



Emerging contaminants related to the occurrence of forest fires in the Spanish Mediterranean



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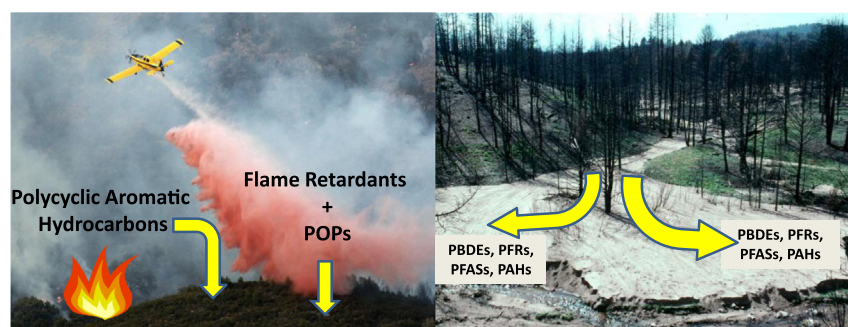
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HIGHLIGHTS

- Assessment of PBDEs, PAHs, PFRs and PFASs distribution in burned and unburned hillslopes
- BDE-85 concentrations were highest so no PBDE mixture was present in the fire extinguisher.
- The fire added significant PAH amounts into the soil (1256 ng g^{-1}), mainly in the upper 2 cm.
- PFRs and PFASs were found in both hillslopes with values up to 352 ng g^{-1} and 17 ng g^{-1} , respectively.
- Contaminants transport downslope in the erosion events depends on the compound nature.

GRAPHICAL ABSTRACT



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ABSTRACT

Forest fires can be a source of contamination because, among others, of the use of chemicals to their extinction (flame retardants, FRs), or by the production of Polycyclic Aromatic Hydrocarbons (PAHs) derived from high temperature alteration of organic matter. Up to our knowledge, this study is the first to assess the direct (PAHs 16 on the USA EPA's priority list), and indirect [tri- to hepta- brominated diphenyl ethers (PBDEs), organophosphorus flame retardants (PFRs) and perfluoroalkyl substances (PFASs)] contamination related to forest fires. The abundance and distribution of these contaminants were monitored on two Mediterranean hillslopes, one burned and one unburned, near Azuébar (SE Spain). Samples were taken in the foot, middle, and top of the slope, at two depths, and in two environments (under canopy and bare soil). Sediments were collected from sediment fences after erosive rainfall events. Most of the screened compounds were found in both, burned and control hillslopes, though significant differences were found between both. In burned soil, low concentrations of PBDEs (maximum Σ 16 PBDEs: 7.3 ng g^{-1}), PFRs (664.4 ng g^{-1}) and PFASs (56.4 ng g^{-1}) were detected in relation to PAHs (Σ 16 PAHs = 1255.3 ng g^{-1}). No significant influence of the hillslope position was observed for any of the contaminants but differences based on depth and vegetation presence tended to be significant, particularly for the PAHs. After the first erosive event, concentrations of PBDEs and PAHs were higher in sediment than in soil (Σ 16 PBDEs: 17.8 ng g^{-1} and Σ 16 PAHs = 3154.2 ng g^{-1}) pointing out the importance of connectivity processes, especially shortly after fire.

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1. Introduction

According to the International Panel on Climate Change, there is already a high degree of confidence that meteorological conditions associated to climate change will be propitious to increasing extreme events (IPCC, 2014). Impacts on land degradation will be manifested in bigger and more frequent wildfires, and greater water stress. Since 1982, the total area affected by forest fires in Mediterranean countries of the European Union (EU) has declined, and the number of fires tends to stabilize. However, in countries like Spain and Portugal, the trend has been slightly upward in the last 5 years (European Commission, 2015), and forest fires are becoming more frequent (Spano et al., 2014; Turco et al., 2016). Based on Spanish Environment Ministry reports (MAAM 2012, 2014), the mean number of fires in Spain during 2001–2010 was 17,127, burning an average of 113,848 ha of forest surface. In Valencian Community (located in the Mediterranean East of Spain), the average area burned in 1991–2010 was of 8706 ha yr⁻¹ but in 1994, 751 fires occurred, one of which burned >25,000 ha. In 2014, there were 92 wildfires in Valencia affecting 1800 ha of woody forest and 116 ha of forest herbaceous.

It is well known that environmental degradation processes are inter-related. One aspect of fire-induced ecosystem degradation that has drawn scientific interest recently is the chemical pollution related to forest fires. This contamination is associated either to the use of chemicals to their extinction known as flame retardants (FRs) (Pepper et al., 2011), or because pyrolysis or incomplete combustion of litter and standing vegetation favours the formation of toxic compounds as polycyclic aromatic hydrocarbons (PAHs) (Yuan et al., 2008). FRs are not only used to extinguish fires but also in the production process of electronics, furniture, clothes and cars (Boyles et al., 2017; Covaci et al., 2003, 2011). Perfluoroalkyl substances (PFASs) are widely used in aqueous film-forming foams (AFFF) but also in industrial and consumer products as protective coatings for textiles and paper, in the production of semiconductors and as polymer additives in herbicide and insecticide formulations and in cosmetics (Hale et al., 2017; Hu et al., 2016). AFFFs are usually spread over pool fires and they have limited post-fire security and are toxic groundwater contaminants (Hinnant et al., 2017). In general, AFFFs containing PFASs are not expected to be used in the extinction of forest fires. They are usually applied for suppression of combustion in industrial and commercial sites, and particularly in fire training areas (Hu et al., 2016). However, the possible presence and degradation of PFASs in relation to forest fires have been never measured. These compounds are persistent, bio-accumulative and/or toxic to wildlife and humans, as well as potential endocrine disruptors (Campo et al., 2016; Duan et al., 2015; Eulaers et al., 2014; Segev et al., 2009). Consequently, penta-BDEs, octa-BDEs, perfluorooctane sulfonate (PFOS) and perfluorooctyl sulfonyl fluoride (POSF) have been included as Persistent Organic Pollutants in the Stockholm Convention, and PAHs are candidates for their inclusion (UNEP, 2010). Additionally, in the EU to comply with the Water Framework Directive (WFD), Directive 2013/39/EU set the Environmental Quality Standards in the field of water policy for Priority Substances and other pollutants, including PAHs, which must be controlled to progressively reduce discharges and losses.

Brominated flame retardants (BFRs) account for a large group of FRs used for firefighting. Polybrominated diphenyl ethers (PBDEs) are the second highest production group of BFRs, and their presence has been reported in different environmental matrixes as water (Wang et al., 2017; Ricklund et al., 2010); sediment (Ross et al., 2009); soil (Akortia et al., 2017) and biota (Eulaers et al., 2014; Boyles et al., 2017). In soils and sediments, photolytic degradation and debromination are possible (Segev et al., 2009; Lee and He, 2010). Debromination can be caused by microbial activity and can be harmful for the environment because lower brominated PBDEs are considered to be more toxic (Rodenburg et al., 2014). The sources of PBDEs contamination are leaching from a wide range of plastics, electronic equipment and textiles (Akortia et al., 2017; Covaci et al., 2011) or their incineration and subsequent

long-range transport in air. de Wit et al. (2006) reported their presence in living organisms and air of the Arctic. BDE-47, -99 and -209 are also present in sewage sludge (de Wit, 2002; Wu et al., 2017). The production and use of the most common PBDEs, penta- octa- and deca-BDE, is nowadays highly restricted in the USA and EU (Kemmlin et al., 2009). Despite PBDEs have been used for fire extinguishing (Alaee et al., 2003; Pepper et al., 2011) there are not published studies on their occurrence in relation to forest fires.

Since the ban on some BFRs, phosphorus flame retardants (PFRs) have been proposed as an alternative (van der Veen and de Boer, 2012). PFRs are widely used as plasticizers and anti-foaming agents in a variety of industries including plastics, furniture, textile, electronics, construction, vehicles and petroleum industries (Wei et al., 2015). Only few reports on possible PFR adverse effects have been published (Araki et al., 2014; Dishaw et al., 2011; Farhat et al., 2014; Kojima et al., 2013; Sun et al., 2016; Wang et al., 2015). Most of the recent research priorities have focused on the occurrence of PFRs in house dust, indoor and outdoor air, surface and ground waters, but data about their presence in soil and sediment after a forest fire are not available.

A small number of studies have already looked into PAH production in forest fires (Kim et al., 2003; Campo et al., 2011; Vergnoux et al., 2011; Choi, 2014). These studies have generally found a significant increase in soil PAH levels after fire, but, not so much to reach harmful levels. However, the increase could still affect local ecosystems (Pizarro-Tobías et al., 2015). In a fire, PAHs are formed by incomplete combustion of litter and standing vegetation (Kim et al., 2003), but they are also probably produced from soil organic matter (SOM). During fire only the upper few centimetres of soil could reach temperatures above 200 °C, at which formation of char and possibly PAHs starts (González-Pérez et al., 2004; Certini, 2005). Incorporation of partly burned organic material into the soil increases both its SOM (Campo et al., 2008) and PAH concentrations (Choi, 2014).

The produced PAHs are either volatilized or retained within the organic material from which they are formed. The volatilized PAHs may also become associated with organic material, as they easily adsorb onto litter, vegetation or floating ash particles (Kim et al., 2003; Choi, 2014). These organic materials are deposited on the surface forming a PAH-rich fire-litter layer (Johnsen and Karlson, 2007) and can become incorporated in the soil, increasing soil PAH levels. PAHs can also enter the soil by moving downwards in gaseous form, or by leaching from the fire-litter layer (Vergnoux et al., 2011; Choi, 2014). Several researchers have noted that most of the PAHs added with fire are removed from a location because the fire-litter, rich in both SOM and PAHs, is eroded downslope (Smith et al., 2011; Zheng et al., 2012; Luo et al., 2013). This eroded material would be transported to the streams as researched by Vila-Escalé et al. (2007), who stated that PAHs were more correlated with organic suspended substances than with total suspended substances.

The main of this study is to establish the impact of a high severity fire in a hillslope of Azuébar, Castellón (Spain) on the occurrence of PAHs and emerging POPs in soil and sediment from the burned area. Samples from coupled slopes (burned and unburned) were analysed and compared for PAHs, PBDEs, PFASs and PFRs concentrations. Furthermore, the specific objectives of the study are to (i) determine whether the fire occurred, in 2014, in Azuébar added significant amounts of PAHs and emerging POPs to the soil, (ii) establish possible relationships between concentrations and position on the hillslope, soil depth and presence of vegetation, and (iii) study the transport of the contaminants downslope. To our knowledge, this is the first report on the presence of flame retardants in burned soils.

2. Material and methods

2.1. Study site and sampling

This work was carried out in the municipality of Azuébar, Natural Park of Sierra de Espadán, in the Province of Castellón, Spain (Fig. 1).

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