

Article

The Impact of Fireworks on Selected Ambient Particulate Metal Concentrations Associated with the Independence Day Holiday

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Abstract: Fireworks are often used in celebrations and are a known transient source of extreme particulate air pollution, and the particles produced by fireworks are known to contain potentially harmful heavy metals. This study investigated ambient particulate metal concentrations associated with heavy firework use during the United States Independence Day holiday in July 2020 and July 2021 in Fullerton, California, located within the greater Los Angeles metropolitan area. For this study, barium (Ba), chromium (Cr), copper (Cu), lead (Pb), and strontium (Sr) were quantified, with Ba, Cu, and Sr being known tracers for fireworks and Cr and Pb being potentially harmful. Total suspended particulates (TSP) were collected with filters and then extracted and analyzed by graphite furnace atomic absorption spectroscopy. Hourly ambient particulate concentrations at a nearby monitoring station exceeded $500 \mu\text{g m}^{-3}$ and $300 \mu\text{g m}^{-3}$ in 2020 and 2021, respectively. Greater concentrations of overall particulate matter and ambient metal concentrations were observed during 2020 when compared to 2021, consistent with studies in the literature that have shown increased firework use in the area, likely due to the COVID-19 restrictions in place in 2020. In 2021, the Ba, Cu, and Sr concentrations peaked overnight on 4–5 July as expected, but the Cr and Pb concentrations peaked in the afternoon on July 5. In 2020, the peak concentrations of Cr and Pb were $510 \pm 40 \text{ ng m}^{-3}$ and $710 \pm 30 \text{ ng m}^{-3}$, respectively, while $4900 \pm 200 \text{ ng m}^{-3}$, $3860 \pm 40 \text{ ng m}^{-3}$, and $1810 \pm 30 \text{ ng m}^{-3}$ were observed for Ba, Cu, and Sr, respectively, among the highest ever observed to our knowledge.

Keywords: pyrotechnics; graphite-furnace atomic absorption spectroscopy; heavy metals; aerosol; air quality



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1. Introduction

Human exposure to high ambient aerosol particle concentrations, both long-term and short-term, increases the risk of adverse health effects in humans [1–7]. The World Health Organization estimates that air pollution (indoor and outdoor) causes nearly 7 million premature deaths per year globally [8]. Particulate matter (PM) can be classified by size, which in turn determines where particle deposition takes place in the respiratory system. PM with an aerodynamic diameter less than or equal to $10 \mu\text{m}$ (PM_{10}) can deposit in the upper respiratory system, while smaller sized particles less than or equal to $2.5 \mu\text{m}$ ($\text{PM}_{2.5}$) can penetrate the deep lung, allowing any potentially toxic compounds in the particles to enter the bloodstream. The long-term and short-term exposure of humans to $\text{PM}_{2.5}$ are connected to premature death, increased preterm birth rates, lung and heart disease, reproductive issues, asthma, stroke, inflammation, and the oxidation of tissues [1–18].

Fireworks are used globally to celebrate various holidays. Fireworks, firecrackers, and sparklers contribute to extreme concentrations of particulate matter in the atmosphere, albeit typically for short periods of time. Studies around the world, including in southern California, have shown that firework emissions increase particulate concentrations to extreme concentrations that may impact human health [16–35].

A unique aspect of firework emissions is that they are a source of potentially toxic metals. For example, studies have found that ambient concentrations of metals, such as barium (Ba), chromium (Cr), copper (Cu), lead (Pb), and strontium (Sr), increase dramatically during firework events when compared to non-firework days and that these metals may impact human health [21,24,28,36–43]. Fireworks contain different metal salts that serve as oxidizers, ignitors, and colorants, and these metals are incorporated into the aerosol particles to which humans are exposed. For example, Ba, Cu, and Sr salts are used to produce green, blue, and red colors, respectively, and are known markers for firework emissions. Other metals, such as lead and chromium, are potentially present as contaminants. These metals and others may cause various environmental and health effects. For example, chronic exposure to lead (Pb) has been shown to affect cognition, especially in children [44–50], and exposure to chromium can be potentially toxic and carcinogenic in humans depending on the oxidation state [51–53]. Metals can also accumulate in soil and groundwater, leading to alternate pathways of potential exposure or potentially harming flora and fauna [25,39,54,54–57]. Therefore, it is important to monitor particulate ambient concentrations of heavy metals during extreme transient pollution events to better understand how they may impact human health and the broader environment.

In the United States, the Independence Day holiday is celebrated with fireworks on the evening of July 4, with large displays during sanctioned public events and heavy use by private households. Within the Los Angeles basin, due to high population density, the July 4 Independence Day holiday is marked by the extremely heavy use of fireworks, firecrackers, and sparklers [33,34]. According to the South Coast Air Quality Management District (SC-AQMD), the California state government agency responsible for air quality monitoring in the Los Angeles basin, July 4 and July 5 are some of the worst days of the year for air quality in southern California, with short-term ambient PM concentrations more than 10 times the United States Environmental Protection Agency (EPA) 24 h limits for PM_{2.5} (35 $\mu\text{g m}^{-3}$ as of time of this publication) [58,59].

Although pollution events from fireworks tend to be short-lived (one to several days), given the high population within the Los Angeles basin, heavy firework use may greatly impact human health, potentially exposing millions of people to high PM concentrations and high concentrations of metals in ambient air. Importantly, recent studies of Independence Day pollution in the vicinity of this study found that race/ethnicity may play a role in exposure, with areas with larger Hispanic populations in southern California having more air pollution, potentially an environmental justice issue for those communities [33,34]. Therefore, ambient measurements of particulate pollution are much needed in the area to better understand how air pollution during firework use may affect public health within certain populations.

The purpose of this study was to perform measurements in Fullerton, California, located within the Los Angeles air basin, approximately 40 km from downtown Los Angeles, during the July 4 Independence Day holiday in 2020 and 2021 to better understand how fireworks can affect air quality, especially with metal-containing particles. Particulate-based barium (Ba), chromium (Cr), copper (Cu), lead (Pb), and strontium (Sr) were quantified, as these are markers for firework emissions or potentially harmful metals. Other constituents such as potassium, magnesium, aluminum, and sulfur are also commonly found in firework emissions [22,28,36] but were not measured in this study due to instrument limitations.

2. Materials and Methods

2.1. Sampling Site and Information

This study took place at two locations in Fullerton, California, located in the northern Orange County within the Los Angeles air basin, approximately 40 km southeast of downtown Los Angeles, one of the largest population centers within the United States. Figure 1 shows a map of the sampling area within the context of California and the Los Angeles area. In 2020, due to COVID-19 restrictions, samples were collected at a private residence approximately 3 km west of the campus of California State University, Fullerton (CSUF). In 2021, samples were collected on the CSUF campus outdoors in the Biology Plant Facility.

Figure 1c shows a detailed view of the sampling sites from 2020 and 2021, along with the location of the nearest regulatory monitoring site at Anaheim operated by SC-AQMD. Wind roses, shown as insets, were generated using data from the SC-AQMD Anaheim site. It can be seen that during the sampling periods, the wind direction was consistently from the southwest, typical of this location at that time of year. The sampling sites were located in a complex environment heavily influenced by ambient background pollution. The sites were located downwind from multiple major freeways and roads, major rail lines, and other potential sources of particulate matter and metal-containing particles [41,60–65]. The 2020 study was performed during the COVID-19 pandemic, with local and freeway traffic reduced. The 2021 study should be considered more representative of ambient conditions in terms of traffic. The years 2020 and 2021 provide a unique opportunity to potentially better understand background sources of metals, such as vehicle and road emissions, due to the COVID-19 pandemic restrictions in place in 2020 that were not in place in 2021, as discussed below.

It is important to note that the sampling site is located within a large metropolitan area that has many potential sources for background pollution, such as vehicular emissions from major roads and freeways, freight and passenger trains, industry, heavy commercial vehicle use near warehouses, photochemical smog, secondary organic aerosol, and primary elemental carbon [60,64–66]. The sampling sites are located approximately 1 km north of a major rail route with regular freight and passenger train services. Both sites are located approximately 2.5 km north of the Riverside Freeway (SR-91) and approximately 6 km northeast of the Santa Ana Freeway (Interstate-5). The 2020 sampling site is located approximately 2.5 km west and the 2021 sampling site is located approximately 0.7 km west of the heavily trafficked Orange Freeway (SR-57). Emissions from SR-57 were not expected to heavily influence the measurements based on wind direction.

Masri et al. [34] studied particulate concentrations in southern California during the Independence Day holiday in 2022 and showed that Fullerton and other cities in the vicinity may have had high concentrations for firework emissions due to the legality of small fireworks. Importantly, Masri et al. [34] also found that race/ethnicity played a role in air pollution during July 4, with areas in the vicinity of this study with larger Hispanic populations tending to be more exposed to potentially harmful pollution, impacting environmental justice in the region.

Figure 2 shows ambient weather conditions at the SC-AQMD Anaheim site during the individual sampling periods for this study, shown as gray shaded areas, with the gray hashed shaded area representing the expected period of heavy firework use. The site is characterized by southwesterly winds, relatively low wind speeds, and mild temperatures.

Table 1 shows details about the sampling time periods used in this study. In 2020, two sampling periods were studied, overnight on July 4–5 and then roughly 24 h later overnight on July 5–6. In 2021, five sampling periods were studied: overnight on July

3–4 (pre-firework), overnight on July 4–5, the afternoon on July 5, the afternoon on July 6, and overnight on July 7–8.

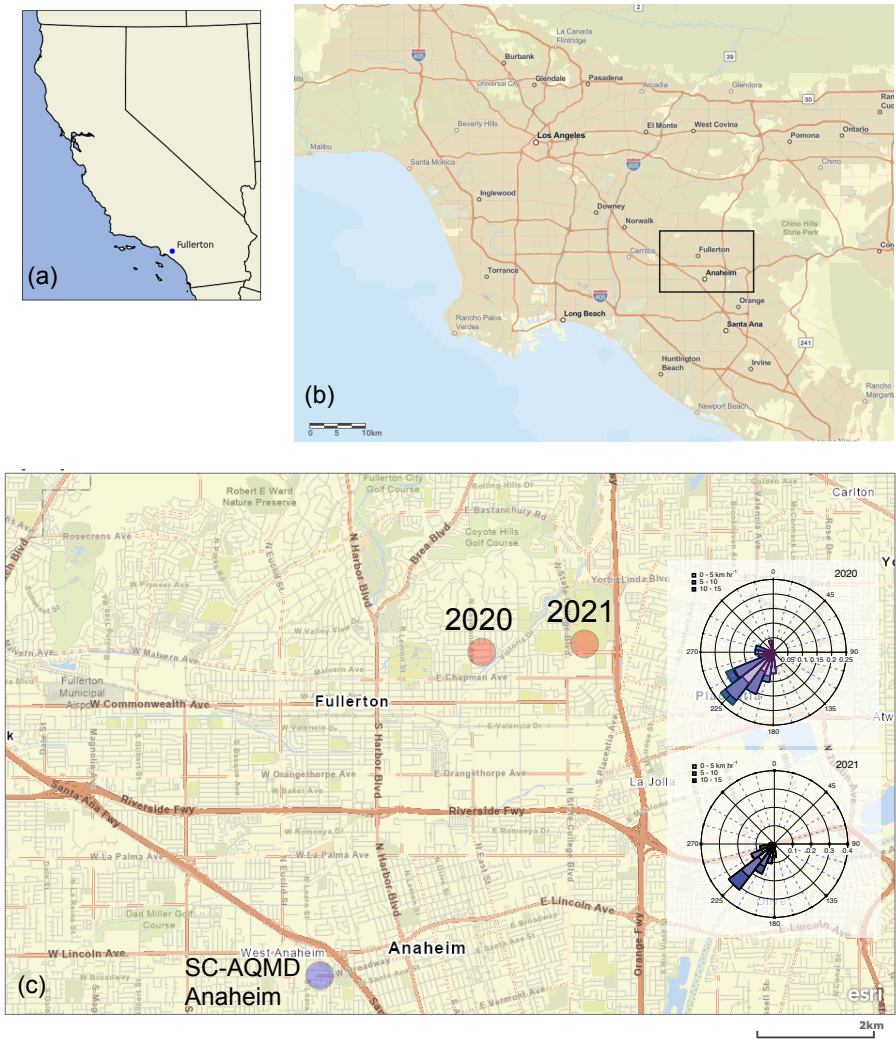


Figure 1. (a) State, (b) regional, and (c) local maps of the sampling site. The rectangle in panel (b) approximates the extent of panel (c). In panel (c), approximate locations of the 2020 and 2021 sampling sites are shown as red circles. For reference, also shown in panel (c) is the approximate location of the South Coast Air Quality Management District (SC-AQMD) Anaheim sampling site (blue circle) and wind rose plots for the sampling periods in 2020 (**top**) and 2021 (**bottom**) as measured at the SC-AQMD Anaheim site.

Table 1. Details of sampling periods for this study.

2020 Sampling Period	Start Time	End Time	Duration (min)
1	Sat. 7/4/2020 20:13	Sun. 7/5/2020 05:28	555
2	Sun. 7/5/2020 20:15	Mon. 7/6/2020 07:11	656
2021 Sampling Period	Start Time	End Time	Duration (min)
1	Sat. 7/3/2021 18:25	Sun. 7/4/2021 04:39	614
2	Sun. 7/4/2021 19:05	Mon. 7/5/2021 02:55	941
3	Mon. 7/5/2021 12:50	Mon. 7/5/2021 16:19	209
4	Tue. 7/6/2021 10:40	Tue. 7/6/2021 15:39	289
5	Wed. 7/7/2021 16:58	Thu. 7/8/2021 01:54	536

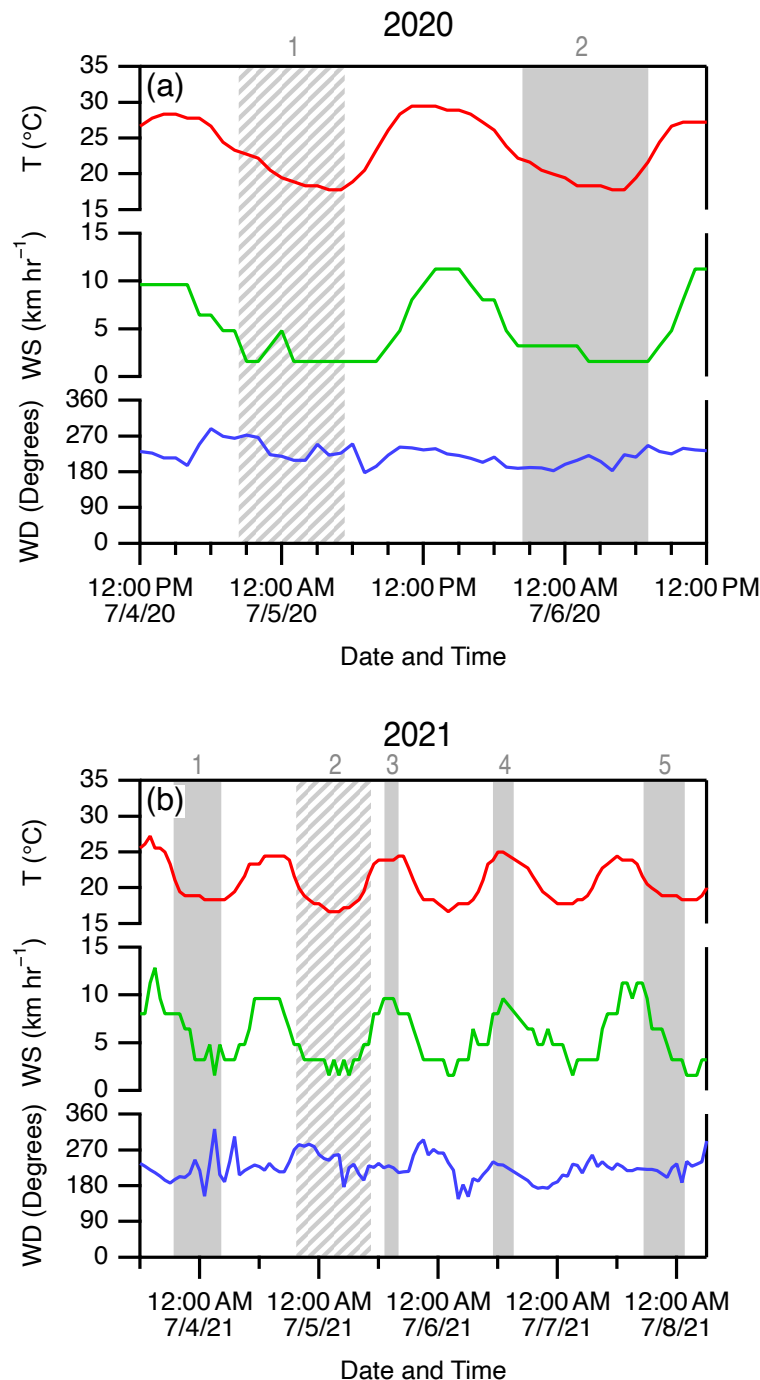


Figure 2. Hourly weather measurements from the SC-AQMD Anaheim site for (a) 2020 and (b) 2021. Gray shaded areas represent sampling periods for this study, while the gray hashed area represents the sampling period associated with heavy firework use. For each year, the temperature (T), wind speed (WS), and wind direction (WD) are plotted.

2.2. Sample Collection

The sampler used in this study was a total suspended particulate (TSP) sampler that collected airborne particles with aerodynamic diameters less than approximately 30 μm , and it is anticipated that the particulate matter generated during heavy firework use is less than 1 μm in diameter [26,35] and should therefore have been effectively sampled. The sampler was a cordless device with a rechargeable battery with a limited battery time of up to approximately 15 h. It was small enough to be portable and durable enough to be deployed outdoors. The TSP sampler was composed of an in-line polycarbonate

filter holder (Pall Corp., Port Washington, NY, USA) connected to a pump (Gillian 5000, Sensidyne, St. Petersburg, FL, USA) by Tygon tubing. Within the polycarbonate filter holder, 47 mm polytetrafluoroethylene (PTFE) filters were placed (Pall Corp., Port Washington, NY, USA). When sampling, the pump operated at a rate of 1.000 L min^{-1} . Before and after sampling, the filter holder was cleaned with trace-metal-grade 1% nitric acid (Fisher Scientific, Pittsburgh, PA, USA) to reduce the potential contamination of each sample. The sampler was placed with the inlet approximately two meters above ground level, facing downward.

2.3. Sample Extraction and Chemical Analysis

For both samplers, PTFE filters were removed from the samplers and stored in plastic petri dishes. The PTFE sample filters were extracted using the US Environmental Protection Agency's Cyclopedia of Methods for Inorganic Pollutants, Method I.O.3 [67]. Briefly, each filter was placed in a glass beaker with 10.0 mL of 70% trace-metal-grade nitric acid (Fisher Scientific), covered with a watch glass, and heated to approximately 90°C for 30 min. The beaker was then rinsed with 10.0 mL of ultrapure water (Nanopure Model D1 1971, Thermo Scientific, 18 $\text{M}\Omega \text{ cm}$ resistivity) and left to rest for 30 min before being transferred into a 25.00 mL volumetric flask and diluted to volume with nanopure water.

The samples were analyzed using the graphite furnace atomic absorption spectrometer (GFAAS) AAnalyst 600 (PerkinElmer, Shelton, CT, USA) with Zeeman effect background correction. Although other studies have used more universal techniques, such as ICP-MS, the GFAAS was chosen in this study due to its high sensitivity and selectivity, the small sample volumes required for analysis ($20 \mu\text{L}$), and the accessibility to the instrument.

Standards of 20, 40, 60, 80, and $100 \mu\text{g L}^{-1}$ made from a multi-element calibration standard (Standard 3, 5% nitric acid, Perkin Elmer) were prepared to calibrate the GFAAS. A matrix modifier was used per the manufacturer's instructions for the analysis of copper and lead. For copper, palladium solution and magnesium nitrate solution were used as the matrix modifier, while for lead, ammonium dihydrogen phosphate solution and magnesium nitrate solution were used. All other instrument parameters were set using the manufacturer's recommendations for each element. More details on the experimental and instrument parameters, such as the furnace temperature program, wavelengths of the lamps, and instrument limits of detection can be found in Rocco (2022) [68].

Liquid concentrations in $\mu\text{g L}^{-1}$ as measured using the GFAAS were converted to ambient ng m^{-3} by determining the volume of air sampled, calculated as the flow rate of the sampling pump (1.000 L min^{-1}) multiplied by the duration of the sampling period. The total mass of each element in 25.00 mL of solution was then normalized to the total volume of air pulled through the filter during the sampling period.

Unused (blank) filters were also extracted to ensure that metal concentrations were due to the particulate matter on the filter. In all cases, metal concentrations from extracted filters were relatively low and near the detection limit of the instrument ($1\text{--}3 \mu\text{g L}^{-1}$). The blank filter concentrations were subtracted from the sample concentrations [68].

3. Results and Discussion

Figure 3 shows the particulate concentrations at the SC-AQMD Anaheim site during the sampling periods of this study, again with shaded areas representing the sampling periods. The measurements show a relatively clean environment prior to firework use and a rapid increase in PM concentrations beginning around sunset on July 4. It can be seen that in 2020, the $\text{PM}_{2.5}$ and PM_{10} concentrations reached nearly $500 \mu\text{g m}^{-3}$, approximately 50 times the current EPA annual average limit of $9 \mu\text{g m}^{-3}$ and 14 times the EPA 24 h average limit of $35 \mu\text{g m}^{-3}$ [59]. In 2021, the peak PM concentrations

were slightly lower than in 2020. We speculate that private firework use was greater in 2020 due to the COVID-19 pandemic shutdown, with more people at their homes, which is consistent with Mousavi et al. [33]. In both years, although the time period where concentrations were extremely high (typically 24–48 h) was relatively short compared to annual averages, the concentrations were extreme and thus greatly impacted air quality during the holiday period.

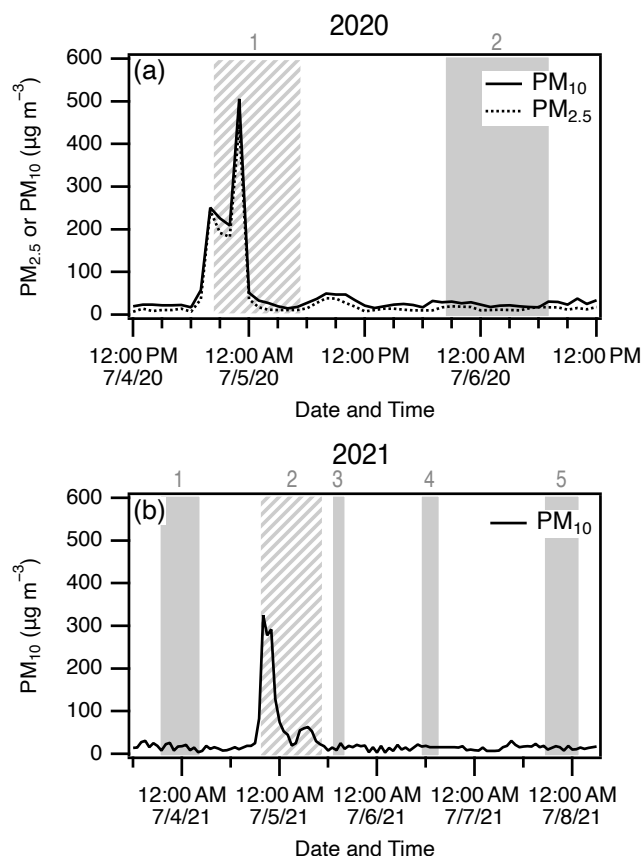


Figure 3. Hourly particulate matter measurements from the SC-AQMD Anaheim site for (a) 2020 and (b) 2021. Gray shaded areas represent sampling periods for this study, while the gray hashed area represents the sampling period associated with heavy firework use. For 2020, PM₁₀ and PM_{2.5} values are plotted, while for 2021, only PM₁₀ measurements were available.

Figure 4 shows the ambient metal results of the 2020 portion of this study. Figure 5 shows the results of the 2021 portion of the study. The data shown in these figures is available in Table A1. The sampling time periods are represented by the width of the boxes shown in Figures 4 and 5 and correspond to the sampling times and dates shown in Table 1. Sampling periods for the 4–5 July measurements for both years are shown as hashed areas, and those time periods were expected to be heavily influenced by firework emissions. The other sampling periods are shown in gray. The 2020 study consisted of two sampling periods, with sampling period 1 taking place in the evening of and overnight on 4–5 July 2020 and sampling period 2 taking place roughly 24 h later. The 2021 study consisted of five sampling periods, with the evening of and overnight on 4–5 July 2021 in sampling period 2. Sampling period 1 was approximately 24 h prior to sampling period 2. Sampling periods 3 and 4 were collected in the midday and afternoon and may not be directly comparable to the 4–5 July sampling period. Sampling period 5 was an evening/night measurement approximately three days after the 4–5 July measurement.

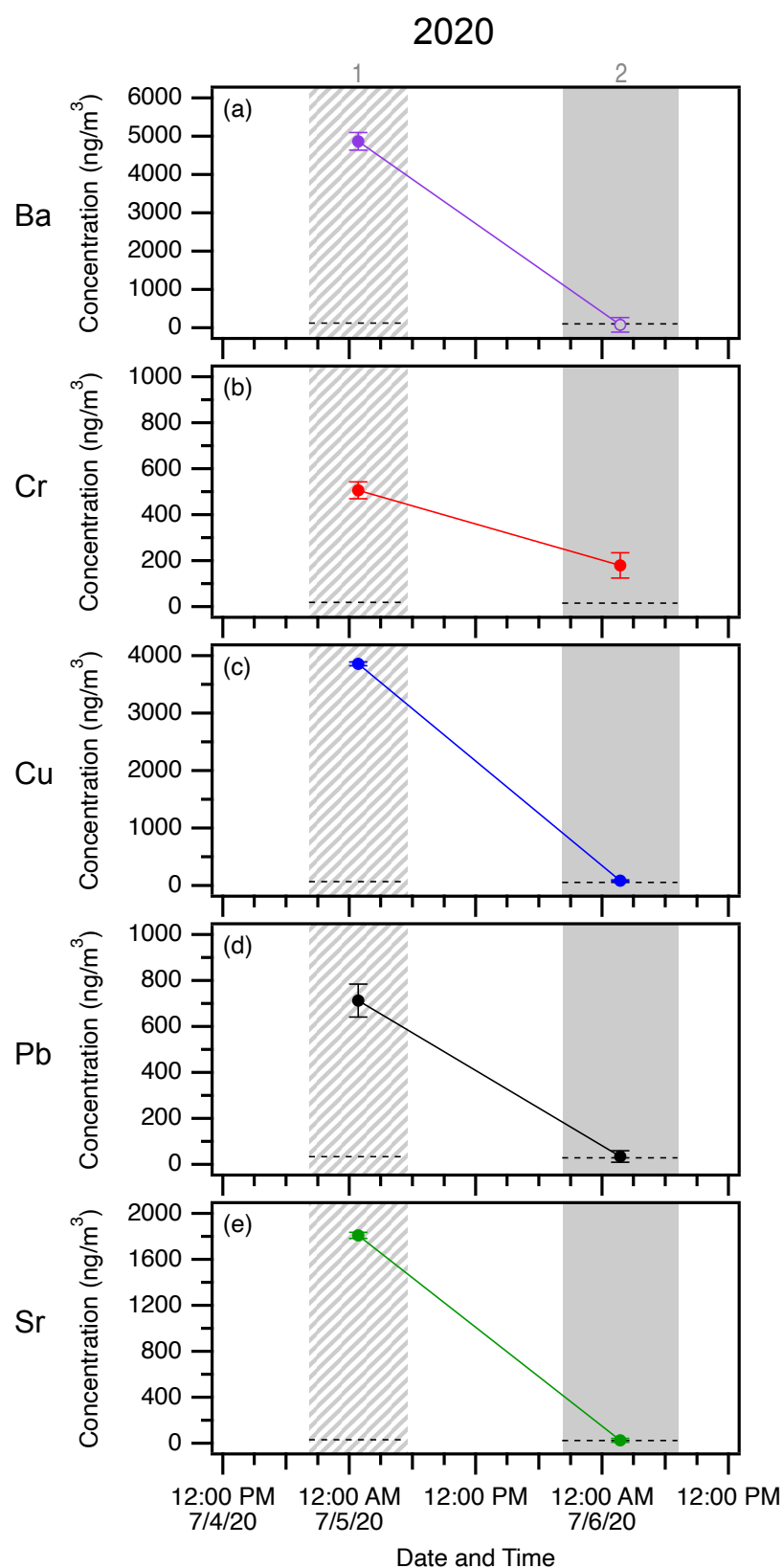


Figure 4. The total suspended particulate (TSP) ambient concentrations of five selected metals in 2020, for barium (a), chromium (b), copper (c), lead (d), and strontium (e). Gray shaded areas represent sampling periods for this study, while the gray hashed area represents the sampling period associated with heavy firework use. Horizontal dashed lines represent the limits of detection and measured values less than the limit of detection are shown as open circles. Note the varying vertical axis limits for each element.

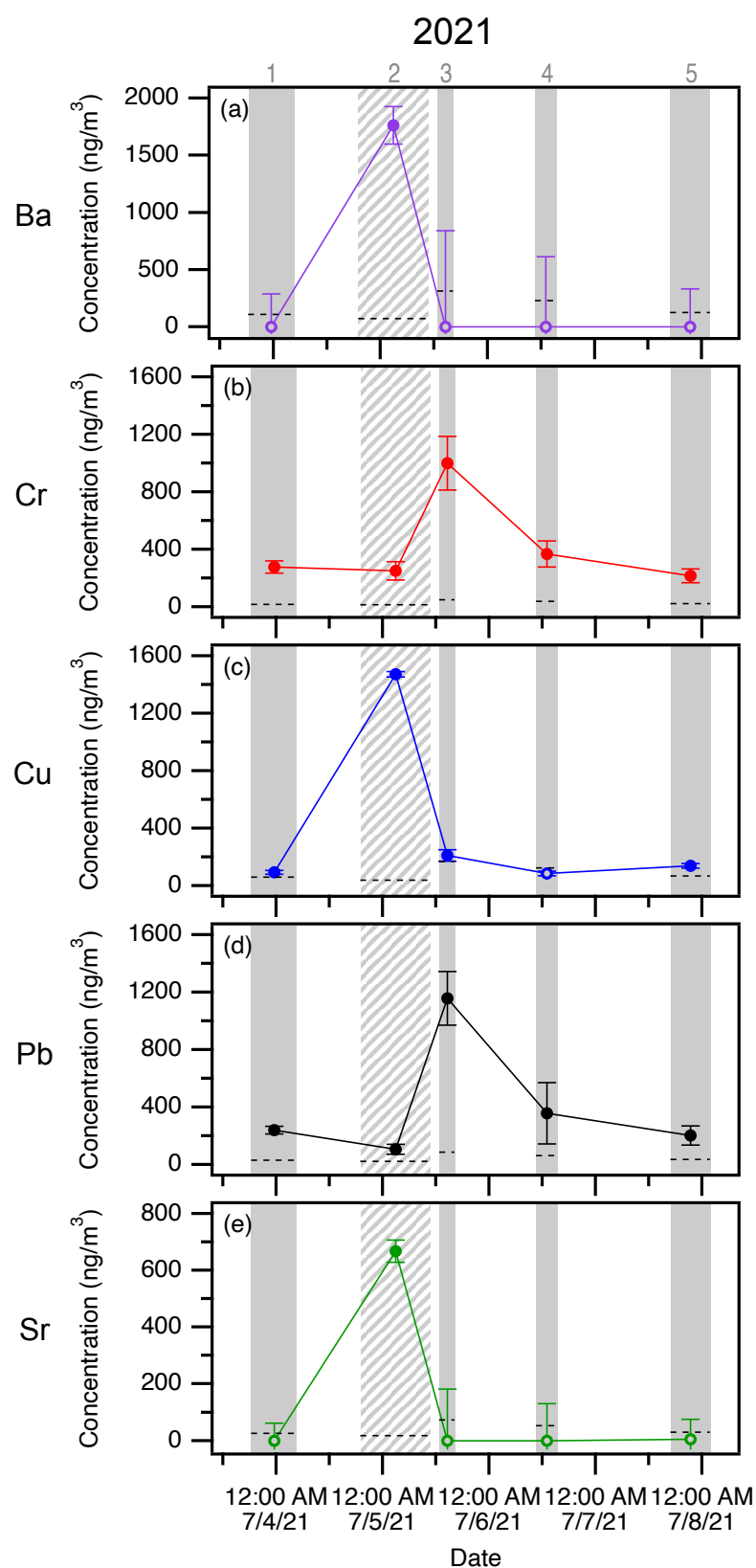


Figure 5. The total suspended particulate (TSP) ambient concentrations of five selected metals in 2021, for barium (a), chromium (b), copper (c), lead (d), and strontium (e). Gray shaded areas represent sampling periods for this study, while the gray hashed area represents the sampling period associated with heavy firework use. Horizontal dashed lines represent the limits of detection and measured values less than the limit of detection are shown as open circles. Note the varying vertical axis limits for each element.

In 2020, it can be seen that the concentrations of each metal were greater on the evening of July 4–5 when compared to July 5–6, indicating that the metal concentrations were likely dominated by fireworks. In general, the metal concentrations were extremely high when compared to the post-holiday time period, with concentrations of $5 \mu\text{g m}^{-3}$ Ba, nearly $4 \mu\text{g m}^{-3}$ for Cu, and nearly $2 \mu\text{g m}^{-3}$ for Sr. Ba, Cu, and Sr are all expected tracers for firework emissions. Cr and Pb concentrations also appear to have been dominated by firework emissions in the 2020 time period, with concentrations of approximately 600 ng m^{-3} decreasing significantly in the post-holiday period.

In 2021, however, it can be seen that the concentrations of Ba, Cu, and Sr were heavily influenced by firework activity on the evening of July 4, but those of Cr and Pb were relatively low during the evening of July 4 but were very high in the afternoon of July 5 before returning to background levels after a few days. It is not clear why the Cr and Pb concentrations peaked after the July 4 holiday, but we speculate that in 2021, these sources may have been dominated by background air pollution at the site or that the transport of particles within the air basin may have influenced the measurements. The 2020 measurements may show relatively low background concentrations of Cr and Pb due to decreased traffic, train, and shipping activity in the area during COVID-19 restrictions. However, it is important to note the even Cr and Pb seem to have been influenced by the July 4 holiday, as the July 5 measurements were significantly above the non-holiday concentrations for those elements. Future studies are planned to better understand the potential sources of background air pollution at this site.

It can be seen in Figures 4 and 5 that metal concentrations were greater in 2020 when compared to 2021. This is consistent with the SC-AQMD PM measurements (Figure 3) that showed greater particulate concentrations in 2020 when compared to 2021. We speculate that the COVID-19 restrictions in place led to greater firework use at private residences in the area. The results are consistent with Mousavi et al. [33], which found that PM concentrations in California were 50% greater during Independence Day in 2020 when compared to 2019, likely due to COVID-19 restrictions. In addition, the 2020 sampling site was located in a residential neighborhood with an extremely high personal use of fireworks. The 2021 sampling site, located on the campus of CSUF, was slightly removed from the residential areas where firework use was taking place, which may have also influenced the total concentration of particles. Future studies are planned to better understand how total PM concentrations may influence metal concentrations during firework periods.

Metal concentrations in this study were either similar to or exceeded metal concentrations found in recent studies from India [36], China [66], the Philippines [22], and Spain [37], although the 2020 results from this study showed extremely high ambient metal concentrations and may be among the highest observed globally, to our knowledge. As mentioned above, this is consistent with the extreme use of personal fireworks during the 2020 COVID-19 restrictions [33]. The high metal concentrations for both 2020 and 2021 in this study may also be attributed to the high population in the area, especially the relatively high percentage of the Hispanic population and legality of fireworks in the area [34]. Future studies will be needed to better understand the consistency of firework emissions in the area and how they compare globally.

The analytical limitations of this study include the limited number of metals analyzed. Although it is highly selective and sensitive, the GFAAS technique requires a different lamp for each element, and measuring multiple metals is time-consuming. The five elements analyzed here were chosen for this study as representative metals including markers for firework emissions. However, we were unable to measure other potential metals such as aluminum, magnesium, and potassium at this time.

Another potential limitation is the relatively low time resolution of the study. In order to collect enough material to be able to observe metals, relatively long filter captures were required. For this study, the collection times ranged from approximately 3.5 h to 15.6 h. Because the production and subsequent removal of particles from the atmosphere occurs on a relatively short time scale (roughly 24–48 h), a higher time resolution may provide important insights into potential exposures of the population to PM during short-lived firework events.

4. Conclusions

The air quality in the Los Angeles basin is greatly impacted by the use of fireworks during the annual July 4 Independence Day holiday. Importantly, fireworks' particulate emissions may contain potentially harmful metals and are subsequently a source of exposure for the population in the area when the particles are breathed in, especially for PM_{2.5} particles, which may reach the deep lung. This study quantified ambient particulate metal concentrations for Ba, Cr, Cu, Pb, and Sr in Fullerton, California, during the Independence Day holidays of 2020 and 2021 by collecting particles on filters, extracting them, and performing atomic absorption spectroscopy. It was observed that these metal concentrations were influenced by firework use and that the metal concentrations were extremely high during a 1–2 day period around the holiday. In 2020, the metal concentrations were among the highest metal concentrations ever observed from firework studies globally, to our knowledge. In addition, it was observed that the metal concentrations were greater in 2020 than 2021, possibly due to increased household firework use in 2020 due to COVID-19 restrictions.

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Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript:

GFAAS	Graphite furnace atomic absorption spectrometer
PM _{2.5}	Particulate matter with diameter $\leq 2.5 \mu\text{m}$
PM ₁₀	Particulate matter with diameter $\leq 10 \mu\text{m}$
TSP	Total suspended particulates
CSUF	California State University, Fullerton

SC-AQMD	South Coast Air Quality Management District
EPA	United States Environmental Protection Agency
PTFE	Polytetrafluoroethylene
Ba	Barium
Cr	Chromium
Cu	Copper
Pb	Lead
Sr	Strontium

Appendix A

Table A1 lists metal concentrations from this study.

Table A1. Results for metal concentrations from this study, listed by the sampling period. * indicates the time period of the heaviest firework use. All concentrations are in ng m^{−3}. nd = not detected (the concentration was less than the limit of detection). Uncertainties are retrieved standard deviations (s_x) based on calibration curves and repeated measurements. Values are rounded to the correct number of significant digits based on the uncertainties.

Year	Sampling Period	Ba	Cr	Cu	Pb	Sr
2020	1 *	4900 ± 200	510 ± 40	3860 ± 40	710 ± 30	1810 ± 30
2020	2	80 ± 200	180 ± 60	80 ± 10	40 ± 30	30 ± 20
2021	1	nd	280 ± 40	90 ± 10	240 ± 30	nd
2021	2 *	1800 ± 200	250 ± 60	1470 ± 20	110 ± 40	670 ± 40
2021	3	nd	1000 ± 200	200 ± 40	1200 ± 200	nd
2021	4	nd	370 ± 90	nd	400 ± 200	nd
2021	5	nd	220 ± 50	140 ± 20	200 ± 70	nd

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