



Review

Risk of post-fire metal mobilization into surface water resources: A review



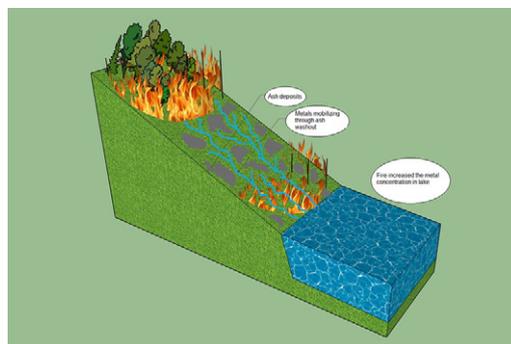
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HIGHLIGHTS

- Forest catchment supply high quality water to a number of communities around the world.
- Forest fire release sequestered metals from soil organic matter and vegetation.
- Post-fire erosion rapidly transports these metals to downstream soil and water bodies.
- Their deposition in the water bodies affects the water quality and aquatic biota.
- This metal contamination may reach to human being as a consumer.

GRAPHICAL ABSTRACT



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ABSTRACT

One of the significant economic benefits to communities around the world of having pristine forest catchments is the supply of substantial quantities of high quality potable water. This supports a saving of around US\$ 4.1 trillion per year globally by limiting the cost of expensive drinking water treatments and provision of unnecessary infrastructure. Even low levels of contaminants specifically organics and metals in catchments when in a mobile state can reduce these economic benefits by seriously affecting the water quality. Contamination and contaminant mobility can occur through natural and anthropogenic activities including forest fires. Moderate to high intensity forest fires are able to alter soil properties and release sequestered metals from sediments, soil organic matter and fragments of vegetation. In addition, the increase in post-fire erosion rate by rainfall runoff and strong winds facilitates the rapid transport of these metals downslope and downstream. The subsequent metal deposition in distal soil and water bodies can influence surface water quality with potential impacts to the larger ecosystems inclusive of negative effects on humans. This is of substantial concern as 4 billion hectares of forest catchments provide high quality water to global communities. Redressing this problem requires quantification of the potential effects on water resources and instituting rigorous fire and environmental management plans to mitigate deleterious effects on catchment areas. This paper is a review of the current state of the art literature dealing with the risk of post-fire mobilization of the metals into surface water resources. It is intended to inform discussion on the preparation of suitable management plans and policies during and after fire events in order to maintain potable water quality in a cost-effective manner. In these times of climate fluctuation and increased incidence of fires, the need for development of new policies and management frameworks are of heightened significance.

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Contents

1. Introduction	1741
2. Metals in the forest catchments	1743
3. Impacts of fire and metal mobilization in forest catchments	1744
4. Ash – Remnant of forest fire	1745
5. Post-fire metal mobilization into surface water resources	1746
5.1. Post-fire mercury mobilization	1749
6. Conclusion	1749
Conflict of interest	1751
Acknowledgments	1751
References	1751

1. Introduction

Catchment management is an overarching premise for water regulation and policies which governs the oversight of the hydrology and hydrochemistry of surface and groundwater resources (Ferrier and Jenkins, 2010). Healthy forest catchments with moderate to high annual precipitation and low contaminant concentrations are essential for the provision of potable water, together with irrigation, industrial and recreational uses around the globe. In addition, they also provide significant aquatic habitats (Neary et al., 2009; Smith et al., 2011). Many urban areas and cities rely heavily on the water provided by forest catchments including two-thirds of the municipalities in the USA and one-third of the world's largest cities including Melbourne, Sydney, Tokyo, Los-Angeles and Rio de Janeiro (CHIFM, 2008). Over 4 billion ha of forest catchments (31% of the total landmass) around the globe store, filter and provide high quality water that provide substantial economic benefits including an estimated US\$ 4.1 trillion annually by limiting the cost for purifying and sanitizing water treatments and associated infrastructure (Bladon et al., 2014). However, the ongoing challenge of supplying substantial quantities of high quality water to communities is significantly increased by the impacts of both natural and anthropogenic activities including forest fire.

Fire is a natural disruption that affects many forested catchments in tropical, temperate and boreal regions (González-Pérez et al., 2004; Neary et al., 2006; Shakesby and Doerr, 2006; Beganyi and Batzer, 2011). It has been estimated that >30% of the land surface is subjected to substantial fire occurrence (Chuvieco et al., 2008) and each year fire burns nearly 650 M·ha of forests around the globe (Shcherbov, 2012). Recent studies highlight that climate change will increase the frequency and areal extent of fire particularly in association with other factors such as rural depopulation, land abandonment and afforestation with fire-prone species (Pausas, 2004; Hennessy et al. 2005; Westerling et al., 2006; Moreira et al., 2009, 2011; IPCC, 2013, Campos et al., 2015, 2016).

Fire can substantially effect a range of physical and biogeochemical properties of soil and is able to both release and redeposit metals on the soil surface (Certini, 2005; Odigie and Flegal, 2011; Campos et al., 2012, 2015, 2016). This may be either as the result of combustion of vegetation and soil organic matter (resulting in the breakdown of metal complex organic compounds) or through the interactions of fire on the soil surface resulting in ash and charcoal deposition (Bitner et al., 2001; Certini, 2005; Goforth et al., 2005; Jovanovic et al., 2011; Costa et al., 2014). Fire also alters catchment behaviour by affecting key hydrological factors, which control infiltration and surface runoff (Ellis et al., 2004; Knoepp et al., 2008; Shakesby and Doerr, 2006). The formation of a temporary water-repellent layer during fire, at or near the soil surface reduces infiltration, which increases overland flow, peak discharge, and overall water yield (when compared to pre-burn conditions) and hence the magnitude and timing of stream flows (DeBano, 2000; Benavides-Solario and MacDonald, 2001, 2005; Kunze and Stednick, 2006; Shakesby and Doerr, 2006; Moody et al., 2009). On the other hand, water repellency is sometimes very low or removed

by fire, specifically in twice burned plots (Keesstra et al., 2016a). This overland flow increases the erosion rate and sediment load consequently increasing toxic and other metal concentrations in downstream water resources. These processes affect water quality parameters including pH, electrical conductivity (EC), turbidity, dissolved oxygen, major ions, nutrient loading (for example nitrate, sulphate, phosphate) and major and trace metal concentrations (Wallbrink et al., 2004; Kunze and Stednick, 2006; Shakesby and Doerr, 2006; Scherer, 2008; Moody and Martin, 2009; Campos et al., 2015, 2016).

Leaching of contaminants, including metals occurs throughout the soil profile and may result in groundwater contamination, specifically where groundwater levels are close to the surface (Reneau et al., 2007; Costa et al., 2014). Wind may also play a significant role in metal mobilization resulting the effects being dispersed over great distances (Breulmann et al., 2002; Schaider et al., 2007). The enhanced metal mobility occurs particularly during or immediately after the first major post-fire rainfall event(s) and has elevated impacts if the fire occurs on a landscape with already raised contaminant levels. This metal mobility has the potential to significantly impact on the quality of water from forest catchments and their associated rivers and water storages and introduces potential health impacts on human and ecosystems (Kelly et al., 2006; Sillins et al., 2009a; Smith et al., 2011; Nunes et al., 2017). However, it should be noted that all forest fires do not necessarily result in major changes such as soil erosion, increased runoff and raised sediments load together with enhanced pollutant connectivity and transport. Exceptions to these severe effects are found in low-intensity prescribed fires and fires which occur on flat lands and areas with low precipitations. Post-fire erosion and runoff and the resulting metal mobility of the area, are influenced by number of factors such as fire severity, post-fire metal concentration in the soil, frequency, intensity and duration of the rainfall, soil surface exposure, hydrological properties of the soil, distance to the water resources, slope of the land, intensity and direction of the wind together with a number of other minor local factors (Smith et al., 2011). In all the environments, the physical dynamics of the location strongly control the localised effects and magnitude of fires impacts.

There is a widely held view that forest fire is a catastrophic phenomenon that affects human lives and properties and which has significant impacts on the atmosphere and terrestrial and aquatic ecosystems (Kelly et al., 2006; Lavorel et al., 2007; Cerda and Doerr, 2008; Burke et al., 2010; Beganyi and Batzer, 2011; Melendez-Perez et al., 2014; Nunes et al., 2017). However, fire is a natural phenomenon of many terrestrial ecosystems and these systems may have adapted to require fire at some level in order to thrive. The effects of fire including moderate increases in erosion, runoff, sediment load and pollutant transport, which are all considered as a part of the natural temporal changes (Neary et al., 2005; Pausas and Keeley, 2009; Pereira et al., 2011). Low to moderate severity prescribed fires and high severity wild fires may even renew essential nutrients for long-term sustainable developments of forest ecosystems (Arocena and Opio, 2003; Pereira et al., 2011). Nevertheless, although fire may be natural and part of an evolved ecosystem, increased rates of fire coupled with increased contamination of surface

materials present a significant risk to catchment systems and may have a significant impact on human populations.

It is recognised that fire is not the end of the forest ecosystem. As it is a natural phenomenon, most forests will recover from fire after several years. In most cases plants will succeed in recolonising the burned forest area and most of the soil properties will return to pre-fire levels and in some cases the event will enhance the forest environment (Certini, 2005). Herbaceous plants and shrubs are reported to be the

type of vegetation that most easily regenerate in the post-fire environment and may quickly dominate the landscape (Costa et al., 2014). After two years regeneration of the trees becomes apparent (Costa et al., 2014). In this respect, Lane et al. (2008) reported a link between vegetation recovery and post-fire alteration in surface water chemistry.

Metals in the environment, particularly Cr, Cd, Cu, Hg, Mn, Ni, Pb, Zn and the metalloids As (herein after called a 'metal') are of great environmental and human health concern due to their toxicity, environmental

Table 1

Some of the human health risks of As and selected metals including their guideline values.

Metals	Guideline value (mg L ⁻¹) – WHO (2008)	Human health risk	Reference
As	0.01	(1) Low intelligent quotient (2) Hypertension and arteriogenic effect (3) Skin lesions (4) Cancer in several organs including bladder, kidney, lungs and liver (5) Chronic iAs ingestion –diabetes (6) Chronic iAs ingestion –cardiovascular disease (7) High level exposure -peripheral neuropathy/vascular disease, and even death	(1) Cobbina et al. (2011) (2) Chen et al. (1996) (3) Valenzuela et al. (2005) (4) Gallaher et al. (2002); Leak et al. (2003); IARC (2004); Valenzuela et al. (2005); Celik et al. (2008); ATSDR (2011) (5) Coronado-Gonzalez et al. (2007); Del Razo et al. (2011); (6) Chen et al. (1996); Mazumdar (2008) (7) USDL (2004); Zukowska and Biziuk (2008)
Hg	WHO – 0.006 (inorganic Hg) Australia - 0.001 (total Hg)	(1) Low intelligent quotient (2) Neurological disorders (3) Create toxicity to kidney (4) Total and structural damages to the brain and central nervous system (CNS) (5) It can cross the placenta and can affect foetuses (6) Methyl Hg may create neurological disorders and mental disability (7) Minamata disease - Impaired hand-eye coordination, memory and speech loss, blurred vision, blindness, muscle weakening and spasms – leads to death in some cases	(1) Cobbina et al. (2011) (2) Cobbina et al. (2013) (3) Gallaher et al. (2002) (4) Cobbina et al. (2013) (5) Bradl (2005) (6) Gallaher et al. (2002) (7) Solomon (2008)
Pb	0.01	(1) Increase in blood lead level (BLL) (2) Decreasing life expectancy (3) Effects on CNS (4) Nerves, skeletal, circulatory, enzymatic, endocrine and immune system effects (5) Hyperactivity, cognitive impairment, reduced intelligence, and increased behavioural problems (6). Cardiovascular, developmental, gastrointestinal, haematological, musculoskeletal, neurological, ocular, renal and reproductive effects (7). It can cross placenta resulting in miscarriage, stillbirths, birth defects and neurological damage	(1) Pirkle et al. (1994) (2) Lacatsu et al. (1996) (3) Gallaher et al. (2002); Leak et al. (2003) (4) Zhang et al. (2012) (5) NTP (2012); Solomon (2008) (6) ATSDR (2011) (7) Solomon (2008)
Cd	0.003	(1) Very low exposure level leads to kidney damage, bronchitis, and osteomalacia. (2) Pulmonary adenocarcinomas, prostatic proliferative lesion, bone fracture, kidney dysfunction and hypertension (3) Cognitive impairment in children (4) Skeletal deformities, bone loss, kidney damage and generalised pain, anaemia (5) Category 1 human carcinogen (6) Affects human nerves, pulmonary renal system (7) Itai-itai disease giving joint, bone and muscle pain, kidney dysfunction	(1) Young (2005) (2) Scholz and Ellerbrock (2002); ADWG (2011); Cobbina et al. (2013) (3) Cobbina et al. (2013) (4) Solomon (2008) (5) Bradl (2005) (6) Kim et al. (2002) (7) Young (2005); Solomon (2008)
Cr	0.05	(1) Dermatitis, lung cancer, nasal septum atrophy, lactate dehydrogenase in bronchioalveolar lavage fluid (2) Mouth ulcers, nosebleeds, kidney disease, low white blood cell count and depressed immune system, miscarriages and variety of cancers. Babies may be born with deformed spines	(1) Offenbacher (1994) (2) Bradl (2005); Wright and Welbourn (2002)
Cu	2.0	(1) Gastrointestinal symptoms at low concentrations (2) Poisoning may occur at high concentrations	(1) Gallaher et al. (2002)) (2) Gallaher et al. (2002)
Zn	3.0	(1) Essential element for human consumption but a very high concentration may have aesthetic and gastrointestinal effects (2) Deficiency reduction in growth, and a high dose creates Cu deficiency (3) Affects human nerves, pulmonary renal system	(1) Gallaher et al. (2002) (2) Chesters (1997)
Mn	0.4	(1) High concentrations may have an aesthetic effect and it stains plumbing fixtures (2) Impairment of neuro-behavioural functions and affects CNS	(3) Kim et al. (2002) (1) Wasson et al. (2003); Townsend and Douglass (2004); White et al. (2006) (2) WHO (2008); Cobbina et al. (2013)
Ni	0.07	(1) It may cause conjunctivitis, eosinophilic pneumonitis and asthma. (2) May induce embryo toxic and nephrotoxic effects, allergic reactions and contact dermatitis (3) It is a potential carcinogen for lungs and may cause lung fibrosis and cancer of respiratory tract in occupationally exposed people	(1) Athar and Vohora (1995) (2) US-EPA (2002) (3) Kasprzak et al. (2003)

persistency and tendency to bioaccumulate and biomagnify in the food chain (Adriano, 2001; Ignatavicius et al., 2006; Kelly et al., 2006; Campos et al., 2012, 2015, 2016; Nunes et al., 2017). Elevated metal concentrations not only degrade the quality of the atmosphere, soil, water bodies and food crops, but also threaten the health and well-being of humans and other animals who reside on the land or consume water and food from the affected area (Wang and Mulligan, 2009; Nabulo et al., 2010; Cobbina et al., 2013). Deleterious health effects such as cancer, hyperactivity, reduced intelligence, reduction in physical growth, gastrointestinal-renal-respiratory disorders, hearing loss, developmental abnormalities, cognitive impacts including mild mental retardation and many others may occur when metals are continuously ingested or inhaled by humans (Table 1) (Chen et al., 1996; Centeno et al., 2007; Mazumdar, 2008; ATSDR, 2011; Del-Razo et al., 2011; Pearce et al., 2012; Cobbina et al., 2013). In aquatic ecosystems metals can alter the structure and composition of resident microbial communities (Nipper et al., 1998; Minshall, 2003; Beganyi and Batzer, 2011) and can accumulate in fish species which may reach the human consumer (Kelly et al., 2006). Elevated metals also interfere with physiological functions of plants with disruption of gaseous exchange, CO₂ fixation, respiration, nutrient absorption, and growth rate (Angelova et al., 1999; Kabata-Pendias, 2011). Increased forest fire frequency combined with intensified post-fire rainfall, wind and the resulting metal mobility is thus viewed as a threat to human and ecosystems health owing to the toxicity and bioaccumulation properties of the metals (Hernández et al., 1997; Pardini et al., 2004; Goberna et al., 2012; Guénon et al., 2013; Muñoz-Rojas et al., 2016). In general, these metals either in deficiency or excess can impact on plants, animals and human health (Hindwood et al., 2003). Therefore, determining the concentration, distribution and mobility of toxic metals in soil and water both pre- and post-fire environment is required to assess the health of the terrestrial and aquatic ecosystems and to devise strategies to minimize impacts.

During fire, mobilization and deposition of metals occur through complex interactions involving combustion of vegetation, mineralization of the soil organic matter and causing interactions of hot ash with the underlying soil (Jakubus et al., 2010; Pereira and Ubeda, 2010; Jovanovic et al., 2011; Odigie and Flegal, 2011, 2014; Costa et al., 2014; Campos et al., 2016). Studies from the USA (Bitner et al., 2001; Sabin et al., 2005; Burke et al., 2010; Stein et al., 2012; Burke et al., 2013; Burton et al., 2016; Nunes et al., 2017), Australia (Leak et al., 2003; Wasson et al., 2003; Lane et al., 2008; Kristensen et al., 2014), Portugal (Campos et al., 2012; Costa et al., 2014; Campos et al., 2015, 2016; Nunes et al., 2017), Chile (Odigie et al., 2016), Lithuania (Ignatavicius et al., 2006) and Korea (Shin et al., 2002), have reported on metals (Cd, Co, Cr, Cu, Hg, Mn, Ni, Pb, Zn and As) in burnt forest soils and ash, and the capacity of rainfall runoff and storm events to produce flows carrying large sediment loads enriched with metals into surface water resources. However, the post-fire impacts on water quality (in terms of metals), including the magnitude of their potential effects, the persistence of their influence and their impacts on organisms (including humans) are not well investigated and documented. Understanding the dynamics of metal concentrations between the land and the water interface is essential for appropriate health risk assessments for quantifying agricultural impacts and for long-term ecosystem management and sustainability planning.

This paper is a review of the state of the art literature dealing with the risk of post-fire mobilization of selected metals into surface water resources. The articles related with this topic have been selected using standard search tools including 'ISI Web of knowledge' and 'Google scholar' interrogated with key words and phrases related to forest fire and metal mobilization. Global examples have been used to illustrate the distribution and mobility of post-fire metal concentrations into water resources and the resulting impacts on ecosystem and human health, in order to promote further discussion on the preparation of suitable management plans in catchment areas during and after significant fire events. The findings of this review will help United Nations

(UN) to achieve their sustainable development goals (Keesstra et al., 2016b).

2. Metals in the forest catchments

The forest floor is a dynamic component of the forest ecosystem and includes organic matter and a mixture of minerals, which act as a potential reservoir of metals, water soluble organic compounds and biogenic substances (Shcherbov, 2012). Metals are geogenically introduced into the terrestrial environment (through weathering and soil erosion, volcanic eruption, atmospheric fall out and cosmic dust), and they find their way into forest and aquatic environments in varying concentrations (UNEP, 2013). Metal concentrations have dramatically increased since the industrial revolution through anthropogenic activities (fossil fuel-based power generation, manufacturing, increased rates and modes of transportation, mining and smelting activities, inorganic agricultural practices, waste incineration and unmonitored land filling) (Nriagu, 1989; Pacyna and Pacyna, 2001; Wei and Yang, 2010). Metals liberated or mobilized by these activities can contaminate air, surface and ground water, terrestrial, lacustrine and marine environments (Boyle et al., 2005). They are often persistent of pollutants in the ecosystems tendency to accumulate in sediments with long persistence times (Pereira and Ubeda, 2010; Jovanovic et al., 2011; Soliman et al., 2015). If these metals remain sequestered in soil or sediment, their impact is limited. However, mobilized and ingested by animals and humans significant damage may follow (Jain et al., 2008).

The deposition and the subsequent accumulation of metals in the forest ecosystem is related to their adsorption by soil organic matter, clay mineral fixation and their association with Mn oxide and Fe and Al hydroxides (Kaschl et al., 2002; Sipos et al., 2005; Kabata-Pendias, 2011). Their behaviour is complex but is dominantly controlled by adsorption and desorption processes including many specific physical, chemical and biogeochemical processes (Reis et al., 2015). Once these metals enter a catchment, they may be (i) concentrated in the organic layers of the soil (especially in the case of Hg, Cu and Pb) (Shcherbov, 2012), (ii) sequestered in sediments, (iii) compounded with soil organic matter (Rea et al., 2000; Biester et al., 2002; Grigal, 2003), and (iv) be absorbed by plants and hence become fixed in the system, significantly reducing their mobility (Hernandez et al., 2003; Biswas et al., 2007; Obrist et al., 2008; Friedli et al., 2009). For example, approximately 90% of the Hg in the forest is found to be accumulated and stored in the surface organic layers of the soil (Grigal, 2003). The binding mechanisms that hold them in place, and the resultant concentration in the forest ecosystem, both-increase with humus formation (Shcherbov, 2012). The presence of other metals, soil pH and redox potential, landscape position, stand age, and biota spectrum all affect humus formation (Sposito, 1989; Munthe et al., 1998). It is generally agreed that Cr, Cu, Pb are strongly binding with soil organic matter, whereas, Cd, Co, Mn, Ni and Zn are weakly binding (Tipping, 1998; Lawlor and Tipping, 2003). Immediately after their deposition metals are generally enriched on the surface layer of the forest soil, then slowly migrate to the lower layers. Consequently, their concentrations decrease rapidly with depth (Xin-Ling et al., 2008; Shcherbov, 2012).

Plants are an intermediate reservoir of metals which are taken from the primary metal sources of soils, water and air and if subsequently ingested by animals or humans, they are able to move into these organisms and can have negative health impacts (Adriano, 1986; Kabata-Pendias and Pendias, 1992; Kabata-Pendias, 2004, 2011). Plant leaves may also extract dust particles including metal aerosols from the atmosphere, and rain events wash these particles from the leaves allowing them to reach to soil, where they are subject to uptake by plant roots (Rea et al., 2000, 2001, 2002; Frescholtz et al., 2003; Fay and Gustin, 2007). This metal intake increases with the mobility of metals and their concentration in the soil. For example, Zn and Cd are very mobile and easily absorbed by plants, whereas Hg, Cu and Pb are strongly bound to soil particles which reduce their immediate

availability (Obrist, 2007; Jovanovic et al., 2011; Rutter et al., 2011; Driscoll et al., 2013). Plants, dependent on species, age and other stress factors, may accumulate metals without any noticeable signs of damage for an extended period and fire liberates metals by either volatilization into the atmosphere (for example Hg) or accumulate on the soil surface within the ash. Thus, the floristic forest ecosystem plays a crucial role in the accumulation and circulation of metals and forest fire is a significant contributor to subsequent metal mobility.

3. Impacts of fire and metal mobilization in forest catchments

During fire, the plant biomass and litter layers are typically consumed and the mineral soil is heated, which affects its physical and biogeochemical properties (Certini, 2005). These changes are either short-term or long-term (Arocena and Opio, 2003) while some may be permanent (Certini, 2005). The changes depend chiefly on the nature of the forest, the severity and frequency of fire events and post-fire climatic conditions (Certini, 2005). It has been established that fire is able to release and remobilize sequestered metals from plants and soil organic matter through a relatively complex set of processes (Biswas et al., 2007, 2008; Burke et al., 2010; Odigie and Flegal, 2011, 2014; Kristensen et al., 2014; Burton et al., 2016; Odigie et al., 2016). To illustrate this process, Parra et al. (1996) reported elevated levels of Mn (471 to 1429 mg kg⁻¹) in the post-fire soil environment in the Sierra de Gredos mountain range, and Costa et al. (2014) reported five-fold increase of Mn concentration in ash (compared to the underlying soil) after the Marao Mountain forest fire. Both of these observations were made in Spain. The increase in Mn (67 to 598 mg kg⁻¹) and Pb (52 to 122 mg kg⁻¹) concentration recorded in ash after the 2010 Ermida Fire in Portugal have also been highlighted (Campos et al., 2016). Moreover, a number of authors have reported that Hg stored in forest soil and biomass was volatilized into the atmosphere in the form of insoluble and unreactive gaseous elemental mercury (Friedli et al., 2001, 2003; Engle et al., 2006; Obrist et al., 2008; Biswas et al., 2007, 2008; Burke et al., 2010; Woodruff and Cannon, 2010; Campos et al., 2015). The mobilized metals are transported to the downstream water resources by runoff and wind activities. For example, Hg introduced into the surface soils by gold mining is mobilized by fire into downstream aquatic ecosystems, where the conditions may favour the formation of the neurotoxic and immune toxic methyl Hg (MMeHg). This may then bioaccumulate in the terrestrial and aquatic food web (Morel et al., 1998; Mahaffey, 1999; Hightower, 2004; Kelly et al., 2006; Mergler et al., 2007; Friedli et al., 2009; Foy et al., 2012). Similarly, a three-orders-of-magnitude increase in Pb and Cd were observed (compared to pre-fire concentration) in an urban fringe watershed (Burke et al., 2013) and an increase in concentrations of As, Fe, Pb, Mn and Ni were observed in water samples collected from the burned area (Burton et al., 2016), both after the Station Fire in California (USA) in 2009.

Soil organic matter and pH are the two important parameters which controls the liberation and mobility of metals in the forest ecosystem. When metals enter the forest, they become associated with soil organic matter as most of the metals (Hg, Cu, Ag and Pb) have an affinity towards it and sequester within it with an immobile phase. Increase in the amount of soil organic matter in the forest ecosystem increases the metal concentrations. Soil organic matter shows variability in concentration immediately after the fire in number of occasions (Castro et al., 2011). During fire, elevated soil temperature is sufficient to decompose the metal-humic acid bond resulting in the release of metals onto the soil, air and water environments. Laboratory study indicates that some of the metals (for example Hg) can completely release from soil organic matter by fire (Friedli et al. 2001) and this metal release depends primarily on fire severity (Biswas et al., 2007). High pH value resulting from neutral or alkaline reactions attributes to the metal accumulation in the post-fire soil environment (Bogacz et al. 2011). However, decrease in pH few months after the fire (in most cases) due to intensive precipitation and runoff increase the metal mobilization. The

accumulation of new acid products of plant decomposition on the soil surface may also intensify this process (Zaideman et al. 1999).

Mobility and availability of metals in soils are generally low under high content of organic matter and clay (Yoon et al., 2006). The organic matter mineralization induced by fire, led substantial amount of metals in ash and soil ready to transport, which could be a source of downstream soil and water contamination (Ignatavicius et al., 2006; Pitman, 2006). This post-fire metal concentration depends on a number of factors: pre-fire metal concentration in the soil, types and density of vegetation in the area, fire severity, maximum temperature reached, duration of the fire, types of soil, plant parts burned and many more (Someswar, 1996; Pereira et al., 2009). The post-fire contaminant soil with metals are characterised by low contents of soil organic matter, low levels of nutrients and pH imbalance (Chiu et al., 2006). The decrease in organic matter and clay and increasing pH during the burning process also increases the mobility of metals (Norouzi and Ramezani, 2013).

The correlation of metal concentration with soil pH and dissolved organic matter (measured as dissolved organic carbon) has been noted and it has been established that pH is largely responsible for metal solubility, speciation (Drever, 1988) and mobility (Violante et al., 2010). Low pH increases solubility and mobility, but decreases the organophilic characteristics of some metals (Drever, 1988). Dissolved Organic Carbon (DOC) forms strong bonds and complexes with metal ions as they have numerous functional groups with negatively charged sites (Ephraim, 1992; Tipping, 1998; Tipping et al., 2011). Mobility of Zn, Cd and Ni are greatly influenced by soil pH and are considerably mobilized when the soil is observed to have low pH; alternatively, the mobility of Cu and Pb is strongly dependent on the solubility of organic matter (Bergqvist et al., 1989). Field based studies in forest catchments also reveal that Cu and Pb show strong positive mobility relationships with DOC (Landre et al., 2009), while Cd and Zn mobility are dependent on pH (Bergqvist, 1987; Lazerte et al., 1989; Watmough and Dillon, 2007; Adkinson et al., 2008). This is supported by Oni et al. (2013), who reported a positive correlation between Cu and Fe mobility with DOC and negative correlations with soil conductivity and sulphate concentration.

The increase in forest fire events is linked to more numerous drought seasons, which also exposes large areas of soil surface suitable for erosion. Post-fire precipitation events and wind activities further increase soil erosion (up to 100-fold) with consequent increased runoff and mobilization of significant quantities of metals into downstream water resources. (Riggan et al., 1985; Cohen et al., 2005; Odigie and Flegal, 2011; Stein et al., 2012; Warrick et al., 2012; Odigie and Flegal, 2014; Campos et al., 2016). This significantly decreases water quality, increases the potential bioavailability of a range of metals and may elevate stress on the aquatic ecosystems (Schneider et al., 2007; Nunes et al., 2017). This is a multifaceted process and further complexed by availability of soil organic matter, soil and water pH, redox status, biogeochemical and hydrological processes, among others (Schindler, 2001; Landre et al., 2009) (Fig. 1).

The metals which reach the aquatic ecosystems may eventually deposit and reside in the bottom sediments of stream beds, river banks and flood plains, where they can adversely affect the ecosystem, especially within benthic populations and cause alterations in their community structure (Nipper et al., 1998; Urbaniak et al., 2008; Zhuang and Gao, 2014). Studies have shown that hydrological changes may trigger the migration of benthic-deposited metals back into water and these may be transported further downstream and act as a point of secondary metal contamination (Crossland and La Point, 1992; Kunze and Stednick, 2006). Water plants and plankton in the waterbody absorb these toxic metals from the benthic zone and this leads to an increase in metal availability to all aspects of the food chain resulting in multiple potential impacts to organisms including humans (Finlay and Callow, 1996). Apart from this food chain dynamic, human and terrestrial biota are able to absorb metals from the environment knowingly or unknowingly by way of direct ingestion and inhalation (Schneider et al.,

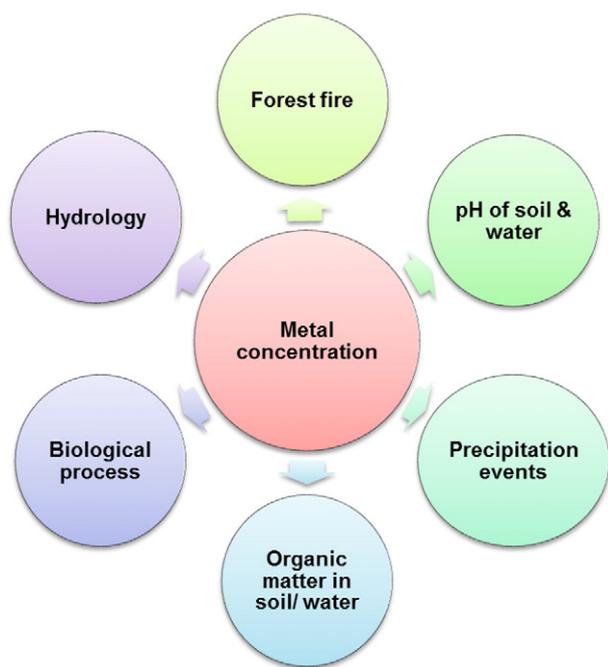


Fig. 1. Some major factors affecting the metal cycling and dynamics in the forest catchments.

2007; Pearce et al., 2012; Martin et al., 2014; Zhuang et al., 2014) (Fig. 2). Thus, metals in the catchments will contaminate the water system and can cause health impacts to human and terrestrial and aquatic biota. Therefore, there is a strong impetus to understand the factors and processes affecting metal dynamics in catchments linked to their prolonged residence time (100 to 1000 years) and their significant toxicity (Macklin and Klimek, 1992; Landre et al., 2009; Huang et al., 2011; Rose et al., 2012).

4. Ash – Remnant of forest fire

‘Soil is the mother of all things’. This shows the importance of soil to life of all living creatures on earth (Doran and Parkin, 1994). Soil play fundamental roles in its functioning, linking the atmosphere, the

subsurface and the aquatic compartments (Faber et al., 2013), and is the critical component of the natural systems both through its contribution to food, clean water and energy security and by mitigating biodiversity loss (Turbe et al., 2010; Kirkby, 2015). The vastness of the planet and the soil resources allowed agriculture to expand with growing population and store vast amount of groundwater resources, which may improve water quality (Amundson et al., 2015). Many factors and properties of soil such as soil moisture, structure, texture, pH, electrical conductivity, soil organic matter are regulating the micro-, meso- and macro-fauna of the soils (Heiden et al., 2016). Most fascinating thing about soil is that it is incredibly diverse both horizontally and vertically (Bardgett, 2016). Soil system is essential to all living beings on the Earth in such a way that the human management of soil with a holistic approach is mandatory. The future of humanity is intertwined with the future of the Earth’s soil resources. Therefore, fifth of the December is celebrated annually as the “World Soil Day” to highlights the importance of soil.

Ash is considered as a part of the soil system. It is a complex mixture of organic and inorganic residues produced by fire which may be subsequently deposited on the soil surface in burned areas (Bento-Goncalves et al., 2012). It is a heterogeneous material composed mainly of particulate carbon, oxides and hydroxides of base cations (Ca^{2+} , Mg^{2+} , P and K^+), silica, phosphorous, nutrients (which promote plant recovery), major and trace metals and other potential contaminants (Pereira et al., 2014). Ash includes components from the accumulation of mineral matter stored in vegetation and in the soil organic horizon prior to burning (Biswas et al., 2007) and is characterised by high alkalinity and electrical conductivity (Goforth et al., 2005; Plumlee et al., 2007; Gabet and Bookter, 2011; Granged et al., 2011; Campos et al., 2012; Wade et al., 2013; Campos et al., 2015, 2016). Ash produced at high temperatures may have a high pH value (Etiegni and Campbell, 1991; Henig-Sever et al., 2001) and a high $CaCO_3$ content (Ulery and Graham, 1993; Goforth et al., 2005; Pereira et al. 2008) which inhibits the solubility of metals (Bodi et al., 2014).

The composition and properties of ash depends on number of factors such as fire temperature, types of fuel, plant species burned, vegetation structure, leaf types, plant parts burned, completeness of combustion and so on (Goforth et al., 2005; Gabet and Bookter, 2011; Bodi et al., 2014). The ash pH is usually higher than the underlying soil (even pH values of 8–14 have been noted) and is associated with increased concentrations of carbonates, oxides and hydroxides (Pereira et al., 2011). A high ash pH enhances the availability of some elements and can

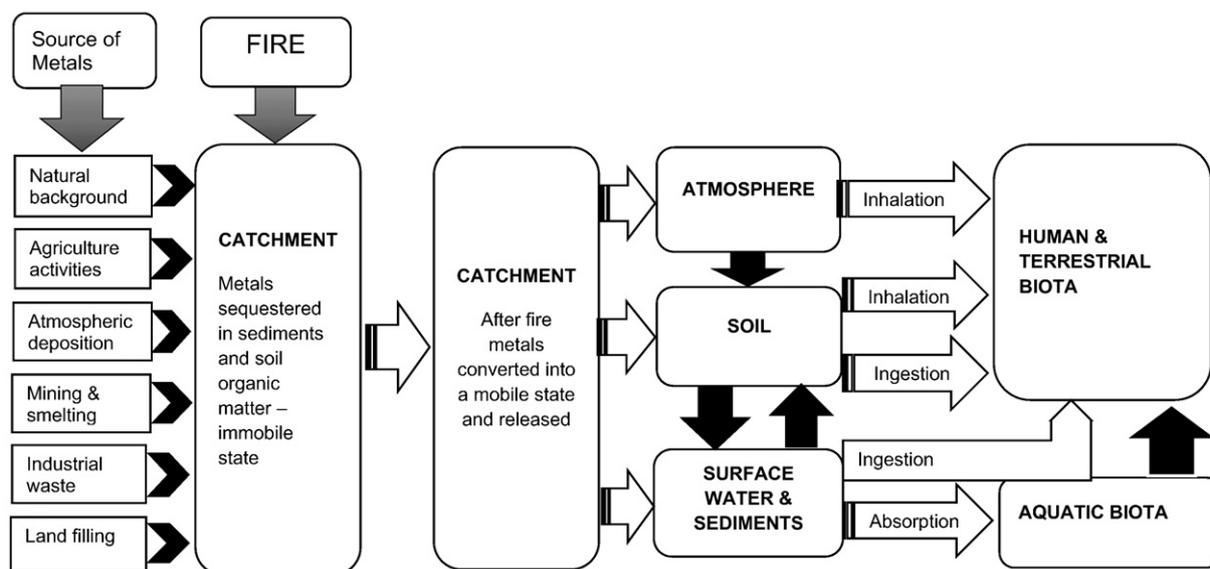


Fig. 2. A conceptual model outlining the sources of metals to the catchment, transport of these post-fire metal to water resources and pathways of exposure. Modified from Schaidler et al. (2007).

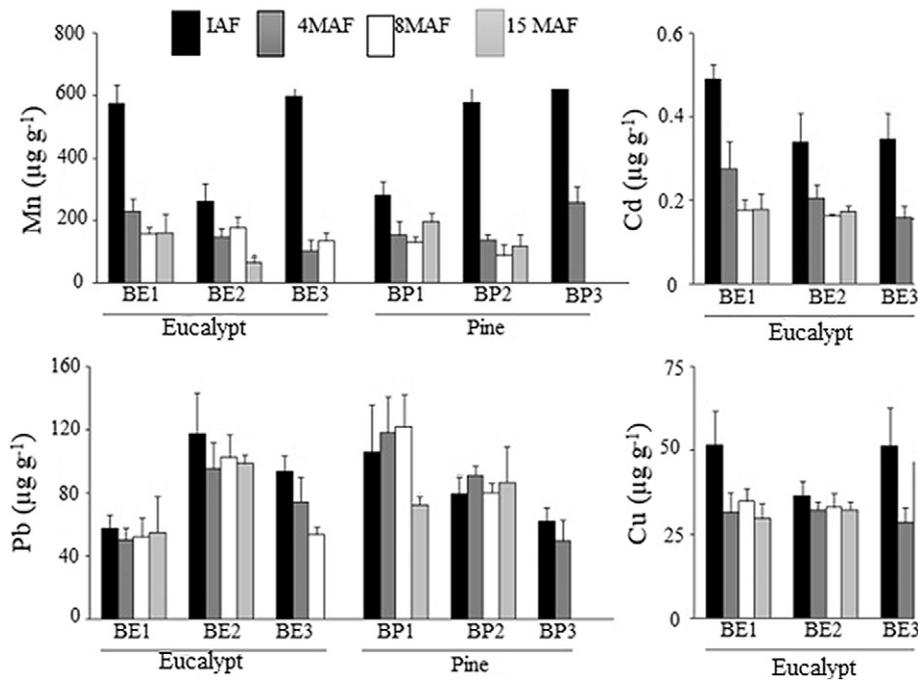


Fig. 3. Concentration of Mn, Cd, Pb and Cu in ash immediately after fire, 4, 8 and 15 months after the fire (IAF, 4MAF, 8MAF, 15MAF respectively) in Eucalypt and Pine plantations after the Ermida Fire (Portugal) in 2010 (Campos et al., 2016). Note the increase in concentration of metals immediately after the fire in most cases.

reduce the availability of others and ash can inhibit the germination of plants due to the alkaline pH and toxic effects of some ions (Bodi et al., 2014; Pereira et al., 2014). In addition, a high percentage of carbon in ash may reflect a higher organic matter content in the soil and litter (Costa et al., 2014). A decrease in clay and soil organic matter together with an increase in pH during the burning process increases the mobility of these metals in the forest soil environment (Yoon et al., 2006).

Ash can provide valuable information about the impacts of fire on the landscape and the colour of the ash is used to estimate the fire severity (Pereira et al., 2014). Black ash corresponds to low to medium fire severity, whereas grey and white ash are the result of high severity fires (Goforth et al., 2005; Pereira et al., 2014). During a forest fire, complete combustion of fuel takes place at or near 500 °C leading to the formation of light grey or white ash which is composed mainly of alkaline earth oxides that over time react with atmospheric CO₂ and water vapour to form soluble hydroxides and carbonates (Ulery et al., 1993). White ash has a significantly higher Munsell hue value and chroma (Goforth et al., 2005). Black ash has a higher concentration of cations probably due to the oxidation of organic matter and incomplete combustion (Goforth et al., 2005; Liang et al., 2006). Goforth et al. (2005) found that variation in colour lightness of ash was correlated with variation in pH ($r = 0.89$), cation exchange capacity (CEC) ($r = 0.82$), total N ($r = 0.74$) and total organic C ($r = 0.72$). Both pH and CEC increased with lighter ash colour, whereas total N and organic C increased with darker ash colour (Goforth et al., 2005).

The thickness of an ash layer is controlled by amount of available fuel and the fire severity. Areas where high fire severity is observed usually present with a thin cover of ash (Pereira et al., 2013). Ash produced at low temperature is more hydrophobic than ash at high temperature (Bodi et al., 2011). A long term ash study in the Mediterranean has highlighted that ash is the key component that determine the post-fire hydrogeomorphological properties of fire-affected landscape (Doerr and Cerdà, 2005).

Major and trace metals such as Mn, Fe, Zn, Cu, Pb, Hg, Cd and As are routinely reported in ash (Khanna et al., 1994; Someshwar, 1996; Ferreira et al., 2005; Pereira and Ubeda, 2010; Campos et al., 2016; Olsson et al., 2017) with Mn returning the highest concentrations (in a number of cases) as it is mostly concentrated in leaves (Parra et al.,

1996; Costa et al., 2014; Campos et al., 2016). For example, Costa et al. (2014) reported high concentrations of Mn in ash (121–448 mg kg⁻¹) after the Marao Mountain forest fire in Spain and Campos et al. (2016) reported high concentrations (57–598 mg kg⁻¹) after the Ermida fire in Portugal (Fig. 3). Al is also observed in high concentrations in some rare cases dependant on vegetation type. For example, after analysing the ash from the Lisbon (Portugal) fire (July 2007) in the area dominated by *Pinus pinaster* and *Quercus suber* trees, Pereira and Ubeda (2010) noted that Al³⁺ was the metal released in the highest quantities and Zn²⁺ was the lowest. Ash monitoring after the 2007 southern California forest fire showed significantly elevated levels of metals (As, Pb, Cu and Zn) that persisted at least a year following the fire (Plumlee et al., 2007).

The concentration of metals in ash varies and is dependent upon a number of factors such as type, density and age of the vegetation, plant parts burned (leaves, bark, litter, etc.), soil types, soil organic matter, pre-fire metal concentrations in the soil, meteorological conditions, maximum temperature reached, fire intensity and duration (Ulery and Graham, 1993; Someshwar, 1996; Pereira et al., 2009; Pereira and Ubeda, 2010; Machado et al., 2015). The variable amounts of metals and other contaminants that are present in ash introduce potentially toxic components into the hydrologic network when the ash is washed into the stream and other water resources in the post-fire environment (Niemenen et al., 2005; Ignatavicius et al., 2006; Pitman, 2006; Smith et al., 2011; Santin et al., 2015).

5. Post-fire metal mobilization into surface water resources

Forest fire is demonstrated to alter physical and biogeochemical properties of the soil and is able to liberate, mobilize and redeposit metals from soil organic matter and vegetation in air, land and water environments. This may potentially impact the health of the terrestrial and aquatic ecosystems (Certini, 2005; Burke et al., 2010, 2013; Smith et al., 2011; Burton et al., 2016; Odigie et al., 2016; Nunes et al., 2017). This is most prominent in forests with already elevated background metal concentrations and fire-suppressed mature forests where metals may have accumulated for extended period of time (Woodruff and Cannon, 2010). Strong and prolonged drought increases the frequency and scale of the forest fire with subsequent post-fire precipitation and runoff being able

to transport large quantities of sediments with metal contaminants to downstream surface water systems, thus changing the water quality and making it unfit for human consumption. This dynamic may exist for a few months or even years after fire and have significant consequences (Gallahar and Koch, 2004; Goforth et al., 2005; White et al., 2006; Smith et al., 2011; Stein et al., 2012; Costa et al., 2014). For example, in the last two decades, fires have affected the water supply catchments of Denver and other south-western US cities, four major Australian cities such as Sydney (2001), Canberra (2003), Adelaide (2007) and Melbourne (2009) including various regional towns by way of contamination (Smith et al., 2011; Martin, 2016). In regional Australia, the eastern Victorian Alpine Fire during 2003 generated debris flow in the upper Buckland River which recorded maximum Fe, As, Cr and Pb concentrations of 2467, 40, 18 and 98 times the respective guideline values (Leak et al., 2003). Similarly, in south-eastern Australia, a single post-fire (within first year) summer storm event (>2 h; 43 mm, peak 15 min with intensity of 80 mm h^{-1}) accounted for 127 t of total suspended sediment yield from a small wet eucalyptus forest mountain catchment (Lane et al., 2008). Moody and Martin (2004) also observed large post-fire (within one year) sediment yield of 50 t ha^{-1} , in a catchment area in the western US.

Many studies report on the presence of metals in burnt forest soils and the capacity of storm events in carrying large sediment loads with metals into surface water resources (Ignatavicius et al., 2006; Rulli and Rosso, 2007; Pierson et al., 2008, Burke et al., 2010, 2013; Burton et al., 2016; Odigie et al., 2016). In 1977, Auclair examined Cu, Zn and Mn concentration in wetland plant tissues and in the soils of *Carex* meadow, in the Qubec Province of Canada and reported that these metals are significantly mobilized by burning (Auclair, 1977).

The Cerro Grande Fire in New Mexico, USA occurred in May 2000, and was one of the major fire events in the last decade. It burned almost 17,200 ha of forest land (Bitner et al., 2001) and 239 residential structures (Wolfe et al., 2004). The post-fire runoff events were monitored and water samples were collected at 40 ephemeral stream sites. The water analysis results (for metals) were compared with 20 years of pre-fire values (Bitner et al., 2001) and concentrations of Fe (560 mg L^{-1}), Mn (102 mg L^{-1}) and Zn (3.6 mg L^{-1}) were elevated at 1870, 255 and 1.2 times the WHO guideline values respectively. Elevated concentrations of As, Ba, Cr, Hg and Pb were also reported (Bitner et al., 2001). Similar to this, the 2002 Hayman Fire in a mature forest (along the Colorado side of the Rocky Mountains, USA) resulted in a 2 to 2500 times increase in concentrations of As, Al, Cd, Fe, Pb and Hg,

which affected the drinking water quality (Ranalli and Stevens, 2004). Comparable results were also recorded at the Rodeo-Chediski Fire in Arizona, USA (Gill, 2004).

One year after the Hayman Fire, a major fire occurred in 2003 in the Lost Creek area of Alberta, Canada, which burned around 21,000 ha of crown land forest (Sillins et al., 2009a). Seven watersheds were examined to determine the effects of forest fire, including stream flow and water quality. Results obtained from this study showed elevated mean concentrations of a number of metals in the burned catchment areas (Sillins et al., 2009b). Two subsequent years of monitoring also emphasized that total Hg concentrations exceeded both chronic and acute provincial water quality guidelines on a number of occasions. For example, an extremely high Hg pulse of $265 \mu\text{g L}^{-1}$ ($0.001 \mu\text{g L}^{-1}$ = maximum acceptable concentration in drinking water) was measured after a moderate precipitation (20 mm) several days after the fire event (Sillins et al., 2009b).

In addition to the above, a number of post-fire observations were recorded in California, USA, between 2003 and 2009. The contaminant loading from these fire events found that the mean Cu, Pb and Zn concentrations in storm water were between 112 and 736-fold higher in burned catchments (Fig. 4) (Stein et al., 2012) and ash fallout also showed a three-fold increase in metal concentrations (Stein et al., 2012). Similarly, elevated concentrations of Fe, Mn and Al were found after the 2010 Fourmile Canyon fire Colorado, USA (Writer and Murphy, 2012).

Recently, Burton et al. (2016) conducted a similar study in California after the 2009 Station fire (USA). Authors collected ash, burned soil samples, pre-storm and storm water samples after the fire and analysed for metals. They reported that the total concentrations of As, Pb, Zn and Ni were higher in those samples collected in burned areas, which indicates that fire played a major role in mobilizing these metals (Fig. 5) (Burton et al., 2016).

The post-fire sediment load and metal delivery in the Arroyo Seco watershed in California, USA, after a fire in 2009 were studied by Burke et al. (2013). A significant increase (three orders of magnitude) was observed in most metal concentrations during the peak discharge time, compared to pre-fire values (Fig. 6). The highest concentrations of Pb and Zn occurred during the rising limb of the hydrograph, immediately following the peak precipitation. Cd peaked earlier than Pb, immediately following the first pulse in precipitation, while the highest concentration of Cu occurred on the hydrographs' falling limb (Burke et al., 2013). The maximum metal concentration occurred during the

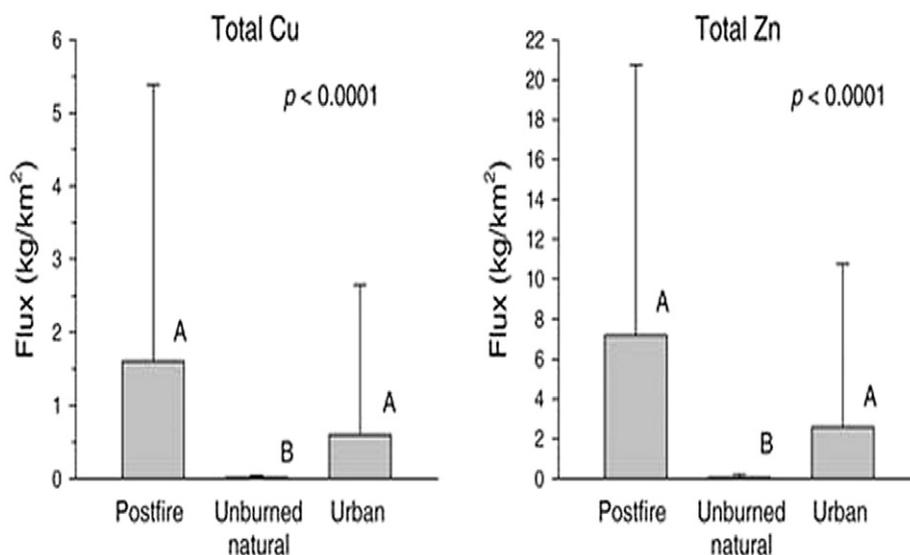


Fig. 4. Variation in concentration of Cu and Zn in pre- and post-fire storm water samples collected after the Southern California forest fire (in between 2003 and 2009). The bars represent the mean flux + standard deviation of individual storms. The letters indicate statistical similarities (Stein et al., 2012). Note the increase in concentration in the post-fire environment.

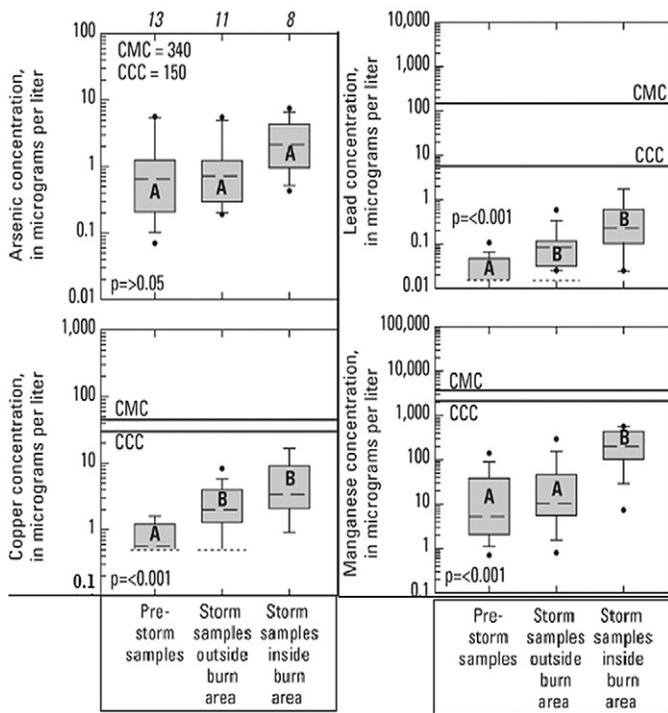


Fig. 5. Metal concentrations in water samples collected during and after the 2009 Station Fire California (CMC- Criteria Maximum concentration for aquatic species; CCC - Criteria Continuous Concentration for aquatic species) (Burton et al., 2016).

first rise in flow, well before the peak discharge and the authors suggest that this pattern should be taken into account in any post-fire storm water sampling program.

2003 is considered a disastrous year in the Earth's recent fire history, particularly in Australia. After a prolonged drought, extensive forest fire events occurred in the east of Victoria in Australia (2003), affecting 1.5 M·ha of land in the Eastern Victorian Alpines. An intense post-fire summer storm event in the catchment area of the Ovens River in North-Eastern Victoria (2003) resulted in an increase in concentrations of As, Cu, Pb, Cr, Fe and Zn that were 4, 32, 33, 40, 47, and >50 times respectively enriched when compared to the pre-fire concentration (North East Water, 2003). In the Upper Buckland River in north-eastern Victoria, a post-fire rainstorm event (38 mm for 2 h) delivered large amounts of sediments (59,000 mg L⁻¹ suspended solids and 1,29,000 NTU) into a surface water system which supplies drinking water to Canberra. Analysis showed elevated concentrations of As (0.28 mg kg⁻¹, six times the guideline value (gv)), Fe (740 mg kg⁻¹, 2467 times the gv), Pb (0.98 mg kg⁻¹, 20 times the gv) and Cr (0.92 mg kg⁻¹, 18 times the gv) with all levels exceeding the WHO drinking water guidelines (Leak et al., 2003).

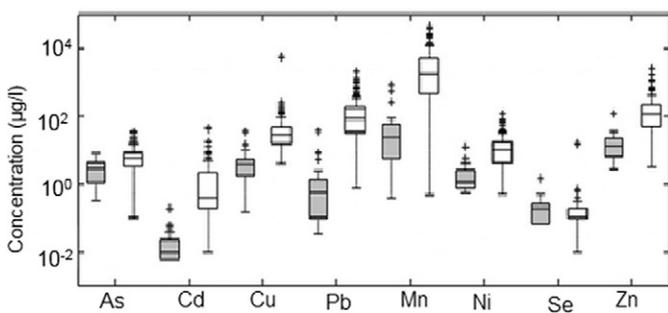


Fig. 6. Box plot show the concentrations of pre (grey fill) and post-fire (no fill) metals loads into Arroyo Seco forest catchment, USA, relative to the 2009 Station Fire (Burke et al., 2013). Note the increase in post-fire metal concentrations.

The fire also affected the Corin reservoir, located >500 km away from the core fire area. There, Wasson et al. (2003) estimated the individual post-fire event loads of Fe and Mn entering the Corin reservoir (197 km²) which supplies water to Canberra and found that three post-fire storms within 2 months of the fire generated the following Fe/Mn loads (kg ha⁻¹): 0.007/0.002 (59 mm/1 day; 25.8 ML inflow), 0.04/0.001 (72 mm/2 days; 280 ML inflow), and 0.02/0.007 (rainfall data unavailable; 153 ML inflow). Similarly, post-fire monitoring of the Fe and Mn concentrations in the Bendora reservoir (supplying water to Canberra) on the Cotter River also demonstrated an increase in concentrations of Fe and Mn compared to all other previous studies by factors of three and five respectively (White et al., 2006). Similarly, Townsend and Douglas (2004) observed increased post-fire loads of annual Fe (1.2 kg ha⁻¹) and Mn (0.22 kg ha⁻¹) transport in a Savanna forest catchment in Kakadu National Park in Australia. The possible deleterious effects of the Eastern Alpine Fire (2003) on the quality of rainwater captured from roofs and stored in tanks for private supply were studied by Spinks et al. (2006). They found that concentrations of Cd (in two sample results; 0.0034 and 0.0067 mg L⁻¹), Fe (five sample results) and Zn (seven sample results) were above the Australian Drinking Water Guideline of 0.002, 0.03 and 3 mg L⁻¹ respectively (ADWG, 2011) and there are indications that the forest fire smoke and ash may have contributed to these increases.

Linked to climate change and prolonged drought, a series of 497 fires (include land and peat bog fires) broke out in Lithuania between July and August 2002 (Sakalauskiene and Ignatavicius, 2003). In August and September 2002 the concentration of metals in Lithuanian rivers increased by 21–80% compared to those averaged over the previous eight-year period (1993–2001). These increased metal concentrations in rivers (Fig. 7) are attributed to forest and peat bog fires in Jul–Aug 2002 and the subsequent mobilization of metals into water resources (Ignatavicius et al., 2006). Similarly, post-fire ash from the *Pinus densiflora* species found in the mountain regions of south Korea contained elevated Cd (0.089 to 0.946 mg kg⁻¹) concentration, which could be mobilized by wind and deposited in eastern coastal waters and subsequently affect the marine ecosystems (Shin et al., 2002).

The impact of wild fire on an ecosystems is sometimes so devastating that it can kill fish in the watershed. Recently Nunes et al. (2017) reported a study which highlighted the impacts of wild fire (the Talhadas fire in Portugal in 2013) which mobilized metals into the aquatic biota with Mosquitofish (*Gambusia holbrooki*) sued as the biomarker. The chemical analysis of water samples revealed concerning levels of metals in both runoff and stream water and were found to be harmful to organisms. Pro-oxidative modifications were the most recent toxic effects observed in the mosquitofish. Metabolic alterations related with toxicant conjugation with glutathione were also affected by the exposure to contaminated surface water and runoff, evidencing the defensive nature of triggered biological response (Nunes et al., 2017). Moreover, dead rainbow trout (*Oncorhynchus mykiss*) have been observed in the Big Tujunga Watershed in California during the 2009 Station fire (Burton et al., 2016).

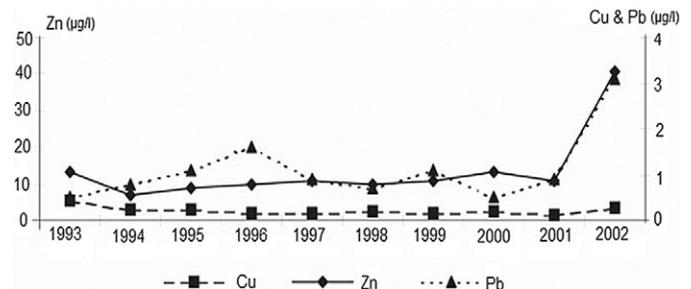


Fig. 7. Showing the increase in concentration of Pb, Zn and Cu in the Lithuanian rivers after a series of fires in Jul–Aug 2002 (Ignatavicius et al., 2006).

Information regarding the historical post-fire metal mobilization is essential to allow identification of the metal contamination of adjacent areas and to understand its impact on the aquatic ecosystem. Such studies were conducted in 2016 by Odigie et al. (2016) in Chile, South America. Temporal change in metals in lake sediments correlated with charcoal abundance was used to locate the post-fire mobilization of contaminants including metals of interest here. Results of the analysis of the age-dated sediments from Lake Thompson in Patagonia, Chile attest to the substantial pyrogenic remobilization of contaminants that occurred in Patagonia during the mid-1900s (Odigie et al., 2016).

Besides direct release onto soils, metals may also be released to the atmosphere during a forest fire. There have been concerns about the remobilization of previously deposited metals on the land and foliage to coastal and other aquatic ecosystems through areal fallout. The Angel's National Forest fire (Nov 1975) burned around 26,000 ha of forest land in the Los Angeles County Basin in the USA and created a dramatic smoke plume of roughly 100 km in width, which deposited a distinct layer of ash throughout the Basin. Distinct increases were measured in dry areal deposition rates of nine metals (Ag, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn) along the sections of the southern California coast in USA, which were affected by the forest fire smoke plume. These metals were possibly mobilized into the adjacent marine and other aquatic environments (Young and Jan 1977). Similarly, during the severe Southern California fire in the USA in 2003, the atmospheric deposition rates of Cu, Pb and Zn rose by factors of 4, 8 and 6 respectively compared with the unburned areas, which could reach soil and water resources by way of rain and subsequent runoff (Sabin et al., 2005).

5.1. Post-fire mercury mobilization

Among the post-fire metal mobilization Hg is of particular concern due to its volatility, toxicity and persistence in the environment (Huang et al., 2011). Forest fires have been recognised as a major source of mercury (re-)mobilization from soil, and living and dead vegetation (Artaxo et al., 2000; Friedli et al., 2001; Wiedinmyer and Friedli, 2007). Fire directly influenced the emission and deposition cycles by mobilizing it from soil and vegetation pools into the atmosphere (Biswas et al., 2007; Burke et al., 2010). The Hg release depends on number of factors including tree species composition (Biswas et al., 2007; Campos et al., 2016), moisture content in the fuel (Obrist et al., 2008), Hg concentrations in the soil and intensity of the fire (Engle et al., 2006). Ash and post-fire soil contains generally low amounts of total Hg compared with pre-fire soil Hg concentrations due to the volatilization of Hg (Biswas et al. 2007; Mergler et al., 2007; Campos et al., 2015). The released Hg from biomass burning is eventually deposited locally and globally, transported to the aquatic ecosystem by runoff and wind and when conditions are favourable it may contribute to the formation of neuro-toxic and immune-toxic methyl mercury (MeHg) specifically mono methyl Hg (MMeHg) (Kelly et al., 2006; Friedli et al., 2009). This may bioaccumulate and biomagnify in the food chain resulting in global health concerns (Caldwell et al., 2000; Amirbahman et al., 2004; Mergler et al., 2007) since monomethyl Hg is the most toxic of the mercury compounds (Clarkson, 2002).

Many researchers (Caldwell et al., 2000; Amirbahman et al., 2004; Ravichandran, 2004; Kelly et al., 2006) found elevated Hg levels in downstream waters of burned watersheds, which it is thought have been transported by suspended sediments and organic matter (Ulrich et al., 2001; Ravichandran, 2004). Caldwell et al. (2000) measured large amounts of Hg mobilization in creek runoff in a freshly burned mixed coniferous forest, as well as noting a six-fold elevation in the concentration of total Hg and a 30-fold elevation in MeHg concentrations in the sediments of the receiving reservoirs. It is noted that this downstream sediment condition suits the formation MeHg. The elevated Hg concentrations in the downstream waters of the burned watershed were also shown to depend, among other things, on the fire severity, topography of the area, and distance to the surface water body (Caldwell

et al., 2000; Amirbahman et al., 2004; Kelly et al., 2006). Areas of poorly drained soils and wetlands are favourable for the formation of MeHg (Grigal, 2003) due to the fluctuating temperature, water levels and the low dissolved oxygen content (Scheuhammer et al., 2007). Kelly et al. (2006) noted a large unexpected pulse of both total Hg and MeHg delivered to a reservoir system in the first post-fire storm event and found that the fish in the downstream reservoir exhibited elevated MeHg concentration that was attributable primarily to the food web restructuring. A study was conducted to examine the spatial and temporal variations of Hg concentrations in sediments of an arid-land reservoir in New Mexico, USA after a forest fire. The concentration of total Hg in the sediments increased from 7.5 ng g⁻¹ to 46 ng g⁻¹ and MeHg increased from 0.428 ng g⁻¹ to 12.46 ng g⁻¹ (Caldwell et al., 2000). The first rainfall after the fire caused an accentuated reduction of mercury levels in ash (30–60%) related to the washing out process. In such case the runoff produced after the rainfall may mobilize Hg into the adjacent terrestrial areas and in the water systems downstream and their study highlighted that forest fires and subsequent rainfall play a key role in the mobilization of Hg in the environment (Caldwell et al., 2000).

Litter fall and the dissolved fraction of organic matter are important factors in Hg transport in undisturbed watersheds (Bushey et al., 2008). This creates particular concern towards Hg mobility where conditions may be conducive for the formation of methyl mercury (MeHg) (Lin and Pehkonen, 1999; Caldwell et al., 2000; Amirbahman et al., 2004; Kelly et al., 2006). Therefore, it is important to determine the amount of Hg released during a fire event to assess the relative contribution of fires to the atmospheric Hg pool and to aid in the investigation of the potential for pulses of Hg to enter aquatic ecosystems following such fires. Increasing temperature in the tropical, temperate and boreal regions, where high Hg pools reside is expected to exacerbate Hg liberation and mobility because these regions are prone to frequent and large fires which is exacerbated by climate change and land-use pattern including afforestation with fire-prone species (Friedli et al., 2009; Campos et al., 2015; 2016).

A summary of the relevant studies linking post-fire mobilization of metals to water resources are provided in Table 2. All the post-fire metal mobilization studies clearly revealed that metals including As and Hg are mobilized after fire events and reach surface water systems primarily through runoff. Among the post-fire metals studied, Hg is the one which has received peak attention mainly because of its toxicity, persistence and health impacts to human and ecosystems and its ability to travel long distances through the air. The formation of MeHg in the wetlands and other surface water systems is also of concern as it is easy to mobilize into the human system through the food chain, where it is neuro-toxic and immune-toxic. Further studies are recommended in order to quantify the extent of mobilization of all the post-fire metals into wetlands and other surface water resources and to further understand their pathways of exposure towards aquatic ecosystem and humans through the food chain. Quantification is of particular significance in areas that are contaminated and hence are already defined as having elevated metal values.

6. Conclusion

Forests are vital elements of the water cycle, in that approximately 4 billion hectares of forest catchments (30% of the total land area) supply potable water resources to many major cities and communities around the world. Thus any kind of contamination, particularly metals have the potential to significantly affect the availability of high quality potable water supplies should those metals become mobilized.

Fire is a common phenomenon in forest catchments, and it can play a significant role in the release of metals usually sequestered in soil organic matter and vegetation. It can thus facilitate their movement into range of environmental contexts and measurably increasing their mobility and bioavailability. In addition, fire events increase soil erosion rate by as much as two to 100-fold, which allows rapid transport of

Table 2
Summary of relevant studies linking forest fire to the remobilisation of selected metals including As.

Authors	Study area/year fire occurred	Fire types/Scale affected (ha)	Metals involved/assessed	Results/remarks
Young and Jan (1977)	Angels National Forest Fire (22–28 November 1975) Southern California, USA	26,000 One of the largest fires in the decade	Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn	Distinct increases were observed in dry aerial deposition rates of nine metals along the sections of the southern Californian coast covered by a forest fire smoke plume
Bitner et al. (2001)	Cerro Grande Fire (2000) New Mexico, USA	17,200	Mn, Cu and Zn	The metal concentrations in the run-off water have been observed to increase after fire (compared with 20 years of pre-fire data)
Wolfe et al. (2004)	Cerro Grande Fire, (May 2003) Los Alamos, New Mexico, USA	19,283 Controlled burn became a wild fire	As, Cd, Hg, Ni and Pb in urine samples of fire fighters	As and Cd levels were significantly elevated among National Guard members which was related to smoke exposure
Ignatavicius et al. (2006)	Lithuania, (Jul/Aug, 2002)	497 forest and peat bog fires	Cu, Pb and Zn	Ce and As levels were elevated in fire fighters In August and September 2002, the concentration of metals in river waters increased by 21–80% compared with those averaged over the previous eight-year period (1993–2001). Forest fires and peat bogs increased metal amounts in the aquatic environment. Metals were released into the atmosphere through volatilization and were trapped in smoke particles or distributed as fly ash after combustion
Spinks et al. (2006)	East Gippsland Fire, 2003 Victoria, Australia	1.5 million	As, Cd, Cr, Cu, Fe, Pb and Zn	Some Cd, Zn and Fe concentrations in the rainwater storage tanks were above the Australian Drinking Water Guidelines
Biswas et al. (2007)	(i).Boulder Creek Fire, (Aug, 2000) (ii).Green Knoll Fire, (Jul/Aug, 2000) (iii).East Table Fire, (Jul, 2003) Western Wyoming, USA	1400	Hg	Hg (22.3–81%) was released during the fires, and greater fire severity was associated with the greater percentage of soil Hg being released. It also depends on the tree species composition of the forest, which affects pre-fire Hg accumulation
Biswas et al. (2008)	Rex Creek Fire, (Aug, 2001) Central Washington forest, USA	20,234	Hg	Hg seems to be re-volatilized during fire. Loss of 6.7 g Hg ha ⁻¹ from the soil has been observed
Witt et al. (2009)	Central Washington forest, USA Northern Minnesota, USA		Hg – Total Hg and methyl Hg	Observed increase in both species of Hg. Canopy type of trees influenced the magnitude of Hg mobilization
Burke et al. (2010)	(i) Topanga Fire (2005) (ii) Pine Fire (2006) (iii) Day Fire (2006) California, USA	(i) 9800 (ii) 100 (iii) 66,000	Hg	Loss of Hg in the freshly burned soil was observed. Four-fold increase in surface Hg concentrations were observed in the Day Fire area. Following the initial volatilization of Hg during fire, the burned soil experienced an accelerated accumulation of Hg in the post-fire environment
Pereira and Ubeda (2010)	Lisbon, Portugal, Spain, (Jul, 2007)	Medium size wild fire – study in a small plot of burned area 27 × 9 m size	Al, Mn, Fe, and Zn	The ash contain high amount of metals and the metal release depends on species distribution and burning severity. Over time the increase in pH, make the metals more mobile and will be released into soil solution.
Stein et al. (2012)	(i) Old Fire, (Oct, 2003) (ii) Simi Vallet Fire, (Oct, 2003) (iii) Day Fire, (Apr, 2006) (iv) Santiago Fire, (Oct, 2007) (v) Station Fire, (Aug, 2009) Urban fringe area, Southern California, USA	(i) 36,940 (ii) 43,790 (iii) 65,840 (iv) 11,490 (v) 64,980	Cu, Pb, Ni and Zn	Mean flux of Cu, Pb, Ni and Zn from burned sites were increased 110, 740, 82 and 110-folds respectively, compared to the respective flux in the unburned (control) site. Contaminant loading from the burned landscapes has the potential to make substantial contribution to the total annual load to downstream areas in several years following the fire
Burke et al. (2013)	Station Fire (2009) Angel's National Forest, Los Angeles, USA	6,66,000	As, Cu, Hg, Al, Fe, Ni, Zn, Cd, Mn and Pb	One (As, Cu), two (Al, Fe, Ni, Zn) and three (Cd, Mn, Pb) orders of magnitude increase were observed in fluxes between largest pre- and post-fire storm events
Burton et al. (2016)	Station Fire (2009) Angel's National Forest, Los Angeles, USA	6,66,000	As, Cu, Fe, Hg, Mn, Ni, Pb and Zn	Filtered concentrations of Fe, Mn and Hg and total concentrations of As, Cu, Hg, Ni, Pb and Zn were elevated as a result of the Station Fire.
Odigie et al. (2016)	Central South-Chile Fire records (20th century)	NA	As, Co, Cu, Mn, Ni, Pb and Zn	Temporal change in the amount of metals (As, Co, Cu, Mn, Ni, Pb and Zn) and their correlation with temporal change in charcoal abundance in age-dated sediments were collected from Lake Thomson, in Patagonia Chile. They attest to the substantial pyrogenic remobilization of contaminants that occurred in Patagonia during mid-1900s.

fire-released metals to downstream land and water resources carried by runoff and wind and which may consequently impact on water resource qualities. Water resources and watersheds contaminated with metals have the potential to pose a significant risk to downstream communities and ecosystems due to the effects of these metals on human and ecosystem health.

Climate change and associated processes include increased frequency of dry and warming weather events, while concomitant human impacts include land-use change (land abandonment, afforestation with fire-prone species) have recently exacerbated forest fire frequency and their extent in many forest catchments. The expansion of fire affected landscapes and the increase severity of events posing a threat to the

sustainability of the landscapes and ecosystems. It is reported that fire and flood events will be more frequent and severe under the predicted climate change conditions, which may significantly increase the mobile metal concentration in the environment. Such an increase in metal concentration and its resulting mobility will impact on water resources and affect the potable water quality. Therefore, greater focus on metal concentration and mobility after fire together with their effects on human and ecosystem health should be reviewed to allow water authorities to predict and mitigate risk. These assessments should be based on the observed fire intensity, areal extent and frequency, the background metal concentrations, possible impacts on catchment hydrological processes, distance to the potable water source, intensity and frequency of post-fire rainfall events, and likely wind activity. This will facilitate the identification of communities with water supplies that might be most vulnerable following fire. Moreover, catchment management authorities and organisations responsible for catchment and water management should take some immediate and reasonable actions to reduce the negative impacts of fire such as: (i) construction and management of fuel breaks around the water catchment areas, (ii) regular conduct of controlled burning in fire-prone areas to reduce the chance and severity of wild fires, and (iii) avoiding the use of fire retardants near waterways and riparian zones in case of fire. These actions will reduce the ingress of contaminants into water reservoirs to minimum level.

Conflict of interest

The authors declare that they have no conflict of interest.

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