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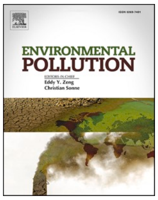

Source-resolved black carbon and $PM_{2.5}$ exposures during wildfires and prescribed burns[☆]

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ABSTRACT

Changes in climate and land-use have significantly increased both the frequency and intensity of wildland fires globally, exacerbating the potential for hazardous impacts on human health. A better understanding of particle exposure concentrations and scenarios is crucial for developing mitigation strategies to reduce the health risks. Here, $PM_{2.5}$ and black carbon (BC) concentrations were monitored during wildland fires between 2022 and 2024, in fire-prone areas in Catalonia (NE Spain), by means of personal monitors (AirBeam2 and Micro-aethalometers AE51 and MA200). Results revealed that exposures to combustion aerosols ($PM_{2.5}$ and BC) were significant and comparable during wildfires and prescribed burns (mean $PM_{2.5}$ during wildfires = 152 μg/m^3 vs. 110–145 μg/m^3 for prescribed burns). Overall, BC/$PM_{2.5}$ ratios showed a large variability as a function of the monitoring scenario, indicating varying contributions from mineral aerosols to the emissions mix originating from fire management and extinction tasks. Specifically, mop-up tasks (final extinction tasks involving stirring top soil using handheld tools) were identified as a significant contributor to $PM_{2.5}$ exposures, with 1-min $PM_{2.5}$ peak concentrations reaching up to 1190 μg/m^3. These results may be especially valuable for emissions modelling. Source apportionment of multi-wavelength BC datasets provided deeper insights into emissions and their impact on exposure profiles: line operators (who control the fire perimeter) were predominantly exposed to biomass burning smoke BC_{bb} (61%) when compared to BC from fossil-fuel combustion (BC_{ff} = 39%), while torchers (in charge of initiating technical fires using fossil-fuel drip-torches) were predominantly exposed to BC_{ff} (77% vs. 23% BC_{bb}). These findings highlight the value of portable monitors in the assessment of wildfire emissions and impacts on human exposure and environment. The combination of these tools, reporting data in real-time and with high time-resolution, is key to the design and implementation of effective mitigation strategies for environmental and health concerns related to wildland fires.

1. Introduction

Due to changes in climate and land-use, wildland fires are increasing globally (Cascio, 2018; Jones et al., 2022; Brown et al., 2023; Burke et al., 2023; Chen et al., 2024), and so are their impacts on environmental and human health. The growing intensity and frequency of wildfires has become an issue of environmental concern (Buechi et al., 2021; Wibbenmeyer and Robertson, 2022; Burke et al., 2023), as well as their impacts on regions which were traditionally not fire-prone (Jones et al., 2024). Whereas the impacts of aerosols on human health are well known (Lelieveld et al., 2019; Oberdörster, 2000; Keywood et al., 2015; Kramer et al., 2023; Burke et al., 2023), recent studies target the specific impacts of wildland fire smoke exposures (Naeher et al., 2007; Youssouf et al., 2014; Miranda et al., 2012; Navarro et al., 2019; Navarro, 2020; Chen et al., 2021; Scieszka et al., 2023; Stowell et al., 2021; Gao et al., 2023; Yin, 2023; Yu et al., 2022; Zhang et al., 2023; Maji et al., 2024; Siregar et al., 2024; Wei et al., 2023), concluding on the association between long-term non-occupational exposure to wildland fire smoke and numerous adverse health outcomes ranging from respiratory and cardiovascular disease to mental health disorders (Aguilera et al., 2021; Matz et al., 2020; among others). In line with these studies, the International Agency for Research on Cancer (IARC) classified the occupation

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of firefighting as carcinogenic, due to occupational exposure to wildland fire smoke (Demers et al., 2022). This is due to the higher toxic potential of $PM_{2.5}$ from wildland fire smoke than from other sources (e.g., construction/demolition, vehicular traffic, etc.) through its potential to cause lung inflammation and oxidative stress (Aguilera et al., 2021).

Because these impacts are expected to continue to increase (Burke et al., 2023), the effective implementation of risk mitigation strategies should become a priority. Environmental impacts may be addressed by means of landscape and vegetation management, which aims to reduce the amount of viable fuel available for combustion (Murray et al., 2023). In terms of human health, mitigation is linked to exposure reduction, which in turn requires a detailed understanding of the aerosols generated during wildland fires. However, aerosol monitoring during wildfires is highly complex from a logistical perspective, as aerosol instrumentation must be portable, able to withstand the harsh conditions of firefighting environments, be minimally invasive to avoid hindering the firefighter's movements and activities, and able to monitor different aerosol properties (e.g., $PM_{2.5}$ mass, carbonaceous aerosols or polycyclic aromatic hydrocarbons (PAHs) concentrations; Naeher et al., 2007; Adetona et al., 2011, 2013). Environmental factors such as wind patterns, topography, and burn intensity play a key role in smoke dispersion and exposure, further highlighting the complexity of wildfire impacts (Siregar et al., 2022). As a result, prescribed burns are increasingly being used as proxies for wildfire scenarios (Fernández-Guisuraga and Fernandes, 2024; Davim et al., 2022; Miller et al., 2020; Cansler et al., 2022), where aerosol monitoring may be carried out in a relatively less complex setting. In addition, prescribed burns are an increasingly used strategy by firefighters for wildfire prevention and management, and are therefore representative of firefighter exposures (Duane et al., 2019; Fernandes, 2013; Morgan et al., 2020).

Specifically, increasing our understanding of the types and sources of aerosols generated during wildfires and prescribed burns is crucial for exposure management and mitigation. Research is available focusing on particulate matter, carbon monoxide and biomass burning markers (Reisen et al., 2011; Reinhardt and Broyles, 2019; Adetona et al., 2011; Wu et al., 2021), but further studies are necessary specifically targeting health hazardous components, such as black carbon (BC). BC is the strong light absorbing component of elemental carbon and acts as a carrier for PAHs and other toxic compounds (Shrestha et al., 2010; Bond et al., 2013). Furthermore, research is especially scarce on source-specific exposures: a variety of $PM_{2.5}$ and BC emission sources contribute to exposure during wildfires, ranging from biomass burning (defined as vegetative material and underbrush), to fossil fuel combustion (from firefighting vehicles and drip torches, which are used for technical firefighting—defined as specialized techniques involving tools like drip torches to intentionally ignite fires for creating firebreaks or conducting prescribed burns (NWCG, 2006)) or mineral dust resuspension (resulting from soil disturbance by wildfire spread and during firefighting activities).

In this framework, the aims of this research are two-fold: (i) to characterize the properties of smoke-derived aerosols, specifically $PM_{2.5}$ and BC, and their impacts on exposure during wildfires and prescribed burns, and (ii) to apply source apportionment methods to understand the contribution from different emission sources on BC concentrations. With firefighters as study subjects, exposure characterization aimed to quantify the $PM_{2.5}$ and BC exposures resulting from various activities, roles, and environments, thereby offering insights into the variability of exposure risks. Subsequently, BC source apportionment focused on discriminating between biomass combustion and fossil fuel contributions. Together, these goals support the development of effective mitigation strategies to minimize the health impacts of wildfire smoke on firefighters. In addition, specifically for the firefighting community, quantifying these exposures provides novel information for developing health protocols and decision support systems.

2. Methodology

2.1. Study locations and subject recruitment

Exposure monitoring was carried out between 2022 and 2024 during 15 prescribed burns in Catalonia (NE Spain). Sampling was performed at different locations including Oristà, Puig-reig, Ódena and Can Feliu in the province of Barcelona; Mont-ras, Torroella de Montgrí and Coll de Banyuls in the province of Girona; Gandesa in the province of Tarragona; and Salàs del Pallars in the province of Lleida. These locations represent a variety of Mediterranean type ecosystems, with a mix of vegetation covers, including shrub, bushes, young trees, and grass. The burn conditions and burnt surface also varied, ranging from 0.3 to 4 ha. This diversity in geographical, ecological, and burn conditions ensures that the findings are representative not only of Catalonia but also of broader Mediterranean biomes, enhancing the relevance of the study to those regions in the world beyond the Mediterranean basin.

Prescribed burns are utilized annually as a management tool, conducted under specific guidelines that include the meteorological prescription window, fuel conditions, and fixed topographies to achieve desired objectives (e.g., elimination of low vegetation, bushes, and young trees; silviculture to prevent fires; and facilitation of the regeneration of certain plant types or pastureland). Conditions and criteria for prescribed burns vary among seasons. Summer-autumn burns are characterized by air temperatures between 24 °C and 30 °C, relative humidity above 35%, and moderate wind speed. On the other hand, winter burns are typically conducted under anticyclonic weather conditions, with air temperatures below 15 °C, relative humidity below 30%, and low wind speed (Bombers, 2019). In this study, these two periods were referred to as PB1 for prescribed burns conducted in September, October and November (summer-autumn), and PB2 for those conducted in January, February and March (winter), respectively. The distinction between PB1 and PB2 was made not only based on the seasonal meteorological conditions but also to account for variations in burn conditions, which may influence exposure concentrations. The meteorological conditions during the prescribed burns were relatively consistent within the window of time when the prescribed burns were conducted, as these conditions must meet specific criteria to permit such activities. However, the time windows for PB1 and PB2 were different, which justified their separate categorization.

From a logistical standpoint, exposure monitoring during prescribed burns entails high complexity, due to the necessary coordination with the firefighter teams and the access requirements. In this sense, the field campaigns during PB1 and PB2 were rather different: direct access to the burn areas was not granted to the research team during PB1, while it was possible during PB2. This largely facilitated a more comprehensive understanding of each case, and allowed for a greater amount of exposure data to be collected during PB2.

For sampling, portable monitors were distributed to the firefighters at the start of each shift, and a post-shift questionnaire was administered to the participating firefighters to collect data on burn characteristics and personal tasks. These tasks defined by the firefighters included lighting, holding (also referred to as fire perimeter controlling), mopping-up and other activities such as truck driving, discharging ⇀ nozzle, post-fire inspection, perimeter inspection, monitoring tasks, control of smoke column, and hotspot control, direct attack firefighting, saw crew work, vehicle mobility and driver tasks, movement with vehicle and chainsaw operations, vehicle patrol and burn operations. Lighting involves the fire ignition process with a dripping torch fueled by a mix of gasoline and diesel (referred to as "torcher"). Holding involves the management of fire within perimeter and use of manual tools to prevent fire spread (referred to as "line operator"). Mopping-up entails the extinguishing of smoldering fire after the major burning phase, by stirring the top-soil layer, using handheld tools (e.g. spades). Truck driving involves operating firefighting vehicles. Nozzle operation is the task of controlling the flow of water or foam through a nozzle, typically during active firefighting. Post-fire inspection refers to the evaluation of fire-affected areas to ensure complete extinguishment and identify potential risks. Perimeter inspection involves monitoring the boundaries of the fire to ensure containment and prevent its spread outside the controlled area. Monitoring tasks include observing and recording fire conditions, air quality, and safety factors. Smoke column control involves managing and monitoring the smoke column to prevent it from becoming a hazard to surrounding areas or impeding visibility. Hotspot control refers to identifying and extinguishing residual fires. Direct attack firefighting involves actively suppressing the fire by applying water directly to the flames. Saw crew work consists of cutting down trees, branches, or vegetation to create firebreaks or clear paths for firefighting efforts. Vehicle mobility and driver tasks involve moving and operating firefighting vehicles to support ground crews or reposition resources as needed. Movement with vehicle and chainsaw operations refers to the action of moving through the fire area with vehicles equipped with chainsaws for clearing vegetation or creating firebreaks. Vehicle patrol and burn operations refer to monitoring the fire area and ensuring the safety and efficiency of the burn operations while traveling in a firefighting vehicle. The operational command center is a centralized location where coordination, communication, and management of prescribed burns take place (NWCG, 2006).

Additionally, monitoring was carried out during 12 wildfires in the region. In the summer 2022, BC records were collected in Catalonia during the Pont de Vilomara (Barcelona) and Castell-Platja d'Aro (Girona) wildfires (Fig. S1 and Table S1 in Supplementary Material (SM)). For particulate matter, records were collected during the Gavarres (Girona), Jonquera (Girona), Tivissa (Tarragona), and Begís (Castellón-Communty of Valencia) wildfires (Table S1). The Begís wildfire was particularly notable for its intensity and the extensive area it burned, covering 19,362 ha.

Records for the 2023 wildfire season were scarce due to the lower incidence of fires; however, some records from the Portbou (Girona), Tivissa (Tarragona) and Mont-roig del Camp (Tarragona) were included in the study (full list is provided in Table S1).

Generally, the exposure records were highly variable and depended on many factors such as the specific conditions of the wildfire or prescribed burn, and the work shift of the firefighter. The exposure periods ranged from 1 to 8 h for prescribed burns and 1–13 h for wildfires.

2.2. $PM_{2.5}$ and BC monitors

At each site, the aim was to monitor the personal exposure of 4–8 firefighters per wildfire or prescribed burn and register their activity, totalling N = 114 for particulate matter and N = 67 for BC across all the events and described activities. In some cases, $PM_{2.5}$ measurements were taken without corresponding BC measurements, and in fewer cases BC measurements were taken without $PM_{2.5}$ data collection, resulting in different sample numbers. Lightweight non-invasive portable monitors were used to monitor personal exposures to $PM_{2.5}$ and BC aerosols. The monitors were carried by the firefighters in their pockets or hanging from their jackets, with the sampling inlet as close to the breathing zone as possible without causing disturbance in the firefighters' tasks.

Black carbon aerosols were monitored using two types of portable Aethalometers, AE51 (single-wavelength) and MA200 (multi-wavelength; AethLabs, San Francisco, USA). Total BC concentrations were monitored at 880 nm. The MA200 monitor determines aerosol light absorption in the range 375–880 nm, and was used to estimate the contributions of BC from fossil fuel and biomass burning emissions (Helin et al., 2018). Four MA200 units and five AE51 units were used in different combinations during the field campaigns, depending on their availability. MA200 were used for the BC source apportionment analysis. Both types of BC monitors collected particles with a 1 min time resolution and at a sampling flow of 100 mL/min.

For $PM_{2.5}$, ten portable sensors (AirBeam2; HabitatMap, Brooklyn, USA) were deployed. The AirBeam2 uses Plantower PMS7003 as particle sensing units, and includes 2 sensing units in parallel per node for quality assurance. It includes a battery, allowing it to operate in standalone mode after configuration, without the need for any additional hardware. During an online record session with AirBeam2, data are communicated via Bluetooth or WiFi to a mobile phone and the onboard GPS enables mapping of the recorded $PM_{2.5}$ concentrations. $PM_{2.5}$ concentrations were integrated into 1-mintue averages.

Before each sampling campaign, the portable devices were intercompared to correct for intra-unit variability. In addition, they were also compared with high-end instrumentation at an EU-reference air quality monitoring station in Palau Reial (Barcelona, Spain) over 4-h to 5-day periods (depending on availability of the monitoring station). BC mass concentrations from the MA200 and AE51 micro-aethalometers were compared with those from a stationary multi-angle absorption photometer (MAAP, Thermo ESM Andersen Instruments), operating at a 1-min time resolution, for quality control. Micro-aethalometer BC concentrations were not corrected with regard to the MAAP concentrations, however, as the MAAP is not a reference instrument and the measurement techniques are not directly comparable. $PM_{2.5}$ concentrations from the sensors were inter-compared with an environmental dust monitor Grimm EDM180, equivalent to EU-reference gravimetric measurements, operated with a 10-min time resolution (Rovira et al., 2022). The $PM_{2.5}$ data from the sensors were corrected with regard to the reference data, as will be shown in section 3.1. The authors acknowledge as a limitation that, during calibration at the Barcelona site, the sensors were challenged with urban aerosols and at lower concentrations than the target aerosol (wildfire smoke, at higher $PM_{2.5}$ concentrations).

2.3. Aethalometer data post-processing using the ONA algorithm

The BC datasets collected with the Aethalometers may generate negative concentrations when monitoring with high time-resolution. Moreover, at short sampling intervals, instrumental noise may cause attenuation (ATN) values to stay constant or even decrease slightly from one period to the next. This noise may lead to erroneously low values at one moment and erroneously high values at the next, or vice versa (Cheng and Lin, 2013). Additionally, due to high concentrations, tape advances in the MA200 Aethalometer can occur as frequently as every 5–7 min, which does not allow the instrument to stabilize and may result in oscillations in the data. The Optimized Noise-reduction Averaging (ONA) algorithm (Hagler et al., 2011) was used in this study to eliminate the negative values and smoothing fluctuations from BC data sets by using a minimum delta attenuation (ΔATNmin) value of zero.

2.4. Source apportionment of BC

Source apportionment was applied to the multi-wavelength BC datasets in order to distinguish between fossil fuel and biomass burning contributions. Two different models were applied: the Delta-C model (Wang et al., 2010), which has been typically used to analyze aerosol absorption in urban areas, particularly in studies assessing traffic-related pollution and wood combustion (Wang et al., 2011, 2012); and the Aethalometer model (Sandradewi et al., 2008b), which has been effectively used in field studies to assess the contributions of biomass burning and fossil fuel combustion to aerosol optical properties in regions with substantial emissions from domestic wood burning and traffic (Sandradewi et al., 2008a; Helin et al., 2018). The Delta-C model, in which biomass burning BC contributions are defined as the difference between the absorbing BC at 375 nm and 880 nm, was previously established as a tracer of wood combustion. This enhanced absorption plays an important role in separating traffic-related emissions (especially diesel) from biomass combustion. However, this method has known limitations as high Delta-C values may be influenced by other absorbing contributors besides biomass burning (such as coal and kerosene combustion and some secondary organic aerosol products) (Harrison et al., 2013; Olson et al., 2015; Zhang et al., 2011; Zhong and

Jang, 2011; Zhang et al., 2017). Due to these limitations, and because the relationship between Delta-C and biomass burning depends on the environmental and combustion conditions (Zhang et al., 2017), the Aethalometer model was proposed (Sandradewi et al., 2008b; Helin et al., 2018). It uses the absorption Ångström exponent (AAE) values at multiple wavelengths to calculate the contribution of fossil fuel combustion (BC_{ff}) and biomass burning (BC_{bb}) to BC (see equations (1)–(10) in SM). This method enhances the accuracy of source apportionment by considering the wavelength dependency of aerosol absorption, making it more robust in diverse environmental conditions (Sandradewi et al., 2008b).

Previous studies determined AAE_{ff} and AAE_{bb} values in various contexts. For instance, Sandradewi et al. (2008c) and Zotter et al. (2016) used auxiliary measurements such as EC/OC and ^{14}C analyses in urban areas impacted by BC mainly from high vehicular emissions and residential wood burning. On the other hand, Fuller et al. (2014) and Titos et al. (2017) employed levoglucosan measurements in urban environments affected by wood burning emissions, as levoglucosan is commonly used as a tracer for biomass burning-derived aerosols (Helin et al., 2018). In the present study, an $AAE_{ff} = 1$ was adopted, following Kirchstetter et al. (2004) and Martinsson et al. (2017). Regarding AAE_{bb}, a range of values from 2 to 3 were tested based on the literature (Martinsson et al., 2017; Massabò et al., 2015; Sandradewi et al., 2008a). The Delta-C method was then used as a benchmark for evaluating the performance of the Sandradewi method (see equation (11) in SM).

2.5. Statistical and data analysis

Statistically significant differences between different scenarios (Wildfire, PB1 and PB2) and different exposure profiles were assessed for $PM_{2.5}$ and BC. Differences among groups were assessed using the parametric one-way ANOVA test for data with normal and lognormal distributions. The normality of the data was evaluated with the Shapiro-Wilk test. When the Shapiro-Wilk test indicated non-normality, the Kruskal-Wallis test was used. For comparisons involving only two groups, the Kruskal-Wallis H-test was equivalent to the Mann-Whitney *U* test. For both the one-way ANOVA and Kruskal-Wallis tests, a p-value <0.05 was considered indicative of statistically significant differences between groups.

The data analysis and plots were performed using Python 3 statistical software. Due to the nature of the data, the whiskers of the boxplots were set to (0, 100) to cover the entire range of the data.

3. Results and discussion

3.1. Portable monitor inter-comparison and calibration

The initial step in the study involved data quality control for the different instruments used. Despite the fact that AirBeam2 monitors were calibrated in urban ambient air as opposed to with wildfire smoke, as stated above, this kind of sensor (Plantower) has been successfully used in the past to monitor wildfire smoke (Barkjohn et al., 2021; Gili et al., 2024). The $PM_{2.5}$ inter-comparison, after co-location at a reference air quality monitoring station, showed r^2 values between sensor and reference data ranging from 0.80 to 0.99. The standard deviation (SD), calculated for the combined dataset of sensor and reference data, ranged from 1.63 to 8.40 (Fig. S2). Based on these comparisons, each individual $PM_{2.5}$ sensor was corrected applying a linear correction calculated based on the reference instrument. No evidence of drifts in the sensor response was detected, based on the inter-comparisons carried out at the end of the monitoring period. Nevertheless, there were no inter-comparisons performed at high $PM_{2.5}$ loadings that are more representative for smoke exposure. At these extreme conditions the data may be biased due to non-linear response of the equipment (Barkjohn et al., 2022).

In the case of BC, the main objective of the comparison was to assess and correct for intra-unit variability, especially due to the fact that 2 different types of portable micro-aethalometers were used. The MA200 units were considered internal reference in this study, and the AE51 units were corrected against them. Regarding the MA200 Aethalometers, inter-comparison with the MAAP instrument yielded r^2 values ranging from 0.86 to 0.89. SD ranged from 1.66 to 1.70 (Fig. S3). Intra-unit comparisons among the MA200 Aethalometers were also carried out, which resulted in an r^2 of 0.99. SD ranged from 1.70 to 1.73 (Fig. S4). As a result, the data from these monitors were not corrected for intra-unit variability. AE51 monitors were inter-compared with one of the MA200 units, obtaining r^2 values ranging between 0.87 and 0.92. SD ranged from 1.68 to 1.75 (Fig. S5). The data from the AE51 monitors were corrected using one of the MA200 as internal standard to ensure comparability across the BC datasets. Subsequently to calibration, the BC datasets were post-processed with ONA, to eliminate the noise in Aethalometer real-time BC data (Hagler et al., 2011).

3.2. BC and $PM_{2.5}$ exposures during wildfires and prescribed burns

3.2.1. $PM_{2.5}$ exposures

Over a 3-year study period, 114 individual $PM_{2.5}$ exposure datasets were collected, covering wildfires and two periods of prescribed burns (PB1 and PB2). The mean $PM_{2.5}$ concentrations across the three scenarios were in a similar order of magnitude, ranging from 110 $PM_{2.5}$ μg/m^3 for PB1 to 152 $PM_{2.5}$ μg/m^2 for wildfires (WF), with no statistically significant differences observed among them (Table 1; note that Table 1 reports the averages for the full dataset, whereas, Figs. 2 and 4 break down the data for each scenario across all tasks for $PM_{2.5}$ and BC. It should also be noted that the time resolution of the $PM_{2.5}$ monitor was 30 s whereas that of the BC monitors was 1 min). As a result, prescribed burns may be considered proxy settings for wildfires, in terms of exposure monitoring. This is especially valuable in terms of experimental research, as it reduces the complexity linked to aerosol monitoring during actual wildfires. The standard deviation of PB2 was higher due to the varying proportion of extinguishing tasks and mop-up tasks recorded.

Fig. 1 shows an example from a prescribed burn at Lloreda, where mop-up tasks were carried out (final extinguishing using handheld tools such as shovels). These tasks generate soil particle resuspension, which contributes to the observed broader peaks in $PM_{2.5}$ concentrations. This is reflected in the higher $PM_{2.5}$ exposures during mop-up tasks (140 $PM_{2.5}$ μg/m^3) compared to those during the initial fire ignition activities (66 $PM_{2.5}$ μg/m^3). On average, mop-up tasks resulted in notable mean $PM_{2.5}$ emissions, with 1-min $PM_{2.5}$ peak exposure concentrations reaching up to 1190 μg/m^3 (Fig. 2), emphasizing the potential health risks associated with post-combustion activities, which may initially seem to be a low-risk task. This result was unexpected and may add value in terms of the need for protective measures during these activities, which are frequently only used during combustion-related tasks (not during post-combustion activities). Thus, this result may be

Table 1
Summary statistics of $PM_{2.5}$ and BC exposure concentrations monitored during wildfires (WF) and prescribed burns (PB). SD: Standard Deviation. N: Number of datasets collected. $PM_{2.5}$ and BC data were not always collected simultaneously; as a result, concentrations are not directly comparable.

		Mean Max	Overall Mean	Mean Median	Mean SD	N
$PM_{2.5}$ (μg/m^3)	WF	798	152	99	161	14
	PB1	666	110	58	129	27
	PB2	1105	145	74	194	73
BC (μg/m^3)	WF	1062	47	18	108	8
	PB1	1671	82	10	221	7
	PB2	1021	49	8	118	52

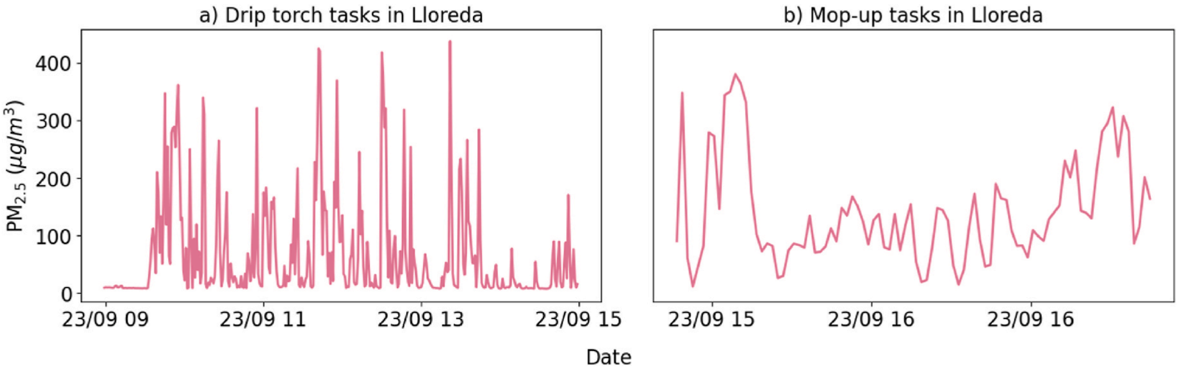

Fig. 1. $PM_{2.5}$ exposure concentrations during a) drip torch fire ignition tasks, and b) mop-up tasks.

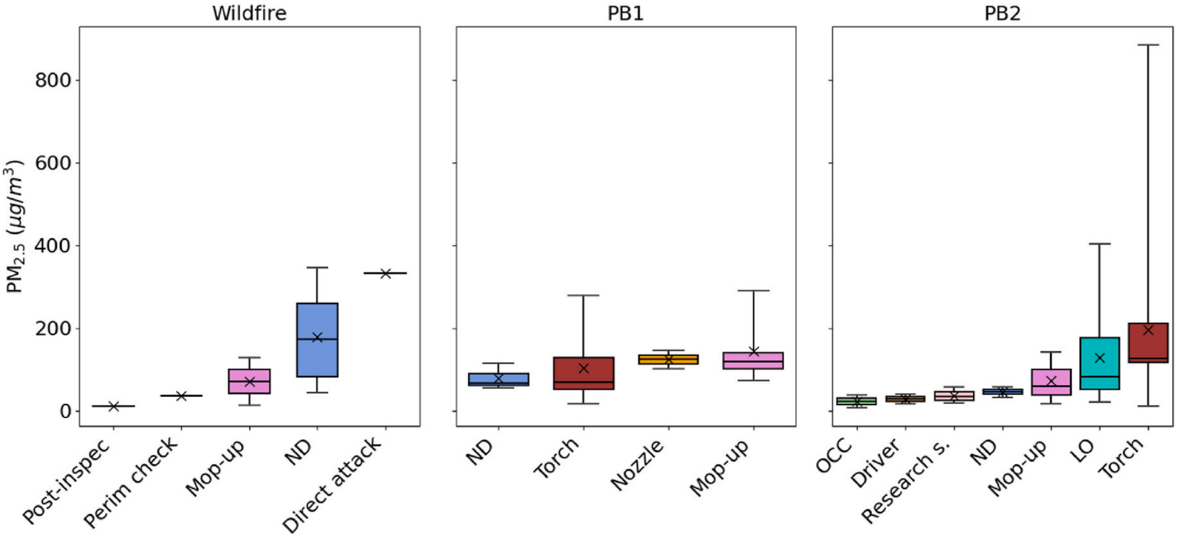

Fig. 2. Mean $PM_{2.5}$ exposure concentrations during wildfires and prescribed burns (PB1 and PB2). Abbreviations are defined as follows: Post-inspec: Post-fire inspection; Perim check: Perimeter inspection, monitoring tasks, control of smoke columns, and hotspot control; Mop-up tasks; ND: No data, Direct attack firefighting; Nozzle: Discharging – nozzle; OCC: Operational command center; Driver: truck driving; Research s.: Research staff; LO: Line operator; Torch: Torchers. Dataset sizes (N) and mean values are available in Table S2.

applicable for a more optimal design of protocols of usage of personal protective equipment, resulting in improved firefighter health.

Other activities reported, which were characterized by lower exposure concentrations, were truck driving (on average, 28 μg/m^3 $PM_{2.5}$), operations at the command center (23 μg/m^3 $PM_{2.5}$), and exposure from research staff (36 μg/m^3 $PM_{2.5}$). However, the size of the datasets for these more specific tasks was smaller than for fire ignition or controlling tasks, which limited our ability to conduct comprehensive statistical analyses. For the main tasks (fire lighting by torchers, and controlling by line operators), there was a higher representation of cases in PB2, with more relevant high exposure concentrations.

During wildfires, exposure concentrations of $PM_{2.5}$ varied considerably across tasks. The lowest mean concentration, 11 μg/m^3, was observed during the post-fire inspection perimeter task. This was followed by perimeter inspection, smoke column monitoring, and hotspot control (36 μg/m^3), and mop-up tasks at 71 μg/m^3. The highest mean concentration was monitored during direct attack firefighting (333 μg/m^3). Additional datasets collected during wildfires, for which task-specific information was not available (ND—no data; Fig. 2), demonstrated a range of concentrations similar to those previously described, with a mean of 179 μg/m^3 (Fig. 2). The variability in $PM_{2.5}$ emissions observed, which impacted personal exposures, is considered valuable for smoke modelling studies as input regarding the source term, which is typically complex to characterize experimentally. Such characterization may provide especially added value for climate models.

While the mean $PM_{2.5}$ exposure concentrations were relatively similar between wildfires and prescribed burns (Table 1) prescribed burns exhibited higher peak values, reaching absolute maximum concentrations of 2526 μg/m^3 $PM_{2.5}$, compared to wildfires (2170 μg/m^3 $PM_{2.5}$), albeit being short-term peaks (Fig. 3a). While the peak concentrations may be higher during prescribed burns, the brief nature of these spikes leads to a lower overall exposure. In contrast, wildfires showed slightly lower but more sustained concentrations, leading to overall higher exposure concentrations over time (Fig. 3b) due to the accumulated dose. This accumulated dose refers to the total amount of contaminants that firefighters are exposed to during their shifts, highlighting the potential need for specialized shift rotations to reduce prolonged exposure, optimize recovery periods, and minimize health risks.

The obtained results of the present study were compared to those in the literature, despite the complexity of this task, due to the intrinsic variability among wildfires and prescribed burns. Examples of $PM_{2.5}$ exposures were reported by Adetona et al. (2011), who investigated the exposure to smoke of firefighters at prescribed forest burns in a south-eastern U.S. forest. Mean $PM_{2.5}$ exposures ranged between 389 and 535 μg/m^3, which were comparable even though higher than our results (129–165 μg/m^3, as average exposures). These authors also found that the exposures of torchers and line operators were similar for $PM_{2.5}$, as is

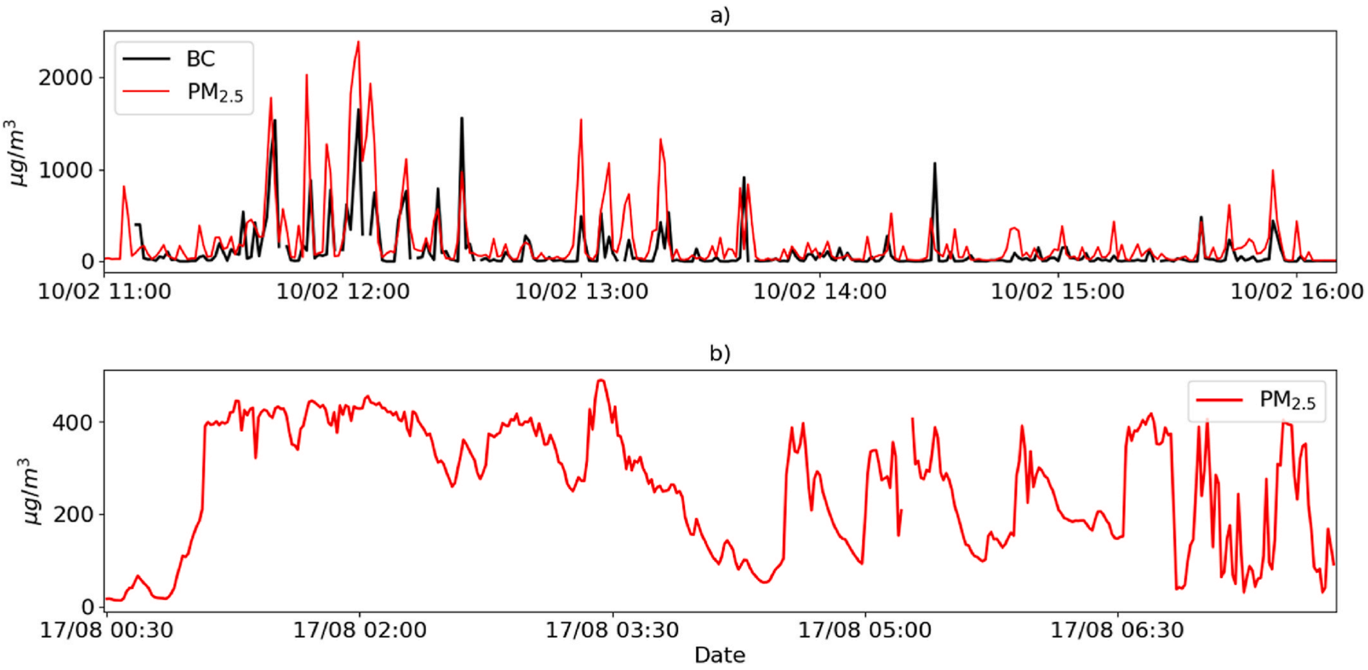

Fig. 3. Top: $PM_{2.5}$ and BC exposure concentrations during a prescribed burn in Can Riera Puig-reig. Bottom: $PM_{2.5}$ concentrations during the Begís wildfire.

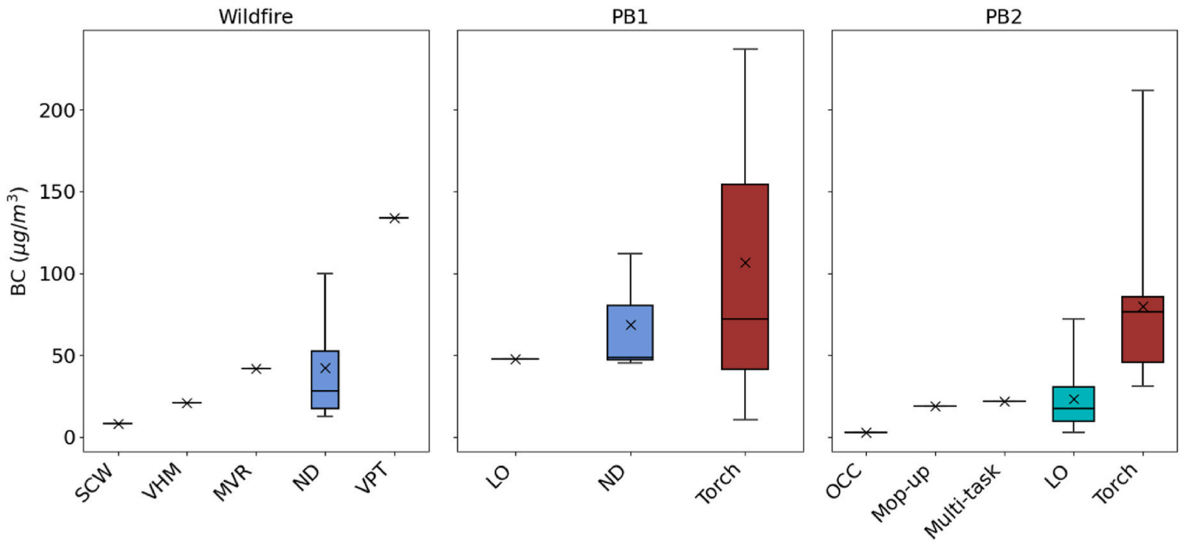

Fig. 4. Mean BC exposure concentrations of other firefighting activities. Abbreviations are defined as follows: SCW: Saw Crew Work; VHM: Vehicle Mobility and Driver Tasks; MVR: Movement with Vehicle and Chainsaw Operations; ND: No Data; VPT: Vehicle Patrol and Burn Operations; LO: Line Operator; Mop-up: Mop-up-tasks; Torch: Torcher; OCC: Operational command center, Multi-task: Multi firefighting task. Dataset sizes (N) and mean values are available in Table S3.

the case in the present work. Wu et al. (2021) conducted a study during prescribed burns in midwestern region of U.S. forests, where the mean $PM_{2.5}$ exposure was 1750 ± 1200 $\mu g/m^3$. Miranda et al. (2010) conducted the first measurements and analysis of firefighter individual exposures to toxic gases and particles in fire smoke experiments in Central Portugal, reporting 1-min peak $PM_{2.5}$ concentrations >18,000 $\mu g/m^3$, which is significantly higher than those observed in the present study. However, the burn durations were relatively short (10–15 min) compared to our study. When averaged over an 8-h exposure duration, concentrations ranged from 551 to 2187 $\mu g/m^3$, which is more comparable to our findings.

3.2.2. *BC exposures*

A total of 67 individual BC exposure records were collected throughout the sampling period (Table 1). The mean BC concentrations ranged from 47 $\mu g/m^3$ for WF to 82 $\mu g/m^3$ for PB1, without statistically significant differences (Table 1). PB1 exhibited higher mean BC concentrations than wildfires, which could be a first indicator of the influence of the use of the drip torch fueled by a diesel-gasoline mixture during prescribed burns. Concentrations between PB2 and wildfires were comparable, however, peak BC concentrations rose to an absolute maximum concentration of 4521 $\mu g/m^3$ during prescribed burns, which was 1.4 times higher compared to wildfires. Although these results were observed under controlled conditions, they underscore the importance of developing adequate respiratory protections for firefighters during prescribed burns, as these high peak concentrations pose significant health risks. Since prescribed burns involve fire that is controlled and less intense compared to wildland fires, firefighters often spend more time closer to the active fire and can be exposed to more smoke. Whereas these differences might indicate higher peak exposures during prescribed burns, on the other hand it is important to highlight that only 8 records were available for wildfires, and therefore these differences should be taken with caution, due to the limited number of wildfire data in this study, which is linked to the complexity of data collection during actual wildfires (the spontaneity of their occurrence, the need for immediate action, limited reaction time, and extreme conditions). Regarding BC exposures based on tasks (Fig. 4), a relatively high variability was observed (e.g., vehicle patrol and burn operations, 134 μg/m^3 BC, vs. movement with vehicle and chainsaw operations, 42 μg/m^3 BC). It should be noted that the concentrations of $PM_{2.5}$ and BC shown in Figs. 2 and 4 were monitored on different days and different times, and therefore they do not correspond to the same specific events. As a result, the tasks represented in these figures are not directly comparable.

As indicated for $PM_{2.5}$, the BC concentrations reported may be valuable to characterize the source term in smoke transport modelling studies. However, the sample size for wildfires was not large enough to confirm the significance of these findings. In the case of prescribed burns, essentially PB2, highest BC concentrations were observed in torchers, followed by line operators. In former studies, Wu et al. (2021) reported mean BC exposures of 72.3 ± 48.8 μg/m^3, which are comparable to the concentrations reported in the present work. They also concluded that torchers were associated with higher BC exposures than line operators.

3.2.3. Source apportionment

Source apportionment models were applied to the BC datasets as a function of the two main BC exposure profiles: torchers and line operators. As described in the Methods section, two different source apportionment methods (Delta-C and the Aethalometer model, Sandradewi et al., 2008b) were applied. To this end, an Ångström exponent of 1 was used for fossil fuel-derived BC (AAE_{ff}), and a range of Ångström exponents was tested for biomass burning aerosols (AAE_{bb}, ranging from 2 to 3) (Martinsson et al., 2017; Massabò et al., 2015). According to literature, a number of authors (Roden et al., 2006; Day et al., 2006, Garg et al., 2015; Martinsson et al., 2017) used AAE_{bb} between 1.3 and 1.9, whereas others used AAE_{bb} between 2.1 and 2.8 (Clarke et al., 2007; Sandradewi et al., 2008b; Massabò et al., 2015; Kirchstetter et al., 2004).

Figs. S6 and S7, and Table S4 show the sensitivity analysis implemented using a range of AAE_{bb} exponents, which in addition to the comparison with the Delta-C model led to the selection of 2.4 as the most convincing exponent for the fresh combustion aerosols modelled in this work. As shown in Table S4, lower AAE_{bb} exponents such as 2 or 2.2 resulted in a significant overestimation of BC_{bb} (101.0%–133.7% of total BC), whereas high exponents, e.g. $AAE_{bb} = 3$ overestimated the contribution from BC_{ff} for the line operators (with up to 60% of BC_{ff}, which was deemed unrealistic). The starting hypothesis was that the majority of BC that line operators are exposed to originates from biomass burning, whereas torchers' exposure to BC_{ff} should be significantly higher in comparison. Additionally, these results were compared with those from the Delta-C method, used as benchmark (Fig. S8), which evidenced a good agreement between models when using $AAE_{bb} = 2.4$.

It should be noted, however, that both models were designed and are typically applied to ambient aerosols, whereas in the present study the monitors were exposed to freshly emitted aerosols (frequently at < 2 m distance from the monitoring instrumentation). In addition, BC concentrations were higher (due to proximity to the source) than typically reported in studies applying these source apportionment models. A primary concern is that the close proximity to the emission sources impacts loading compensation on the filter. In the MA200, the aerosol loading on the filter is non-negligible during the period after a tape advance (Virkkula et al., 2007), potentially causing larger underreporting in the UV channel than in the IR channel. Moreover, aerosols have varying combustion efficiencies, and additional factors such as heat and moisture, smoldering versus high-efficiency burning, different fuel mixtures, are variables not accounted for in the model (Sandradewi et al., 2008c).

Table 2 reports $PM_{2.5}$, and the calculated BC_{bb} and BC_{ff} exposure concentrations obtained from the Aethalometer model, separately for line operators and torchers. Results evidenced that, on average, exposures to $PM_{2.5}$ concentrations were comparable for line operators (129 μg/m^3) and torchers (165 μg/m^3), and the same was true for peak concentrations. However, the composition of the $PM_{2.5}$ aerosols was largely different for both types of activity: 62% of $PM_{2.5}$ was made up of BC in the case of torchers, while this was 22% for line operators. This result suggests a major contribution for different types of aerosols in the case of line operators, such mineral aerosols sourcing from soil resuspension during general activities and especially during mop-up tasks, as well as higher organic carbon contributions in biomass burning.

Part of the differences in BC exposure could be related to the different tasks and potential exposure to source emissions, such as biomass burning and fossil fuel combustion. Subsequently, the results obtained from the Aethalometer model calculations allows to study the relative contribution to total BC concentrations from biomass burning (BC_{bb}) and fossil fuel combustion (BC_{ff}). The results in Table 2 indicate that line operators were exposed to 61% of BC_{bb} and 39% of BC_{ff}, as opposed to torchers for whom exposure to BC_{ff} was dominant (77%, vs. 23% BC_{bb}). This suggests that the use of drip torches drives exposure to BC from fossil fuel combustion. More specifically, the exposure concentrations of BC_{ff} for torchers were 8 times higher than those for line operators (p-value <0.001), indicating that torchers were heavily influenced by the use of drip torches to ignite fires. Regarding $PM_{2.5}$, the ratio between torchers and line operators was only 1.3, which was expected as line operators are more exposed to organic carbon containing biomass burning smoke and possibly soil dust particles. This later exposure is influenced by resuspension of particles caused by perimeter monitoring activities, where manual tools are used to prevent the fires from spreading, leading to the generation of suspended dust.

Finally, the Aethalometer model also allowed for the comparison between fossil fuel and biomass combustion contributions to BC during wildfires and prescribed burns. As expected, the primary contributor to BC during the wildfires monitored was biomass combustion, accounting for 60% of total BC. However, there was a relatively high contribution of fossil fuel emissions, likely sourcing from other sources such as drip torch, trucks and other machinery used in the area. In prescribed burns (PB1 and PB2), the primary contributor to BC was fossil fuel emissions, with 54% attributed to BC_{ff}, due to the influence of drip torches on BC exposures. This distinction between the sources of BC in wildfires and prescribed burns highlights the different factors at play in fire management and the aerosol emission profile of controlled prescribed burns versus uncontrolled wildfire events.

Table 2

$PM_{2.5}$, BC_{bb} and BC_{ff} exposure concentrations in prescribed burns for line operators and torchers. BCtot: total BC. N: Number of datasets collected. SD: Standard Deviation.

		Max	Mean	%BC/BCtot[a]	SD	N
Line operator	$PM_{2.5}$ (μg/m^3)	1139	129		196	26
	BC_{ff} (μg/m^3)	394	12	39%	40	16
	BC_{bb} (μg/m^3)	269	19	61%	42	16
Torcher	$PM_{2.5}$ (μg/m^3)	1145	165		207	51
	BC_{ff} (μg/m^3)	1554	95	77%	206	10
	BC_{bb} (μg/m^3)	538	20	23%	61	10

[a] Ratios calculated for each individual exposure dataset and subsequently averaged for the instances where both $PM_{2.5}$ and BC data were measured simultaneously (N = 11 for LO and N = 5 for T).

4. Health implications and conclusions

This work aimed to increase our understanding of the types and sources of aerosols generated during wildfires and prescribed burns, which is crucial for exposure management and mitigation. To this end, firefighters' exposure to PM$_{2.5}$ and BC aerosols was monitored during wildfires and prescribed burns. PM$_{2.5}$ and BC concentrations were substantial and comparable during prescribed burns and wildfires in a Mediterranean environment, suggesting that prescribed burns may be considered proxy settings for wildfires in terms of exposure monitoring. This is especially valuable in terms of experimental and modelling research, as it reduces the complexity linked to aerosol monitoring during wildfire events. Nevertheless, peak PM$_{2.5}$ concentrations were higher during prescribed burns, while wildfires ultimately resulted in greater overall PM$_{2.5}$ doses, due to the longer duration of exposures. In terms of BC, peak 1-min concentrations were relatively higher during prescribed burns when compared to wildfires.

Considering the different firefighting tasks, mop-up tasks were identified as an important contributor to high PM$_{2.5}$ exposure, although torchers and line operators were exposed to higher peak concentrations. BC was about 62% of PM$_{2.5}$ in the case of torchers, while this was 22% for line operators. This result suggests exposure contributions for different types of aerosols among the tasks, probably related to resuspension of mineral aerosols from soil during mop-up tasks as well as different compositions of smoke from biomass burning (bb) and fossil fuel combustion. Source apportionment of BC aerosols provided deeper insights into the fire smoke exposure of line operator and torcher. Line operators were exposed to 61% of BC_{bb} and 39% of BC_{ff}, as opposed to torchers for whom exposure to BC_{ff} was dominant (77%, vs. 23% BC_{bb}). This suggests that the use of drip torches drives exposure to BC during specific firefighting activities, and should thus be assessed in terms of exposure risk and hazard potential (Aguilera et al., 2021). Former studies showed higher pro-inflammatory responses in torcher compared to line operators after performing their tasks which could be related to enhanced exposure from fossil fuel combustion products in combination with biomass burning smoke (Adetona et al., 2017), however the relationship of the implication of toxic compounds and toxicity mechanisms is not conclusive, especially in terms of long-term effect related to firefighting occupation, such as respiratory diseases, cardiovascular diseases, systemic inflammations, and cancers (Navarro, 2020, and references therein). Although the exposure of firefighters to smoke has been observed through lung function decline and the detected of biomarkers in urine and cells, epidemiological studies often show no difference in long-term exposure effects in firefighters compared to the general population. Nevertheless, model calculations estimated that wildland firefighters that were exposed to an average PM$_{2.5}$ concentration of 510 $\mu g/m^3$ during their fire extinction tasks had increased risks of 8%–43% for lung cancer mortality and 16%–30% for cardiovascular diseases (Navarro et al., 2019). The results in the present study showed PM$_{2.5}$ levels around 150 $\mu g/m^3$, so is not unlikely that these concentrations can provoke acute and chronic effects. The International Agency for Research on Cancer (IARC) recently classified occupational exposure as a firefighter as "carcinogenic to humans" (Group 1) based on "sufficient" evidence for mesothelioma and bladder cancer in humans. Meta-analysis of epidemiological studies estimated a 58% higher risk for mesothelioma and a 16% higher risk for bladder cancer among firefighters compared to the general populations, which are most probably linked to exposure of firefighters to PM air pollutants, such as asbestos particles, and soot particle related polycyclic aromatic hydrocarbons (IARC, 2023; Demers et al., 2022). The present study showed high exposure levels of BC, which could evidence the presence of high levels of these toxic compounds. Therefore, further studies are necessary to monitor the occupational exposure of firefighters to type of chemicals. This study highlights the reliability of portable monitors and their value in the assessment of wildfire emissions and impacts on human exposure. The use of multi-wavelength aethalometers provides special added value, as it enables the application of source apportionment methods to identify and quantify emission source contributions. The combination of these tools, reporting data in real-time and with high time-resolution, is key to the design and implementation of effective mitigation strategies for environmental and health concerns related to wildland fires.

CRediT authorship contribution statement

Jordina Gili: Writing – original draft, Visualization, Software, Resources, Methodology, Investigation, Formal analysis, Data curation. **Aina Maín:** Software, Resources, Methodology, Formal analysis. **Barend L. van Drooge:** Writing – review & editing, Supervision, Methodology, Investigation, Conceptualization. **Mar Viana:** Writing – review & editing, Supervision, Resources, Methodology, Investigation, Conceptualization.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2025.125660.

Data availability

Data will be made available on request.

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