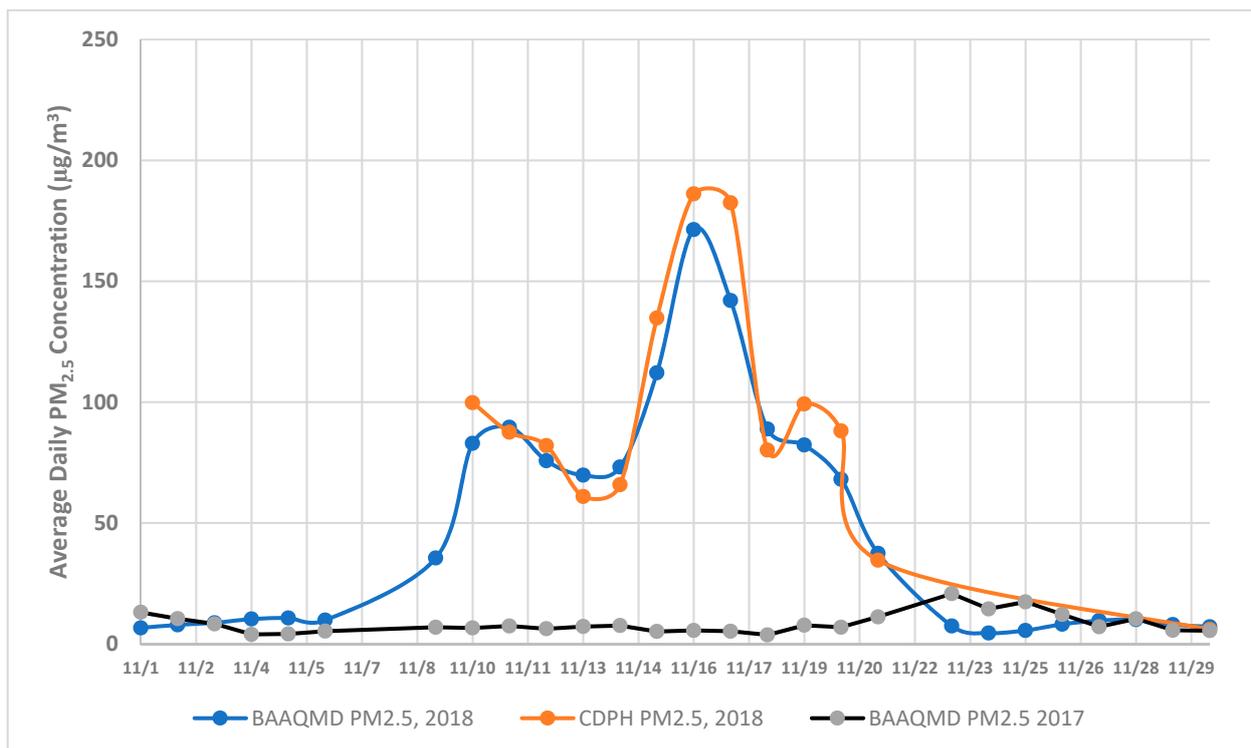
### 3.2. Comparing $PM_{2.5}$ Values during the Wildfire and Non-Fire Periods

E-BAM data from the fire period and data from a non-fire period at the same site in 2015 are shown in Figure 6. The wildfire-period measurements demonstrated much higher  $PM_{2.5}$ , with an hourly average peak value of approximately  $270 \mu\text{g}/\text{m}^3$ . The  $PM_{2.5}$  daily average during the fire period was  $101.6 \mu\text{g}/\text{m}^3$ , compared with the 2015 average  $PM_{2.5}$  of  $12.7 \mu\text{g}/\text{m}^3$ . The  $PM_{2.5}$  data acquired 25 days after the fire episode demonstrated a similar  $PM_{2.5}$  concentration to the 2015 non-fire concentrations ( $12 \mu\text{g}/\text{m}^3$ ).



**Figure 6.** Comparison of the Richmond hourly average  $PM_{2.5}$  ( $\mu\text{g}/\text{m}^3$ ) during the November 2018 fire period with that in a non-fire November in 2015 from the same site, and with that in the San Pablo site in November 2019.

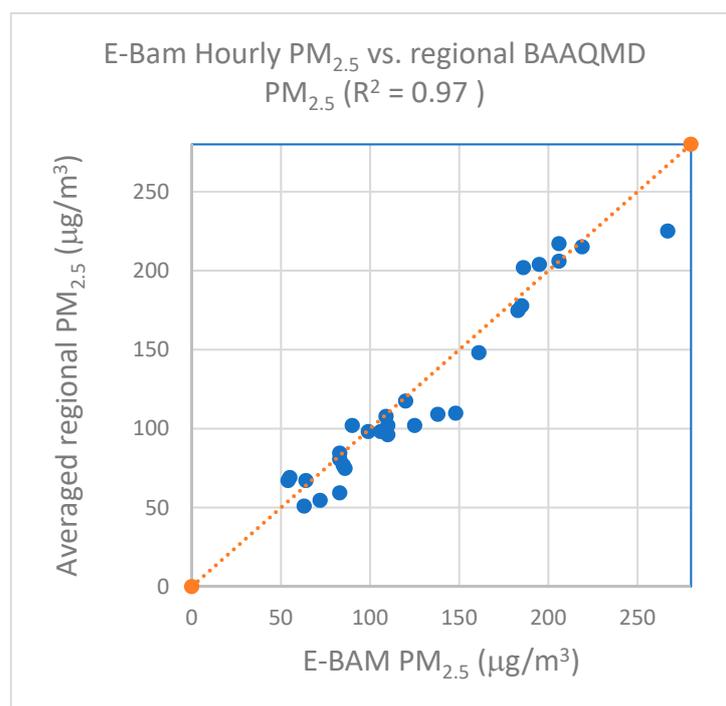
Figure 7 shows a PM<sub>2.5</sub> comparison between the CDPH E-BAM, a regional BAAQMD BAM (Berkeley) during the fire episode, and BAAQMD BAM (Berkeley) during a non-fire period for the same month in 2017. The E-BAM measurement demonstrated a similar trend to that of BAAQMD's measurement at Berkeley. The peak PM<sub>2.5</sub> concentrations during the fire period were almost 10-fold higher than the PM<sub>2.5</sub> concentrations in the same month of 2017. The highest daily peak value reached approximately 200 µg/m<sup>3</sup>. Poor air quality (PM<sub>2.5</sub> above 150 µg/m<sup>3</sup>) lasted for many days, as shown in Figures 6 and 7. The observation and measurement here were also consistent with early BlueSky modeling results [52].



**Figure 7.** Comparison of November PM<sub>2.5</sub> concentrations from the Richmond E-BAM with those from the BAM at BAAQMD Berkeley station during the 2018 Camp Fire, and with the BAAQMD Berkeley PM<sub>2.5</sub> from November 2017 with no fires.

These data are consistent with the heavy visible haze observed during the fire period within the Bay Area, as shown in Figure 2. That picture, taken on 12 November 2018, demonstrates near darkness during daylight hours. Other qualitative observations during this period include frequently smoky odors and visible, macroscopic ash.

Figure 8 shows that the CDPH-measured E-BAM PM<sub>2.5</sub> values had a high correlation ( $R^2 = 0.97$ ) with the PM<sub>2.5</sub> data reported for nearby regional BAAQMD (average of San Pablo, 5.4 km north, and Berkeley, 9.5 km south) stations. This suggests that the wildfire PM impacts in this region were relatively spatially homogeneous. This homogeneity may have been caused by the extended trapping of the smoke from the Camp Fire within an inversion layer over the San Francisco Bay Area [52,53].

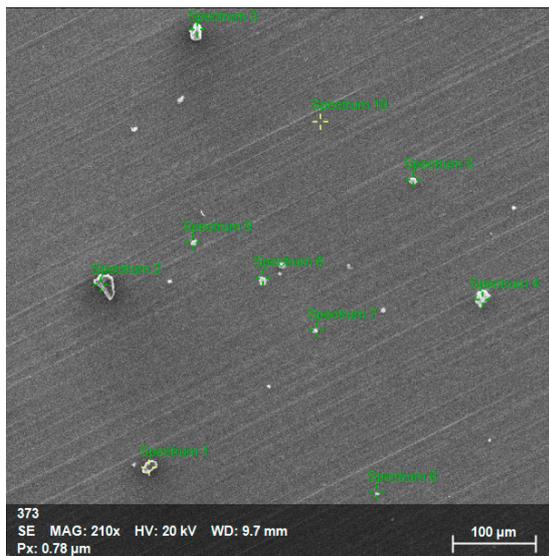


**Figure 8.** Comparison of E-BAM data with BAAQMD measurements. (The orange dot line is the diagonal line).

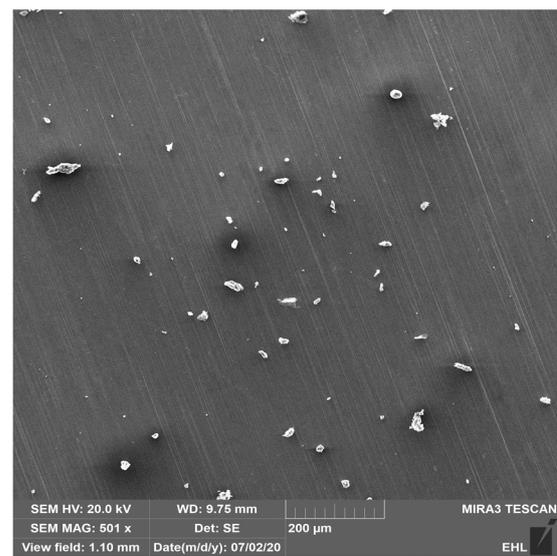
### 3.3. PM Morphology and Elemental Profiles

Carbonaceous PM and inorganic dust and ash collected using the OFPS were analyzed using SEM to examine particle morphology and elemental composition to help understand any unique types of particles collected during the wildfire period. Figure 9 shows typical low-magnification images of particles collected using the OFPS. As expected, a wide range of particle sizes were present, with visible particles or ash suspended in the air during the wildfire episode. The EDS analysis of the suspended largest particles (Figure 9a) showed that besides carbon, over 90% of these particles' compositions comprised inorganic components, such as aluminum, silicon, and iron, as shown in the spectrum and the normalized mass concentration table. Although these inorganic elements are consistent with ash, they also are common elements in road dust or PM from building construction and demolition.

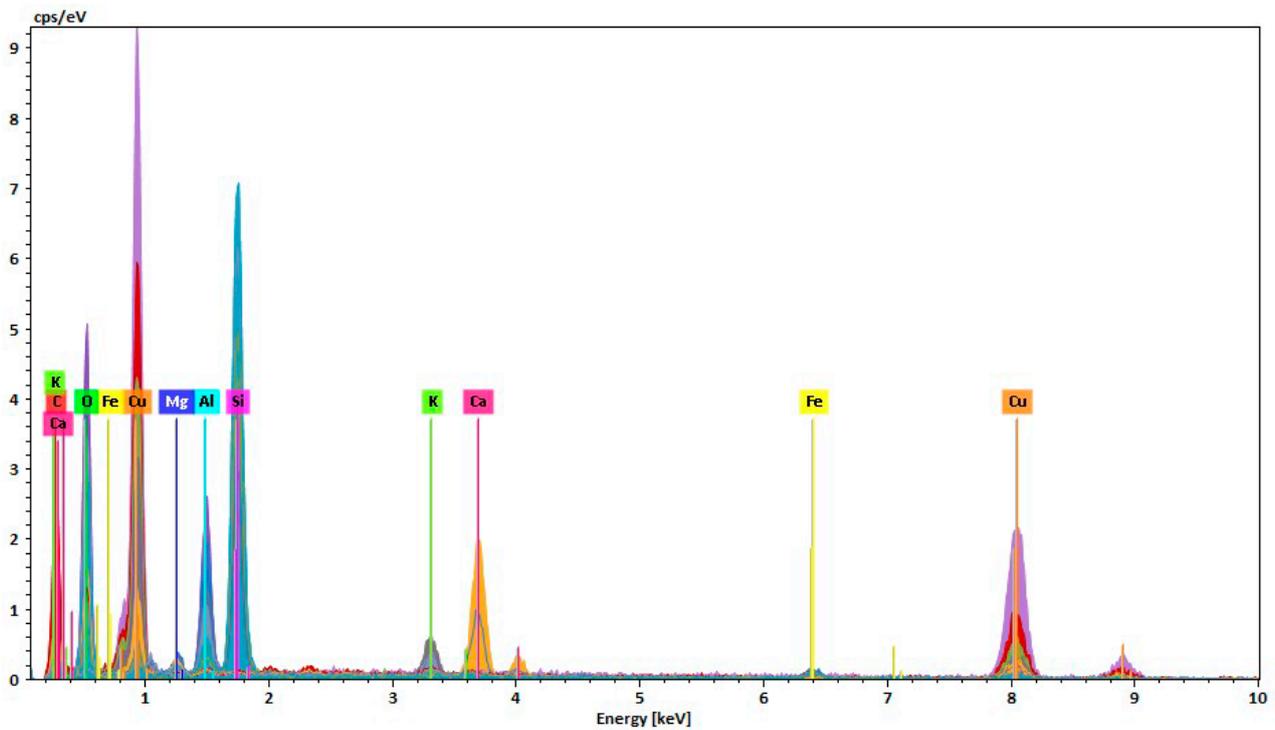
Examined under a higher magnification, the samples collected during the fire episode contained an abundant number of spherical particles consistent with organic carbon tar balls [27], as shown in Figure 10. Different sizes (250 nm–3 µm) of spherical particles and agglomerates are shown in Figure 10, and most of them had carbon as their major constituent. It is possible that some of the larger, super-micron spheres were biogenic rather than tar balls. Besides individual tar balls, the last two images in Figure 10 appear to be large agglomerates of tar balls, indicating that many different sizes of smoke particles co-existed in the air due to agglomeration.



(a) 210 X

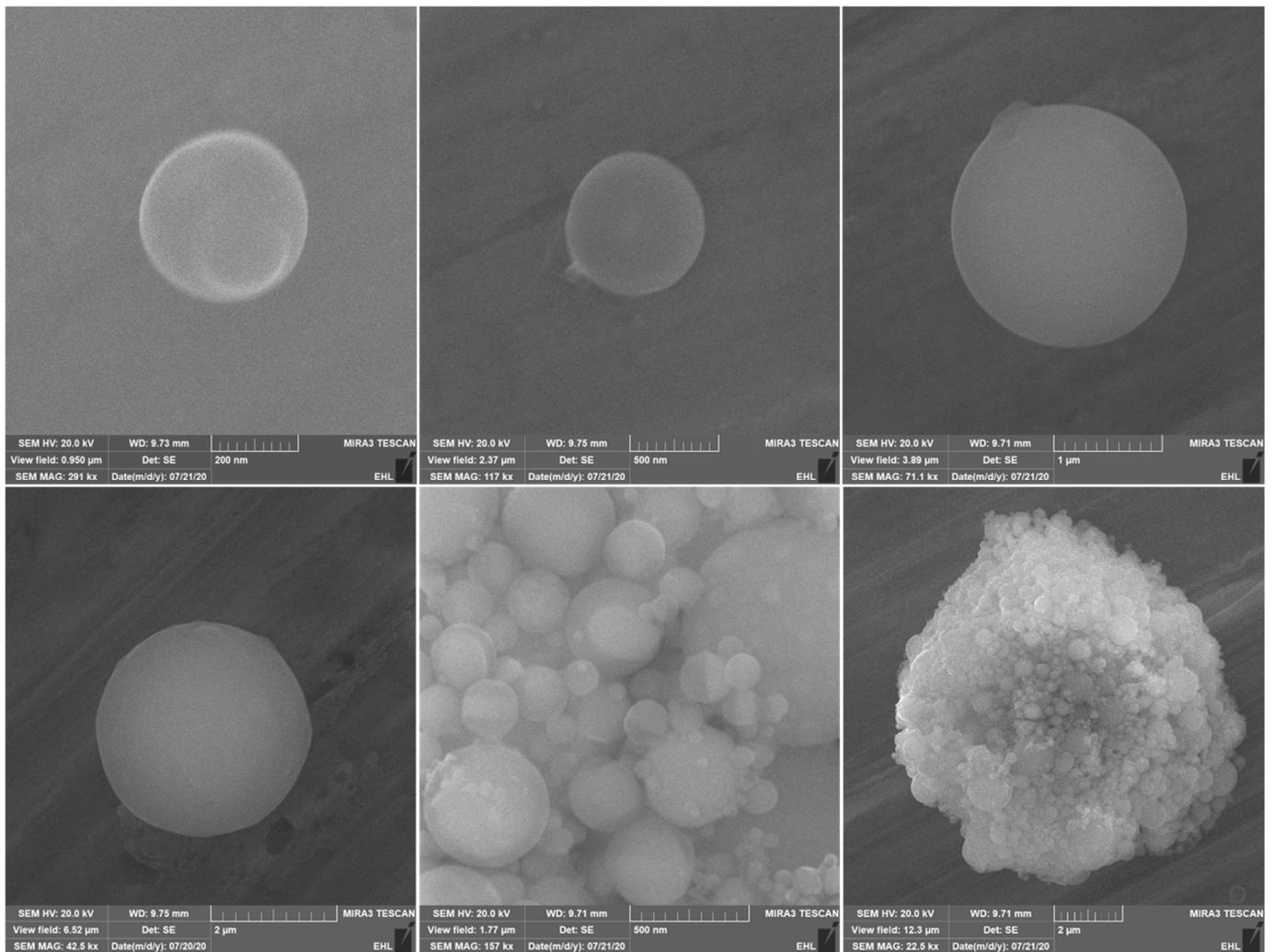


(b) 501 X



(c)

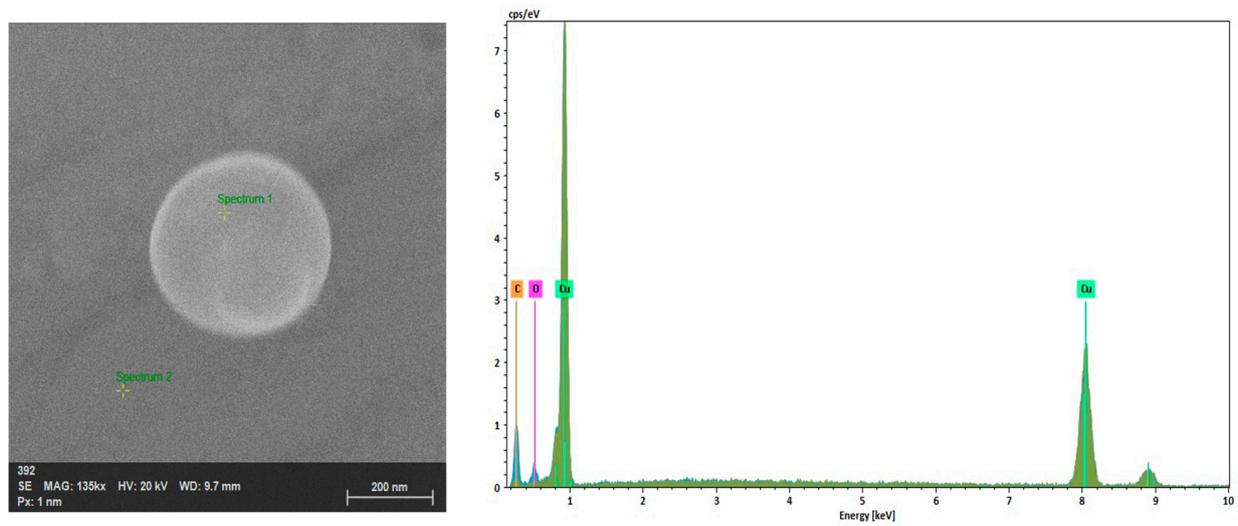
**Figure 9.** (a,b) SEM images of large particles collected using the open-face passive sampler during the fire event at different magnifications; (c) elemental compositions for the measured particles in (a). Note: the copper was mainly from the copper tape substrate.



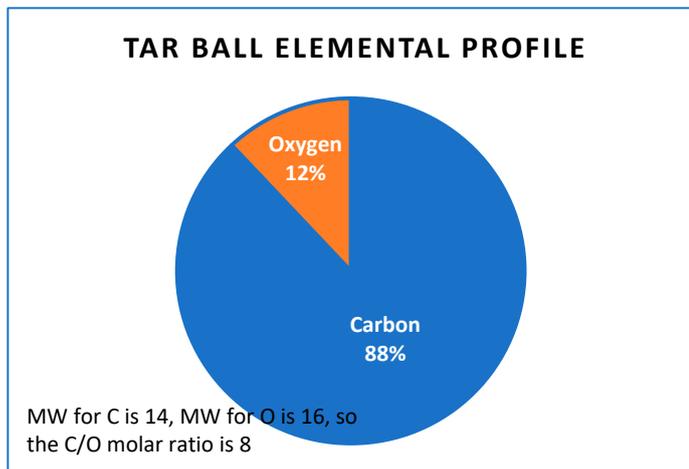
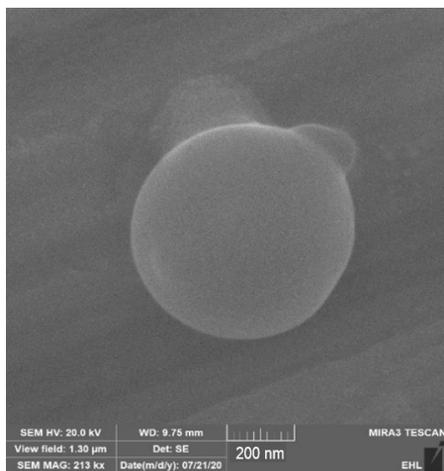
**Figure 10.** SEM images of spherical particles collected using OFPS.

In addition to carbon and oxygen (Figure 11), some tar balls also included sulfur (S), as shown in Figure 11c. This is reasonable since sulfur is also present in wood and grass [54]. For the small tar ball that only contained carbon and oxygen (Figure 11b), the measured molar ratio of carbon to oxygen was 8, which is different from the value of 6 previously reported [55]. We repeated the measurement multiple times on many different particles and obtained values between 7.1 and 8.4, with an average value of 7.9 and a standard deviation of  $\pm 0.57$ . Note that since copper substrates were used for this measurement, there was no carbon substrate effect on the carbon concentration calculation, a potentially more accurate and reliable method for carbon content determination [43].

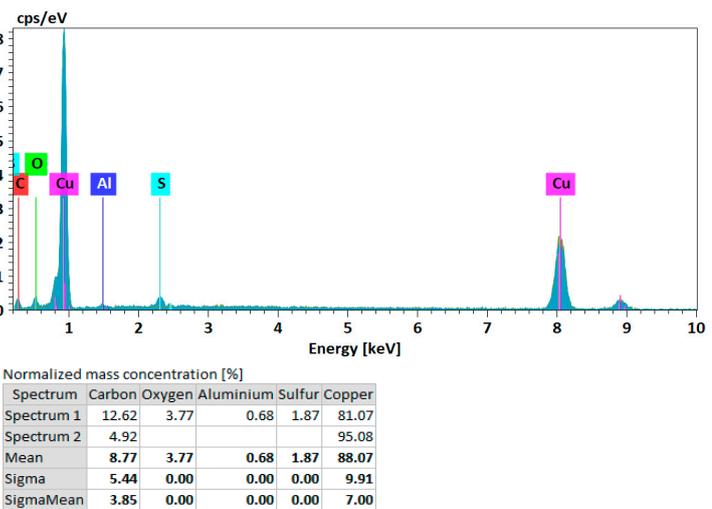
To construct an elemental profile of the wildfire episode PM, over 100 spherical particles were randomly selected and analyzed using EDS, and the overall average element compositions are shown in Figure 12. Carbon and oxygen were the dominant elements, including tar balls and other carbon-rich particles. The other major elements in the wildfire-generated particles included silicon, calcium, aluminum, iron, magnesium, and sulfur. Minor elements included potassium, chloride, sodium, titanium, phosphorus, barium, and zinc, as shown in Figure 13.



(a)



(b)



(c)

**Figure 11.** Typical elemental images, spectra, and profiles of tar balls. (a) Only carbon and oxygen were detected for small tar balls. (b) Carbon/oxygen ratio calculation for tar ball. (c) Sulfur had been detected for some tar balls. Note: Cu was from the substrates.

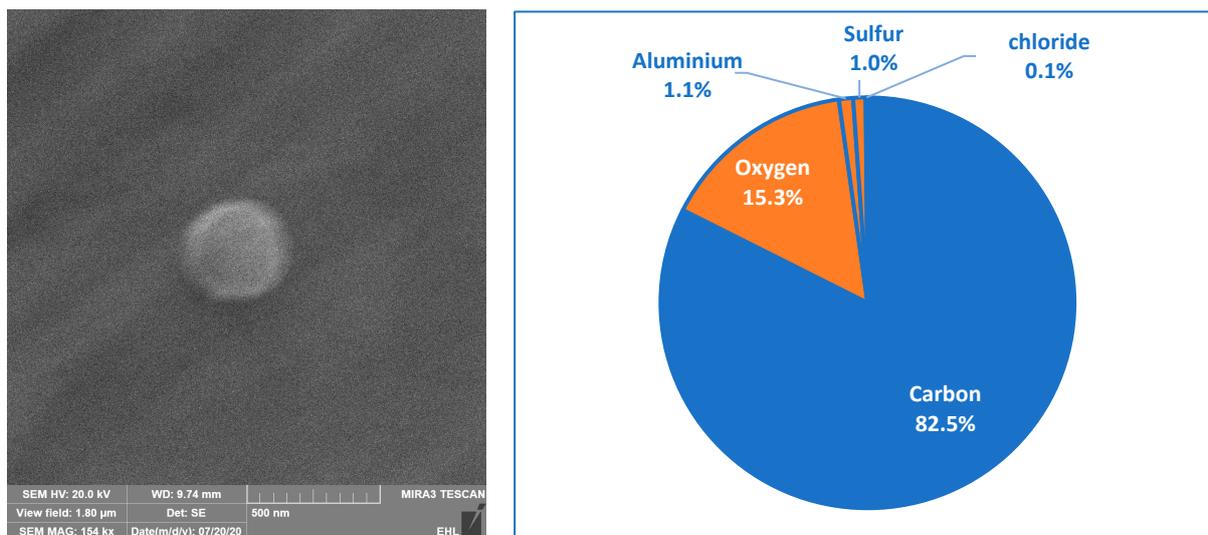


Figure 12. Elemental profile of tar balls during the wildfire episode and example tar ball image.

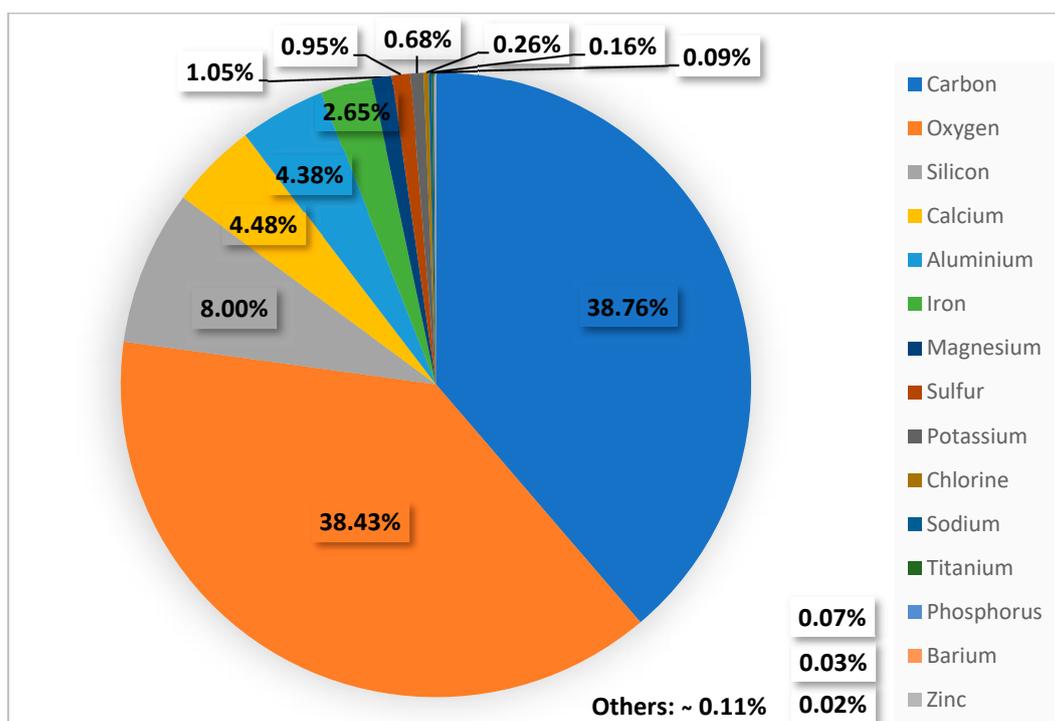
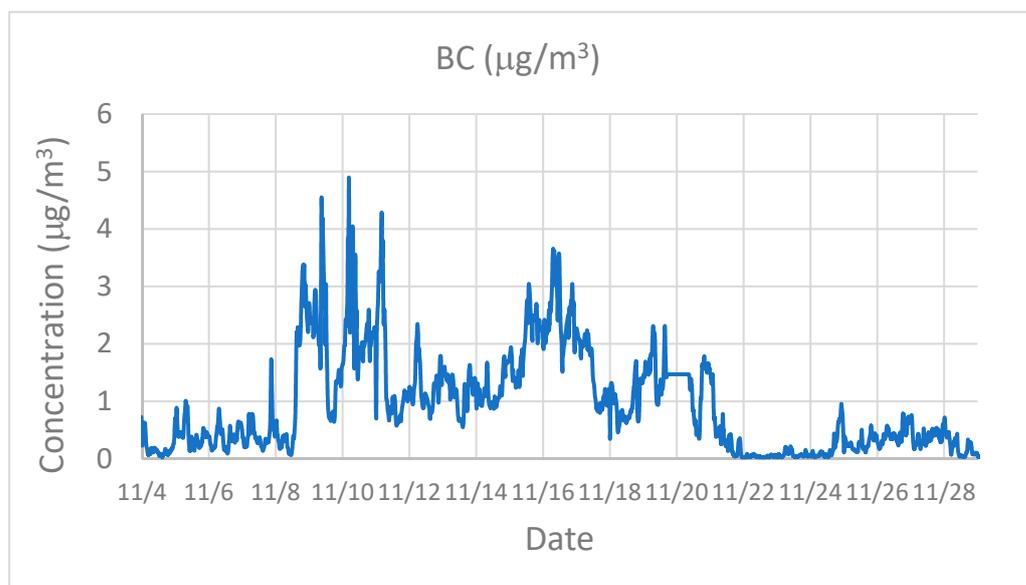


Figure 13. Overall particulate matter elemental profile during the wildfire episode.

In comparison, the total carbon percentage of ambient PM under normal conditions has been reported in other urban regions to be 12–18% [56], while the elemental analysis of PM sampled during non-fire season in the San Francisco Bay Area showed that it was 14% [43]. The higher carbon content of 38.76% detected in the wildfire PM during the Camp Fire period likely indicates enhanced incomplete combustion products.

### 3.4. Black Carbon Enrichment during the Wildfire Episode

Black carbon (BC) is a product of the incomplete combustion of organic materials and wildfire-generated PM [57]. Figure 14 shows the data from BAAQMD’s Richmond site measurements.



**Figure 14.** Black carbon concentration in Richmond during the Camp Fire.

In non-fire conditions, the average black carbon concentration in Richmond was  $0.38 \mu\text{g}/\text{m}^3$ . In contrast, the average black carbon concentration during the Camp Fire was  $1.33 \mu\text{g}/\text{m}^3$ , which is approximately 3.5 times higher. As shown in Figure 14, the highest black carbon peak value was above  $4.9 \mu\text{g}/\text{m}^3$  during the Camp Fire, which is about 13 times higher than typical background peaks. Sustained, high levels of black carbon could potentially cause health impacts for both humans and plants [58,59].

#### 4. Conclusions

Air measurements conducted in an urban area during a wildfire episode yielded high PM and VOC concentrations even though the fire was located over 240 km away. Several toxic VOCs not detected in non-fire samples were identified in the wildfire period, including sec-butylbenzene, 1,2,4-trimethylbenzene, n-propylbenzene, o-xylene, and styrene. Other VOCs such as 1,3,5-trimethylbenzene, ethylbenzene, toluene, and benzene were present in the non-fire air, but they were found in increased concentrations during the wildfire episode. BTEX concentrations were several times higher during wildfires. The increased VOC concentrations measured during the wildfire are potentially hazardous to human health and are known to cause skin irritation and asthma-like symptoms upon inhalation. Due to the limitations of our target compound list, the reported VOCs in this study may not represent all VOCs emitted during the Camp Fire, including even more toxic oxygenated VOCs. More wildfire gas-phase monitoring methodologies need to be developed in the future to better characterize VOCs directly emitted from wildfires and their transformation in the atmosphere.

The highest daily average  $\text{PM}_{2.5}$  value during the fire period approached  $200 \mu\text{g}/\text{m}^3$ , and the highest hourly average was approximately  $270 \mu\text{g}/\text{m}^3$ . The daily average  $\text{PM}_{2.5}$  concentrations in the Bay Area during the entire fire episode was  $102 \mu\text{g}/\text{m}^3$ . SEM analyses revealed that the PM included a high concentration of particles consistent with organic carbon tar balls. These results suggest the necessity to measure and monitor not only PM but also gas phase composition during wildfire season to inform public health. The detailed chemistry of wildfire smoke from different combustible fuels, locations, and temperature profiles needs to be studied further [32]. Different conditions may result in different compounds [37,51], especially for the oxygenated, aged smoke carried over long distances and throughout long fire seasons into heavily populated, urban areas [10].

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