



# Effects of wildfire on mercury mobilisation in eucalypt and pine forests



Isabel Campos<sup>a,b</sup>, Carlos Vale<sup>b,c</sup>, Nelson Abrantes<sup>a,\*</sup>, Jan Jacob Keizer<sup>a</sup>, Patrícia Pereira<sup>b,d</sup>

<sup>a</sup> Department of Environment and CESAM, University of Aveiro, 3810-193 Aveiro, Portugal

<sup>b</sup> IPMA – Portuguese Institute for the Sea and Atmosphere, Av. Brasília, 1449-006 Lisboa, Portugal

<sup>c</sup> CIIMAR, Rua dos Bragas, 4050-123 Porto, Portugal

<sup>d</sup> Department of Biology and CESAM, University of Aveiro, 3810-193 Aveiro, Portugal

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## ABSTRACT

Wildfires and subsequent rainfall play an important role in the redistribution of major, minor and trace chemical elements, which could be mobilised from burnt soils and ashes. In particular, the fire-induced release of mercury (Hg) into the environment is relevant to study due to its volatilisation and toxicity. However, the impact of wildfire followed by rainfall on mercury redistribution is poorly documented. Hence, eucalypt- and pine-covered hillslopes (1–3) were surveyed in two burnt areas (Ermida and S. Pedro do Sul, Portugal), as well as in a non-burnt eucalypt slope (Ermida). Top-soils (0–2 cm) and ashes were sampled 4 and 14 weeks after the fire, the latter following an episode of heavy precipitation. In order to clarify the effect of temperature on Hg release from the soil, a heating experiment was also conducted. Results revealed that 30% of the Hg retained in eucalypt soils was released by the fire, corresponding to a loss of 1.0–1.1 g Hg ha<sup>-1</sup> of burnt soil. Levels in burnt eucalypt soils were twice the values registered in burnt pine soils for both fire severities. Mercury in ashes pointed to similar trend between the two types of vegetation. Results from the heating experiment with soil suggest that temperature reached in the wildfire was insufficient to destroy the most stable bonds of Hg-ligands. Mercury concentrations were not correlated with organic matter content, both in soils and ashes. It was estimated that rainfall caused a loss of 1.0 g Hg ha<sup>-1</sup> from ashes and an uptake of 0.5 g Hg ha<sup>-1</sup> of soil washed out. The difference between the previous estimations, 0.5 g Hg ha<sup>-1</sup>, corresponds to the quantity of Hg transported to the surrounding area or eventually introduced into aquatic systems. Hence, the results obtained in the present study highlight the importance of wildfire and subsequent rainfall in the mobilisation of Hg in the environment.

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## 1. Introduction

Forest ecosystems undergo recurrent wildfires with serious impacts resulting from the destruction of vegetation coverage and severe changes in the properties of soil surface layers (e.g. Cerdà and Lasanta, 2005; Verma and Jayakumar, 2012; Rubenacker et al., 2012). It is well known that the magnitude of these changes depends mainly on the nature of the soil, type and biomass of the vegetation coverage, fire severity, ash production, and post-fire climate conditions such as timing and intensity of precipitation events (e.g. Ulery et al., 1993; Neary et al., 1999; Certini, 2005; Maia et al., 2012; Verma and Jayakumar, 2012). Wildfire can have profound effects on the functioning of the soil system (physical, chemical and biological elements), which can affect the sustained productivity and soil recovery of the burnt area (Certini, 2005; Doerr and Cerdà, 2005; Terefe et al., 2008; Verma and Jayakumar, 2012; Guénou et al., 2013). Both the direct effects of fire and also the overall changes to the ecosystem encountered in post-fire situation can lead to short-, medium- and long-term changes in the soil. One of the most intuitive and important soil changes, during

burning is the alteration of organic matter content (Certini, 2005). Organic matter (LOI) is a key factor for forest soil since it has direct and/or indirect influences on the physical, chemical and biological characteristics of the soil. Fire affects soil organic matter both quantitatively and qualitatively and its changes may range from its almost total destruction to increased contents in surface layers due to external inputs, as partly charred litter and leaves, forest necromass and residual ash (González-Pérez et al., 2004; Knicker et al., 2005; Certini, 2005; Mataix-Solera et al., 2011). Among the effects of wildfires, it is important to recognize and underline its impact on the chemical availability of trace elements and pathways (Navrátil et al., 2009; Jakubus et al., 2010; Aref et al., 2011). In fact, wildfires can release deposited metals on soil surface, either directly by combustion of vegetation and soil organic matter mineralization and leaching from ash–soil interactions (Ulery et al., 1993; Antilén et al., 2006; Jakubus et al., 2010; Pereira and Úbeda, 2010). Although ash produced by forest fires is a very heterogeneous material (depending strongly on plant species, type and part burnt, and combustion completeness), it primarily consists of oxides and hydroxides of base cations such as calcium, magnesium and potassium, but also includes trace metals (Ulery et al., 1993; Pereira et al., 2011a; Bodi et al., 2014). These compounds can be transported by overland flow and leached into the soil profile, and may have impacts

\* Corresponding author.

E-mail address: [njabrantes@ua.pt](mailto:njabrantes@ua.pt) (N. Abrantes).

on surface and groundwater chemical composition (Reneau et al., 2007; Cerdà and Doerr, 2008; Smith et al., 2011). Research studies on ash resulting from wildfires have been mainly focused on soil erosion, physical and hydrological properties of the ash (e.g. Doerr and Cerdà, 2005; Quintana et al., 2007; Cerdà and Doerr, 2008; Balfour and Woods, 2013; Gabet and Bookter, 2011; Pereira et al., 2015). The chemical composition of wildfire ash has received comparatively little research attention (Ulery et al., 1993; Pereira et al., 2011b, 2012; Santín et al., 2012), especially in what concerns its trace metal constituents (Engle et al., 2006; Pereira and Úbeda, 2010; Pereira et al., 2011a).

The toxic element mercury (Hg) is of particular concern due to its volatility, toxicity, persistence and the relevance of the atmospheric compartment in the global Hg cycle (Cheng and Schroeder, 2000; Huang et al., 2011), showing a potentially harmful impact on the environment and on human health (Alloway, 1995; Cheng and Schroeder, 2000; Huang et al., 2011). Water, soil and vegetation take up Hg species following wet and dry deposition, being the uptake by vegetation an important pathway by which metals enter the food chain (Friedli et al., 2001; Roy and McDonald, 2013). Once deposited, Hg may undergo methylation by biological processes into the highly toxic methyl mercury, a potent neurotoxin with a strong tendency to bioaccumulate and then biomagnify within the food chain (Boening, 2000; Caldwell et al., 2000; Gnamus et al., 2000; Amirbahman et al., 2004). Various studies have quantified anthropogenic emissions of Hg (e.g. Pacyna et al., 2003; Pacyna and Pacyna, 2002). However, natural emissions, particularly associated with wildfires that have been accounted as one of the important Hg non-point sources, are less well documented (Sigler et al., 2003; Biswas et al., 2008; Navrátil et al., 2009; Wiedinmyer and Friedli, 2007). Forest soils accumulate Hg that is strongly sorbed to organic matter (Schwesig and Matzner, 2000; Biester et al., 2002; Grigal, 2003). The behaviour of Hg in soils is very complex and is mainly controlled by adsorption and desorption process, involving several chemical, physical and biological processes (Reis et al., 2015). Its environmental and toxicological effects are however, highly dependent on Hg forms, as different species interact differently with soil constituents, exhibiting different behaviours, solubilities, mobilities and bioavailabilities (Biester et al., 2002; Bollen et al., 2008). Therefore, the identification of mercury species in soils contributes to understand the Hg behaviour and fate in the environment and its toxicity (Biester and Scholz, 1997; Rumayor et al., 2013). Methods based on species release from the matrix according to their desorption temperatures have been used to determine the speciation of Hg species in solid samples, including thermo-desorption (Biester et al., 2002; Rumayor et al., 2013, 2015; Reis et al., 2012, 2015).

During wildfires, Hg stored in vegetation, organic litter and soil tends to be associated with organic-rich ash and is released to the atmosphere predominantly in the form of gaseous elemental form ( $\text{Hg}^0$ ), with up to 15% in the form of particulate mercury. Mercury released from fires can have local, regional and global impacts, since  $\text{Hg}^0$  can be readily transported to significant distances from the original source, whereas particulate Hg is likely to be deposited locally (Friedli et al., 2003a, 2003b; Sigler et al., 2003; Wiedinmyer and Friedli, 2007). The amount of Hg released during a wildfire is limited by Hg accumulation in the ecosystem, particularly in the soil, prior to burning (Biswas et al., 2007, 2008). Large Hg losses result from the low volatilization temperatures (100–300 °C) of Hg species. For example, during some wildfires, elevated soil temperatures are sufficient to decompose Hg-humic acid bond (200–330 °C; Biester and Scholz, 1997). Experimental burns performed on soils showed a 79% decrease in Hg content after burning (Mailman and Bodaly, 2005). Recent studies have reported Hg release from soils burned during both prescribe fires (Woodruff et al., 2001; Harden et al., 2004; Engle et al., 2006) and wildfires (Biswas et al., 2007, 2008; Engle et al., 2006; Navrátil et al., 2009; Burke et al., 2010).

Fires, partitioning of Hg in soil and biomass, influence Hg pathways (Biswas et al., 2007, 2008; Witt et al., 2009). Additionally, runoff and wind can mobilise Hg bound to eroded soil, litter and ash particles,

leading eventually to the transport of Hg to aquatic systems (Caldwell et al., 2000; Amirbahman et al., 2004). Engle et al. (2006) measured higher concentrations of Hg in surface ash one year following both wildfire and prescribed burns, and suggested that it was related to the sorption of atmospheric Hg over the subsequent year, reflecting Hg's affinity for carbon in ash. However, very few studies have reported such effects on Hg pathways in burnt soils and ashes. Therefore, there is a need to better understand the terrestrial Hg cycle and, in particular, to investigate wildfires as a potential non-point source of Hg. Understanding this is important to predict the fate of atmospherically derived Hg in terrestrial ecosystems with important implications for the loading of Hg to water bodies (Obriest et al., 2009), which is reinforced by the knowledge about solute released due to the runoff and erosion, after wildfires (Lasanta and Cerdà, 2005).

Hence, this work aims to: (a) clarify the effect of a wildfire on mercury mobilisation from soils and ashes; (b) assess the influence of rainfall on the mercury pathway; and (c) evaluate the differences in Hg concentrations in soils and ashes between two vegetation types. For this purpose, soil and ashes were surveyed from eucalypt and pine forests burnt at different severities, as well as from a non-burnt eucalypt stand. Furthermore, in order to clarify the effect of temperature on Hg release a thermo-desorption assay of Hg from soils was performed in the laboratory.

## 2. Material and methods

### 2.1. Characteristics of the study areas

Two recently burnt areas located in north-central Portugal: Ermida (E) (40° 43' 33" N, 8° 20' 60" W, 170 m a.s.l., Aveiro district) and S. Pedro do Sul (S) (40° 45' 35" N, 8° 10' 04" W, 370 m a.s.l., Viseu district) (Fig. 1) were selected to conduct the present study. The wildfires destroyed 295 ha in Ermida (26th July 2010) and 5066 ha in S. Pedro do Sul (6th August 2010) (DUDF, 2011). The burnt areas were predominantly covered by eucalypt (*Eucalyptus globulus* Labill.) and maritime pine (*Pinus pinaster* Ait.) plantations. The mean annual temperature varies within similar ranges, 12–19 °C and 13–15 °C, at E and S, respectively. Both areas are characterised by a humid meso-thermal climate, with wet winters and prolonged dry and warm summers (Köppen Csb; DRA, 2002). Over the past 30 years, the annual rainfall at E and S ranged from 960 to 2531 mm and 523 and 1757 mm, respectively (SNIRH, 2013). The soils of the two study areas are mapped as humic Cambisols and, to a lesser extent, Leptosols or Luvisols, developed over granite, mainly for S. Pedro do Sul, and schist, mainly for Ermida (Cardoso et al., 1971).

Fire severity was assessed according to the methodology described in Shakesby and Doerr (2006), Keizer et al. (2008) and Keeley (2009). Hence, fire severity assessment was based on the degree of consumption of the canopies of 10 randomly selected trees as well as of the litter layer (partial or total), and on ash colour (black, grey and/or white), which was classified by visual inspection. The ash samples were ranked by shade (from dark to light) by comparing the samples with each other, without using the standard of greyness for comparison (Stronach and McNaughton, 1989; Parson et al., 2010). The fire severity at each slope was determined at 5 points along a transect (Fig. 1). In Ermida, the fire consumed the tree canopies only partially but the litter layer entirely, and produced black ashes; in S. Pedro do Sul, the fire totally consumed both the tree canopies and the litter layer, and produced grey-white ashes. On the basis of these observations, fire severity was classified as moderate in the case of Ermida and as moderate to high severity in the case of S. Pedro do Sul.

### 2.2. Sampling design

Three hillslopes were selected in Ermida (E), both in eucalypt (BE) and pine (BP) burnt stands: E-BE1, E-BE2, E-BE3; and E-BP1, E-BP2, E-

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