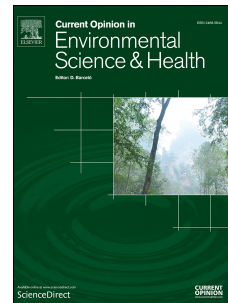


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Forest fires as drivers of contamination of polycyclic aromatic hydrocarbons to the terrestrial and aquatic ecosystems

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1 **Forest fires as drivers of contamination of polycyclic aromatic hydrocarbons to the** 2 **terrestrial and aquatic ecosystems**

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7

8 **Keywords**

9 Forest fires; PAHs; Soil; Water; Fate; Environmental pollution

10

11 **Abstract**

12 Forest fires are a well-known source of polycyclic aromatic hydrocarbons (PAHs), playing an
13 important role on their formation and redistribution across the terrestrial and aquatic
14 compartments. Fire-induced inputs of PAHs to the environment are of major concern due to
15 their toxicity, high persistence and tendency to bioaccumulate. This paper presents a synthesis
16 of the most important work on the role of wildfires and time since fire in the production and
17 mobilization of PAHs on soil and water. Furthermore, it also assesses their toxic effects on
18 aquatic species. The post-fire PAHs fluxes vary depending on a variety of factors, such as
19 vegetation composition and plant's part burnt, fire severity and post-fire hydrological
20 conditions. In general, off-site effects are particularly notorious during the initial post-fire
21 period, although not necessarily limited to it. This review highlights the role of forest fires in
22 the production and mobilization of PAHs, acting thus as a diffuse source of PAHs
23 contamination to the terrestrial and aquatic systems, thus warning to the need to be
24 considered in future monitoring/management programs on the environmental impacts of
25 wildfires.

26

27 **1. Introduction**

28 Wildfires are recognized as a natural and beneficial phenomenon and even an important
29 evolutionary driver of forest ecosystems playing an important role to keep and shape the
30 ecosystem dynamics, promoting biodiversity and productivity [1,2]. However, under the
31 changing climate conditions due to global warming and driven by human land-use change, the
32 current and future wildfire regimes have been causing widespread concerns due to their socio-
33 economic and environmental impacts such as loss of lives and properties, costs of suppression,
34 and damages to ecosystems and the services they provide [3-5].

35 The major impacts of wildfires range from the on-site effects as the destruction of vegetation
36 and organic matter, changes in soil structure, physical, chemical and biological properties and
37 geomorphological and hydrological responses [6-11] to off-site effects as the contamination of
38 fire-affected watersheds [12-16]. Arguably, the effects of the fire-induced production and
39 mobilization of ubiquitous polycyclic aromatic hydrocarbons (PAHs) [17-23] assumes particular
40 attention and raise environmental and biological concerns due to their toxic, mutagenic,
41 carcinogenic and teratogenic potential, persistence within ecosystems and tendency to
42 bioaccumulate [24-27]. For this reason, USEPA (US Environmental Protection Agency) has
43 listed sixteen PAHs as priority pollutants: naphthalene (NAP), acenaphthylene (ACY),
44 acenaphthene (ACE), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene
45 (FLT), pyrene (PYR), benzo(a)anthracene (BaA), chrysene (CHR), benzo(a)pyrene (BaP),
46 benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), indeno(1,2,3-cd)pyrene (IND),
47 dibenzo(a,h)anthracene (DBA) and benzo(g,h,i)perylene (BGP). Naphthalene, ANT, FLT, BbF,
48 BaP, IND and BghiP are also included in the list of priority substances of the European
49 Commission [26]. Since PAHs produced during combustion in wildfires are emitted into the
50 atmosphere, they can be redistributed between the vapor and particulate phases and are
51 subsequently transported away over long distances and/or deposited to the terrestrial and
52 aquatic environments, through dry or wet deposition [28] as well as mobilized between those
53 compartments (Fig. 1). They can be deposit onto the soil surface, either directly by combustion
54 of vegetation or mineralization of organic matter, or indirectly through interactions of ashes
55 with the underlying soil, and by litter fall. Furthermore, they can be leached into soil profile or
56 transported by overland flow, impacting surface and groundwater bodies. These post-fire
57 pollutant fluxes will vary depending on a variety of factors such as soil type, topographic
58 conditions of the terrain, vegetation composition, fire intensity, and post-fire climate
59 conditions, such as timing, wind and intensity of precipitation events [8-10,14-15,17-19,21,39].
60 Although PAHs are strongly adsorbed to organic particles they can volatilize and degraded by
61 abiotic and biotic processes [29]. Therefore, it is important to consider the risks of PAHs
62 contamination posed by wildfire to the environment.

63 The present review tackles the role of wildfires as a potential source of PAHs contamination to
64 the terrestrial and aquatic ecosystems, highlighting the current research on this topic.

65

66 Figure 1: Transport of polycyclic aromatic hydrocarbons (PAHs). (Modified from [58])

67

68 **2. Effects of wildfires on the production and mobilization of PAHs in forest** 69 **ecosystems**

70 *2.1. Wildfires impacts on the terrestrial ecosystems*

71 An important consequence of wildfires with hydrological, geomorphological and ecological
72 implications is the deposition of a layer of ash on the soil surface [30]. Although ash properties
73 change according to the fire severity, type and part of burnt vegetation, temperature and time
74 of contact a [30-32], they are mostly composed of oxides, hydroxides and carbonates; the
75 main inorganic components are magnesium (Mg), silicon (Si), potassium (K) and calcium (Ca),
76 and, to a lower extent by sodium (Na), phosphorous (P), sulphur (S), and other elements
77 aluminum (Al), vanadium (V), chromium (Cr), manganese (Mn), iron (Fe), cobalt (Co), nickel
78 (Ni), copper (Cu), zinc (Zn), arsenic (As), cadmium (Cd), mercury (Hg) and lead (Pb) [9,16,31,33-
79 38]. In addition, wildfire ashes may also contain organic compounds, such as polycyclic
80 aromatic hydrocarbons (PAHs) and occasionally polychlorinated dibenzodioxins, dibenzofurans
81 (PCDD/Fs) and polychlorinated biphenyls (PCBs) [17-19, 34-35].

82 Worth stressing to this respect is that the ash layer is typically very prone to post-fire
83 mobilization and export by wind and water, leading to the leaching of those compounds into
84 the soil and groundwater as well as to their transport by overland flow to water bodies.
85 Likewise, the presence of ash after a fire can have huge impacts on terrestrial and freshwater
86 ecosystems [30-31].

87 Indeed, several authors [17-19, 34-35] reported significant amounts of the Σ_{16} PAHs, ranging
88 from 458 ngg⁻¹ to 14078 ngg⁻¹, in wildfires ashes from different geographical regions,
89 vegetation types and fire severity. After the 2017 Thomas Fire in southern California Wang et
90 al. [34] reported levels of Σ_{16} PAHs in ashes from orchards between 2840 ngg⁻¹ to 4450 ngg⁻¹.
91 Campos et al. [18] in a study conducted in Portugal, in eucalypt and pine forest reported a
92 temporal decrease 15 months after the fire on the Σ_{15} PAHs from 458 ngg⁻¹ to 275 ngg⁻¹ and
93 from 695 ngg⁻¹ to 285 ngg⁻¹, respectively. A similar trend (from 11007 ngg⁻¹ to 1169 ngg⁻¹) was
94 reported by Simon et al [17] in the levels of Σ_{16} PAHs in ash from pine forest collected in South
95 Korea 19 and 492 days after the forest fire. In a study conducted by Harper et al. [36] in ashes
96 from six wildfires types [different species, fire severities and countries: Australia (AUS), United
97 Sates of America, Canada, Spain and United Kingdom (UK)] found significant variations in the
98 concentration of Σ_{16} PAHs, ranging from 1155 to 14078 ngg⁻¹, the highest total being found in
99 the moderate severity fire in the UK and the lowest in the moderate to high severity fire in the
100 AUS. Campos et al [18] also found that ash contents showed a clear tendency to be lower
101 (eucalypt: 315 ngg⁻¹; pine: 429 ngg⁻¹) following a high severity fire than following a moderate

102 severity fire (eucalypt: 458 ngg⁻¹; pine: 695 ngg⁻¹). Fire severity seems to play some role in the
103 total ash PAHs concentrations. Important to note that in, general, ashes were dominated by of
104 2-3 ring (NAP, PHE, FLU, ACY) PAHs followed by 4-ring (FLT, PYR) [17-19,35]. In contrast, Wan
105 et al. [34] found a considerable amount of higher ringed PAHs (≥ 4 rings) which can be due to
106 the combustion of irrigation pipes in orchards.

107 The incorporation of ash into the soil profile has been referred in many studies as the main
108 cause of changes in soil physical and chemistry properties [18,30,34]. Indeed, increases in
109 PAHs in soils after ash deposition have been reported in several studies [18,21,34,39,40].

110 Despite wildfires being identified as a major source of PAHs, with the smoke of biomass
111 burning revealing high PAH levels [28,41], their recognition as a source of diffuse
112 contamination to terrestrial ecosystems has received less attention [17,18,20,21,34,39,42,43].

113 Overall, the former studies suggest that wildfires leads to the deposition of PAHs on the soil
114 surface, bounded to ash and charcoal particles, and in an increase of PAHs contents of the
115 topsoil, bound to soil organic matter. Indeed, PAHs concentrations in burnt soil immediately
116 after the fire were markedly higher than unburnt soil concentrations, as reported by Tsibart et

117 al. [42] and Abakumov et al. [43] in soils of drained peatlands in Moscow, Russia and sandy
118 podzol soils of Nadym, Russia, respectively. Campos et al. [18] in a study conducted in eucalypt
119 forest in Portugal and Choi [40] in a pine forest in South Korea, also reported enrichment in
120 burnt soils of approximately 5 times relatively to the unburnt soils 1 month after the fire. Chen
121 et al. [39] assessed that the levels of PAHs of unburnt soil (247 ± 58 ngg⁻¹) < white ash/burnt soil
122 (515 ± 333 ngg⁻¹) < black ash/burnt soil (893 ± 285 ngg⁻¹). In line with the former authors, Campo

123 et al. [20] and Rey-Salgueiro et al. [21] in studies conducted in recently burnt forests in Spain
124 also reported increased levels of PAHs in burnt soils compared to unburnt sites. The implied

125 substantial wildfire-increased in PAHs contents is supported by the formation of PAHs by
126 combustion of plant biomass during wildfires [44,45]. Possibly, the PAHs of the topsoil
127 originated, at least in part, from the ash layer, itself. Namely, PAHs concentrations on ashes
128 immediately after the fire (as described early) were markedly higher than topsoil
129 concentration [17,18,34,39]. For example, Campos et al. [18] reported values of Σ PAHs: 458 vs.

130 148 ngg⁻¹ (eucalypt) and 695 vs. 242 ngg⁻¹ (pine), for ashes and burnt topsoil, respectively,
131 while Simon et al. [17] found values of 11007 vs. 294 ngg⁻¹, respectively. In contrast, Simon et
132 al. [17] and Wan et al. [34] reported similar values between the unburnt and burnt soils.

133 However, with time since fire (especially after the first rainfalls), PAHs contents tend to
134 decrease especially due to soil erosion by wind and water, and by leaching from the ash layer
135 into the soil and to the groundwater as well as the vaporization and degradation of 2-3 ring
136 PAHs [17,18,40]. As in the case of ashes, fire severity can influence the levels of PAHs in burnt

137 soils. For instance, in the study conducted by Campos et al. [18] values tended to be lower
138 after a high severity fire than after a moderate severity fire, both in the case of eucalypts sites
139 (-10%) and of the pines sites (-30%). Chen et al. [39] also reported the same trend.

140 Similar to the ash profile, burnt soils were characterized by the dominance of 2-4 ring PAHs
141 (NAP, PHE, FLU, FLT and PYR) [17,18,20,21,39,40], which is in accordance with the most
142 produced aromatic compounds during wildfires [28,41] and by pinewood and needles
143 combustion [45]. For example, Campos et al. [18] observed a temporal decrease (from 69% to
144 44% over the first 4 months) in the contribution of the 3-ring PAHs mainly PHE and FLU) to the
145 total PAHs while the 4-ring (FLT) increased. In general, this pattern was in line with other
146 authors, who also reported a decreasing trend in the fraction of 2-3 rings PAHs (NAP, PHE) and
147 an increase in the fraction of 4-ring PAHs over time [17,40].

148 In spite of this decrease with time since fire, post-fire PAHs loads can pose a long-term threat
149 to the environment implying a degradation of the land. Clearly, the results from the former
150 studies consistently indicate that forest fires can be a significant non-point diffuse source of
151 PAHs to the terrestrial systems.

152

153 *2.2. Wildfires impacts on the aquatic ecosystems*

154 The risks of post-fire contamination are not limited to the burnt soils but can also affect
155 surface and groundwater bodies within and downstream of the burnt area. Transport of PAHs
156 from soil to water by surface runoff, either in dissolved or particulate form is likely to be
157 significant in recently burnt areas, as fire typically enhances overland flow generation and the
158 associated transport of ash and soil particles to downstream surface water bodies [10,12-16],
159 impacting their water quality [22,45-51]. Furthermore, PAHs deposited in the bottom
160 sediments of water bodies can resuspend periodically, leading to lasting water quality
161 issues [52]. These contaminants can also be leached into the soil profile impacting
162 groundwater [22,53,54].

163 In the surface water, PAHs can volatilize, photodegrade, oxidize, biodegrade by aquatic
164 organisms, bind to particulates or accumulate in aquatic organisms (see Fig.1), depending on
165 their physicochemical properties. Because of their low solubility and high affinity for organic
166 carbon, PAHs in aquatic systems are primarily found sorbed to particles that either settle to
167 the bottom or as suspended in the water column [56]. In sediments, PAHs can biodegrade or
168 accumulated in benthic organisms [56].

169 Although the importance of considering the impacts of the inputs of PAHs to freshwater
170 systems, only few studies have been addressing this issue. Olivella et al. [46] and Vila-Escalé et
171 al. [47] observed an increase in post-fire inputs of PAHs to streams one month after a fire in

172 Catalonia, Spain, although, in the first case was attributed to atmospheric deposition of ashes
173 (in absence or low rain after the fire) and in the second, to post-fire erosion. In both studies,
174 the 3-ring (PHE) and 4-ring (PYR) were the dominant PAHs in water samples. Furthermore, the
175 total concentrations of the PAHs decreased with time since fire, approaching the background
176 levels. Worth stressing is that the concentrations of PAHs measured remain within the
177 European drinking water limits and never reached levels of toxicological concern. In line with
178 the previous studies, Schäfer et al. [49] monitored the levels of PAHs in 9 streams in Victoria,
179 Australia following a wildfire and found that compounds with 3-ring PHE and FLU) and 4-ring
180 (PYR) were the predominant (levels of PAHs ranged from 0.1 to 9 ngL⁻¹). In a study conducted
181 by Stein et al. [48] in Southern California it was observed that the PAHs flux was 4 times
182 greater from burned areas than from adjacent unburnt areas.

183 A study conducted by Mansilha et al. [22] in Caramulo Mountain, Portugal during 19 months,
184 showed increases of 1.2-4.0 times higher than in the unburnt control samples, with the 2-ring
185 (NAP) and 5-6 ring (BbF, BaP, BkF, BghiP and IND) as the most detected PAHs. The decay
186 process of PAHs with time since fire was also observed in this study getting closer to the ones
187 of the control samples by the end of the first year. Mansilha et al. [22,53,54] also determine
188 the influence of wildfires on PAHs levels in groundwaters in Estrela and Gerês Mountains and
189 Braga, in Portugal and found increases of 1-to 6-fold higher in systems within the burnt area
190 than in the control sites, and a temporal reduction over time after intense precipitation
191 conditions. These studies corroborate that wildfires can affect water quality and can threaten
192 downstream drinking water supplies. Despite this, the detrimental impacts of the post-fire
193 inputs of PAHs to the freshwater communities have been vaguely explored.

194 First steps to overcome this research gap were given by Campos et al. [50] and Vera et al. [35],
195 whose studies demonstrate that PAHs released or mobilized by wildfires were capable of
196 exerting significant toxicity to the bacteria *Vibrio fischeri*, the microalgae *Raphidocelis*
197 *subcapitata* and the macrophyte *Lemna minor*. By contrast, no significant effects were found
198 towards the cladoceran *Daphnia magna*. In contrast, Harper et al. [19] found effects on the
199 immobilisation of *D. magna* when exposed to 3 of the 6 ash types. Furthermore, Nunes et al.
200 [51] used a biomarker approach to find early warning signs of toxicity triggered by the
201 exposure of the fish *Gambusia holbrooki* to aqueous runoff and stream water collected from a
202 burnt catchment in Portugal. Pro-oxidative modifications involved in glutathione metabolism
203 were the most relevant deleterious effects found, while no signs of neurotoxicity were
204 observed. More recently, and in the same water samples as Nunes et al. [51], Carvalho et al.
205 [56] showed the adverse effects on microbial communities and invertebrates in stream detrital
206 food webs and they observed that the chemical composition and source of runoff explained

207 the different effects observed. In this overall context, Pradhan et al. [57] investigated the leaf
208 consumption behaviour and the responses of oxidative and neuronal stress enzymatic
209 biomarkers in the freshwater invertebrate shredder *Allogamus ligonifer*, after short-term
210 exposure. The shredding activity was severely inhibited, and the enzymatic activity suggested
211 oxidative and neuronal stress in the shredders upon exposure to post-wildfire runoffs. These
212 results suggest that post-fire contamination can induce sublethal effects on invertebrate
213 shredders with impacts on key ecological processes in streams.

214 All the reported findings, appear to justify concerns around the impacts of wildfires inputs of
215 PAHs on water quality and aquatic biota, emphasizing the need to better understand their
216 potential toxicity.

217

218 **Final Remarks and Conclusions**

219 It has been unequivocally demonstrated that forest fires are important non-point diffuse
220 source of PAHs to the terrestrial and aquatic ecosystems as well as their numerous
221 ecotoxicological effects over multiple aquatic organisms. However, despite the recognized
222 risks that the ubiquity of PAHs in the forest ecosystems poses not only to the environment (soil
223 and water ecosystems), but potentially also to human health, several research gaps remain in
224 understanding their terrestrial and aquatic impacts.

225 A major challenge is to understand the transport and fate of PAHs in forest ecosystems and
226 accordingly, which mechanisms are in stake. Fire induced PAHs loads can affect soil quality,
227 fertility and productivity, especially when wildfires occur with a frequency or severity outside
228 of the historic range of variation. Due to the persistence of PAHs in the environment, they can
229 pose a long-term threat, which may lead to severe damage and land degradation that may not
230 be identified until it is well advanced. This way, long-term studies are critical to better
231 understand the delayed and persistent effects of fire on PAHs fate and mobilization in the
232 terrestrial compartment. Furthermore, there is often a lack of connection between PAHs
233 delivery from the land surface and within stream transport processes. This is of major concern,
234 since contamination with PAHs represents a serious threat when they are transported and
235 reach the aquatic systems such as rivers and water supplies reservoirs. In this regard, it is
236 important to assess the fate and the medium- to long- term impacts of PAHs in the quality of
237 waterbodies, mainly due to the problem associated with the resuspension of sediments as well
238 as to assess the effectiveness of post-fire management techniques in mitigating post-fire PAHs.

239 Notwithstanding, for an adequate assessment, monitoring and predicting of the fire-induced
240 pollution risks, it is necessary to increase the knowledge on the impacts of post-fire PAHs on
241 the terrestrial and aquatic systems under several possible risk scenarios.

242

243 **Declaration of competing interest**

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

246

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253

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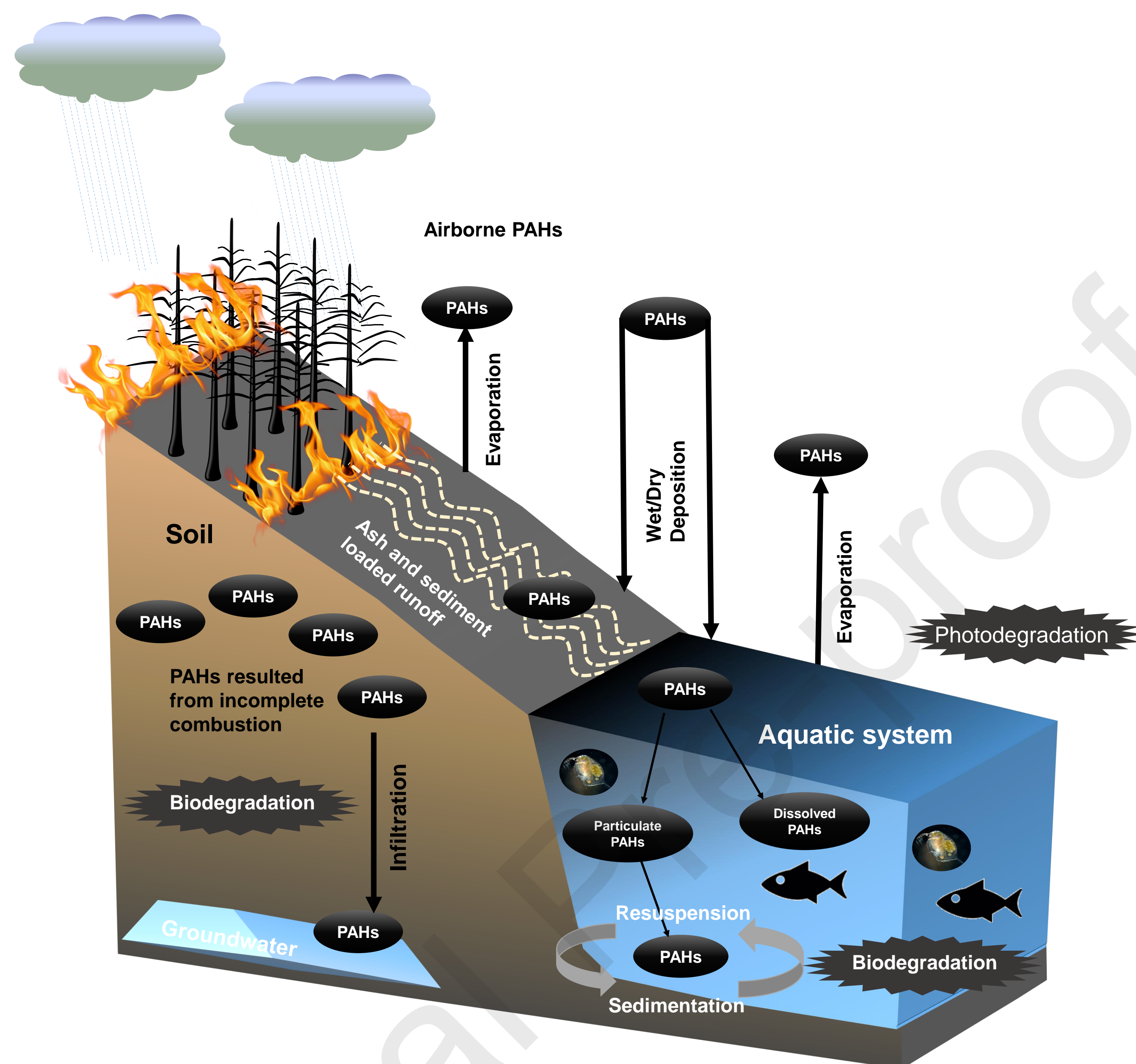


Figure 1: Transport of polycyclic aromatic hydrocarbons (PAHs). (Modified from [58])

Declaration of interests

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

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