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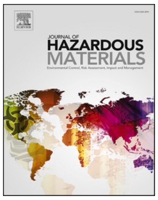

Mobilization of mercury by sediment transport after a prescribed fire in NE Portugal: Insight into size classes and temporal variation

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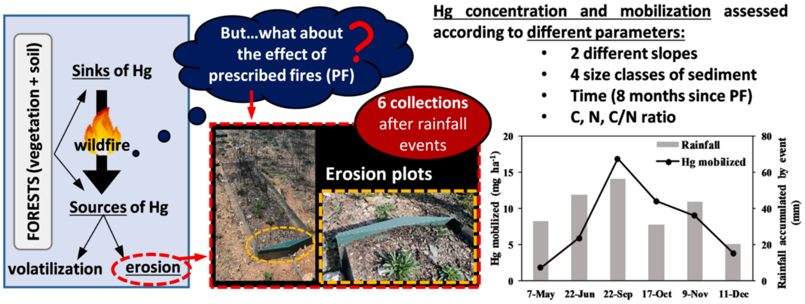
HIGHLIGHTS

- Hg mobilization is an unintended consequence of prescribed burning in SW Europe.
- Higher Hg concentrations in the fine sediment are partly explained by the C/N ratio.
- Hg levels in sediments could increase with exposure time to atmospheric deposition.
- Hg mobilization was higher during the early post-fire rainfall events.
- Early erosion control measures may reduce the arrival of Hg to aquatic ecosystems.

GRAPHICAL ABSTRACT



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ABSTRACT

Terrestrial ecosystems are important sinks for atmospheric mercury (Hg). It is well known that high severity wildfires can mobilize Hg in the surroundings of burned areas due to changes in ecosystem stability, but it is unclear whether this also occurs after lower severity fires, such as prescribed fires. The present study determined Hg concentrations and mobilization rates in different size fractions of sediments collected after a prescribed fire in a scrubland area. Sediments, collected from eight erosion plots on six occasions, were analysed for total Hg, C and N in several size classes (<0.2 mm, 0.2–0.5 mm, 0.5–2 mm and >2 mm) and Hg mobilization rates (Hg_{ST}) were calculated for each size fraction. Average total Hg were 38, 57, 94 and 126 $\mu g\ kg^{-1}$ for size fractions > 2, 0.5–2, 0.2–0.5 and < 0.2 mm, respectively. Total Hg was negatively correlated with C/N ratio, involving the humification degree of organic matter of sediments in Hg retention. In the last event (eight months after fire), sediments had 45–106 % more Hg, depending on size fraction, compared to the initial event. Mercury mobilization rates varied between 32 and 78 $mg\ ha^{-1}$, with the fraction 0.5–2 mm accounting for 46 % of the mobilized Hg. The results revealed that prescribed fires can mobilize Hg, so their use to prevent wildfires must be done with caution.

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1. Introduction

In the biosphere, terrestrial ecosystems are considered the most relevant sinks of atmospheric mercury, a global pollutant derived from natural and anthropogenic sources [1]. Among their components, the soil constitutes one of the largest pools in the Hg global biogeochemical cycle [2]. However, the content of Hg in soils is highly conditioned by the overlying vegetation, which is key in the air-soil transfer of atmospheric Hg [3] and thus, forest soils showed higher Hg concentrations than scrubland and barren soils [4,5]. The ability of these landscapes to retain Hg is affected by environmental disturbances such as wildfires [6–8], as burning of aboveground biomass and soil contribute to the re-mobilization of Hg retained in the terrestrial ecosystems [9–11]. In fact, wildfires can cause an increase in Hg concentration in burned soils due to the post-fire local deposition of Hg [12]. Global emissions of Hg from biomass burning were estimated to be more than 600 Mg Hg year^{-1} [13,14] but, in the coming decades, an increase in the frequency, spreading and severity of wildfires are expected as a consequence of global warming, changes in land use due to increased rural abandonment, and afforestation with fast-growing species [14].

Prescribed fires are increasingly being used in forest management policies as a tool to mitigate wildfire risks and consequences [15]. Although multiple ecosystems benefits are reported from the use of this prevention technique, such as the control of invasive species or the reduction of fuel loads, the use of prescribed fires as a prevention tool is still under debate [15]. The application of prescribed burns, under suitable weather conditions and the control of experts, make them less damaging to forest ecosystems than wildfires. In terms of Hg losses during fire, gaseous elemental mercury (Hg^0) starts to volatilize at 50 °C and Hg bound to soil organic matter (SOM) is released around 200–300 °C because of the thermal decomposition of humic acids [16]. Mercury losses due to wildfires strongly depend on fire severity [9], and prescribed fires, commonly characterized of moderate to low severity, are able to disturb Hg cycle in terrestrial ecosystems [10,11,17].

Many of the studies performed to date have focused on the emission of Hg into the atmosphere as a direct consequence of wildfires [13, 18–21]. However, during a wildfire, not all the Hg stored in the aboveground biomass or retained in the uppermost soil layers is volatilized to the atmosphere and thus, an unknown amount of Hg remains bound to ash and burned soil particles [8,11]. During rainstorms, the surface layer of burned soils is susceptible to be mobilized, as sediment, by water or wind erosion. In fact, soil erosion is considered one of the worst environmental aftermath of forest fires [22]. Projections for the coming decades estimate that rainfall erosion in Europe would increase by almost 20 % because of climate change [23]. Despite southwest Europe is being a region highly threatened by wildfires, few field-based research was carried out to assess the mobilization of Hg with sediments in burned areas. This is an issue of concern, as post-fire runoff would transport sediment-bound Hg downstream until reaching freshwater bodies [8,24], where it could be methylated and biomagnified through the food chain, posing high toxicity risks to animals and even humans [25].

We hypothesized that after a prescribed fire, carried out to reduce fuel load and minimize the risk of a wildfire in a scrubland area in NE Portugal, Hg could be mobilized through erosion, and Hg mobilization rates may differ depending on the sediment size and composition. Thus, the present study aims to assess the mobilization of Hg by eroded sediments over eight months after the prescribed fire. For this purpose, the amount of sediment mobilized was quantified, and total Hg concentration was analysed in different size classes. Some parameters related to organic matter (C, N and C/N) and the accumulated precipitation were considered for discussing the results obtained concerning the concentration of total Hg and its mobilization rates by sediment transport.

2. Material and methods

2.1. Study area

The study area is located within a protected area covering 750 km^2 in the NE Portugal (Montesinho Natural Park). In the last decades, the decline of agricultural and livestock activities in the area resulted in the abandonment of crop fields and favoured the spreading of scrubland. As a preventive measure against wildfires, the management plan for the Natural Park includes the application of prescribed burning to control scrub growth. The area that was subjected to prescribed fire is a scrubland located at 800 m above sea level in a plateau zone cut by deep valleys through which small streams flow (41°54′0″N, 6°40′52.6″W; Fig. 1). The climate in the study area is temperate with dry and warm summers, showing a mean annual temperature of 12 °C and mean annual rainfall in the range 800–1000 mm [26]. The lithology of the area is composed of schist resulting in shallow and stony soils which can be classified as Epileptic Umbrisol (Loamic, Eutric) [27]. The vegetation covering the burned area was the typical of scrublands in the Mediterranean biogeographical region with a predominance of species of the genus *Erica*, *Cistus*, *Genista* and, to a lesser extent, *Lavandula*.

Characteristics of the prescribed fire such as the date it occurred, the extent of the burned area, and rough temperatures reached during the fire, as well as the severity degree, can be found elsewhere ([10,28]; Fig. S1; Text S1).

2.2. Sample collection and processing

To assess Hg mobilization due to soil erosion after the prescribed fire, eight erosion plots of 4 m^2 (1 m wide and 4 m long) were installed in the burned area one week after the burning, with no raining events occurring until their deployment in the field. The erosion plots were distributed in two locations following the natural runoff pattern of the field (Fig. 1). Four plots were established on the top of the burnt area (average slope of 6.9 %) and the other four plots on an adjacent steepest area (average slope of 16.5 %). The erosion plots were delimited on the sides and the upper part by partially buried metal plates, sticking out at least 15 cm from the ground. The lower part was an obtuse-angled triangle (from the top view), consisting of a metal structure with a base that rests on the ground and a movable cover that keeps the sample protected from external disturbances. The obtuse angle (> 90°) of the triangle, which marks the lower end of the erosion plot, has a drainage outlet that connects through a hose to a plastic drum placed in a pit (Fig. 1). A double 2 mm filter was placed in the opening (lower end of the plot) to reduce the sediment loss through the hose. Periodic visits to the study area during samplings confirmed that filters were never clogged, which prevented the occurrence of standing water in the erosion plots.

The collection of sediment samples mobilized by runoff was carried out after 6 periods of precipitation (hereinafter called rain or rainfall events) occurred along 8 months since the installation of the erosion plots. It was assumed that each rain event, make up of several individual rainfall events, ended when more than 24 h elapsed without precipitation. Every sampling campaign was conducted during dry days following each rain event and the whole area was visually inspected during every visit, confirming that no erosion occurred in the surroundings where prescribed fire was not applied. The accumulated precipitations by event were 33, 47, 57, 31, 44 and 20 L m^{-2} from the first to the sixth, respectively. Precipitation data were obtained from the Portuguese Institute for Sea and Atmosphere [29]. After each rainfall event, the sediment accumulated in the lower part of the plot was collected using a brush and a trowel and placed in individualized bags. Sediment samples mostly consisted of soil particles and aggregates, burned plant debris and gravels. Once in the laboratory, after removing gravels and stones, all sediment samples were dried in a forced-air oven at 40 °C. Then, each bulk sediment sample, both the collected from the lower part of the plot and from the drum, was weighed for estimating sediment transport (ST) by runoff. It should be noted that the number of erosion plots used for this study was reduced from 8 to 6 after the second collection, as two of them (one at each slope) were assigned to a parallel experiment (not described in the present study).

In order to make a more detailed study of the Hg potentially mobilized with sediments after the prescribed fire, the bulk sediment samples were separated by sieving into the following four size classes: < 0.2, 0.2–0.5, 0.5–2 and > 2 mm. These size classes roughly corresponded to those defined by Burke et al. [30] consisting of coarse sand (0.5–2 mm), medium sand (0.25–0.5 mm) and fine sand (<0.25 mm), the latter including silt and clay. Organic matter would be present in all size classes, as physical separation only allows for size differentiation, but does not provide a separation between organic (plant/ash) and mineral (soil) remains. Before separation in size classes, an aliquot of each bulk sample was dried in an oven at 105 °C to express the results at constant dry weight. The subsamples resulting from the separation were weighed and ground with a mechanical agate mortar. The mill was properly cleaned with diluted HCl and deionized water between samples to reduce the risk of cross-contamination.

2.3. *Chemical analysis of sediments*

The concentration of total Hg (THg) was measured in the ground samples of sediment by thermal combustion and atomic absorption spectroscopy (US EPA method 7473) using a Milestone tri-cell Direct Mercury Analyzer (DMA-80). All the samples were analysed in duplicate, with any additional measures if the initial coefficient of variation exceeded 10 %. At the beginning and end of each analytical run and every twelve measurements, a standard reference material (SRM) was analysed for quality assurance and control. The SRM analysed was BCR-142R (sandy soil; certified Hg value of $67 \pm 11\ \mu g\ kg^{-1}$), obtaining 95 % as recovery percentage. Approximately 100 mg of sample were weighed in nickel vessels for Hg analysis in both the sediments and the SRM.

The concentrations of total organic carbon (C) and nitrogen (N) were analysed in the ground subsamples of all size classes by combustion using a Thermo-Finnigan 1112 Series NC elemental analyser. The C/N ratio was calculated as a proxy for organic matter quality or its humification degree.

2.4. *Calculations and statistical analyses*

All the calculations were made on a constant dry weight basis. Thus, sediment transport (ST) by runoff, as $kg\ ha^{-1}$, was calculated from the total dry mass of sediment collected divided by the area of the plot ($4\ m^2$).

The amount of Hg mobilized by sediment transport (Hg_{ST}), in $mg\ ha^{-1}$, was calculated for each size class by multiplying the corresponding sediment transport by its total Hg concentration.

Statistical treatment was made using the IBM SPSS Statistics 25 software for Windows. The influence of different size classes ($n = 4$), rainfall events ($n = 6$) and field slope ($n = 2$) on sediment transport (ST), total Hg concentration and Hg mobilization through sediment transport (Hg_{ST}) was assessed with the non-parametrical Kruskal-Wallis test (H). The Spearman rank test (r_s) was applied to assess the statistical significance of correlations between the parameters studied. Statistical significance was considered when $p < 0.05$, unless otherwise specified.

3. Results and discussion

3.1. *Hg concentration in sediment size classes*

The mean concentration of total Hg (THg) in the sediment size classes over the eight months of study increased with decreasing size following the sequence: $37.5\ \mu g\ kg^{-1}$ (> 2 mm) $< 57.1\ \mu g\ kg^{-1}$ (0.5–2 mm) $< 93.9\ \mu g\ kg^{-1}$ (0.2–0.5 mm) $< 126.4\ \mu g\ kg^{-1}$ (< 0.2 mm). For the finer sediment classes (0.2–0.5 mm and <0.2 mm), averages of THg were greater than the values reported for these fractions in the organic horizon and in the 0–3 cm layer of the mineral soil in the same study area before the prescribed fire [10], suggesting that the prescribed fire contributed to Hg accumulation in soil surface. This is consistent with a significant local re-deposition of Hg emitted during fire [12], as well as the ability of ash to adsorb ambient Hg in burned areas [11]. In addition, atmospheric Hg would be directly deposited on the soil surface due to the loss of the vegetation cover after the prescribed fire. Under natural conditions, plant cover would act as a temporary Hg reservoir by uptaking it from the atmosphere [3], thus reducing the direct deposition of atmospheric Hg on the soil surface. According to the Kruskal-Wallis test, total Hg concentrations in sediments varied significantly among size classes ($H = 126.430$; $p = 0.000$; $n = 158$), with the highest values in the finest classes, which coincided with earlier studies that reported a higher concentration of Hg in the clay and silt size fractions of soil

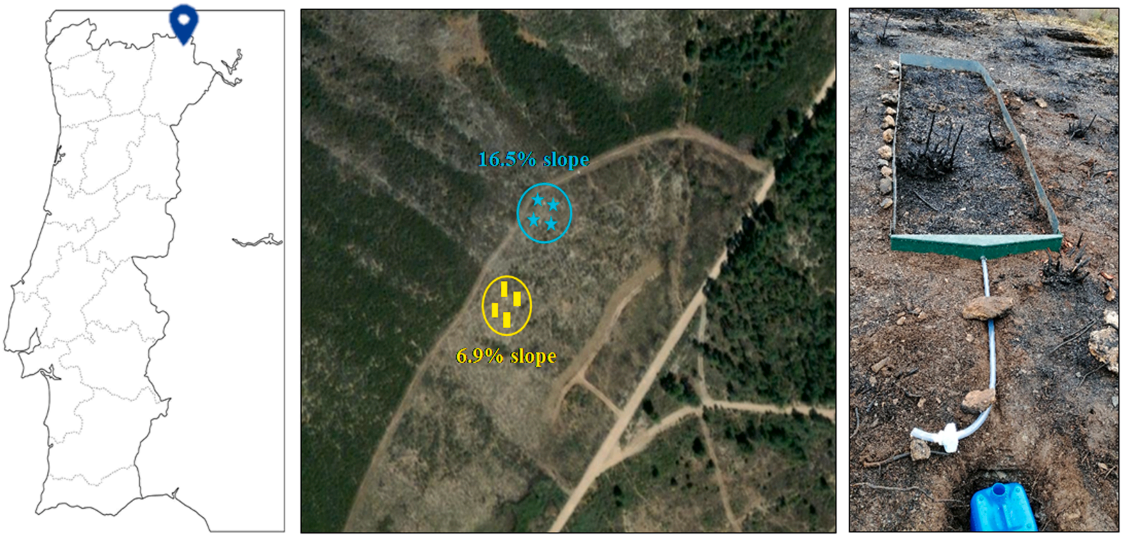

Fig. 1. Location of the study area in NE Portugal (left), distribution of the erosion plots in lower (6.9 %, yellow rectangles) and higher (16.5 %, blue stars) slope locations in the prescribed burning area (centre) and example of the plots installed after the prescribed fire (right).

samples [31–35]. In general, the lowest size soil particles have greater specific surface area and contain more organic matter, clay minerals and metal oxyhydroxides [36], providing numerous and strong binding sites for heavy metals [33,37,38]. This would justify the increasing trend of total Hg as sediments size becomes smaller. The variation in THg among size classes in sediments coincided with the results obtained for soil samples collected in burned areas [10,30].

The close relationships between Hg and soil organic matter in forest soils [39–43], and the fact that the sediments mobilized after a wildfire mostly come from the uppermost (and organic) soil layers, led to perform an analysis of the relationships between THg, total organic C and the C/N ratio in the study samples. Mean values of total organic C (TOC) in the sediment samples covered a wide range of values (204–595 g kg^{-1}; Table 1), similar to the levels reported in the O horizon of the same study area (298–523 g kg^{-1}; [10]). The size of sediment was a statistically significant factor of variation for TOC ($H = 64.007$; $n = 158$; $p = 0.000$), which agrees with Girona-García et al. [44], who found that total C differed among size fractions in soils collected at different times after a prescribed burn. Total organic C and total Hg were negatively correlated ($r_s = -0.363$; $p = 0.000$; $n = 158$), with the finest sediment class (<0.2 mm) showing the highest THg but not the highest TOC, which was observed in the > 2 mm size class (Table 1). This suggests that the quantity of TOC was not the only responsible for the amount of Hg present in sediment samples, similarly to that reported in previous studies [10,36].

A possible explanation for our results is that coarser sediment classes (>0.5 mm) were mainly composed of partially charred plant debris (Fig. S2) which still maintain a substantial amount of total C that is transferred to sediment samples, a fact that was evidenced in low to moderate severity fires [45]. Contrarily, finer sediment classes would be mostly composed of particles and aggregates of the mineral soil instead of burnt plant debris, which is supported by the relatively low C/N ratio (21−25) in the class < 0.2 mm (Table 1). These results suggest that the distribution of THg among different size classes in sediments after prescribed fire would be related to a greater extent to OM quality than to its quantity.

Table 1

Average and standard deviation of the concentrations of total Hg, total organic C, total N and C/N ratio in sediment size classes in the six rainfall events.

Rainfall event	N	Sediment size class (mm)			
		< 0.2	0.2-0.5	0.5-2	> 2
Total Hg (μg kg^{-1})					
1	8	100 ± 21	72 ± 10	34 ± 3	26 ± 4
2	8	114 ± 13	79 ± 11	48 ± 17	32 ± 12
3	6	125 ± 2	92 ± 7	60 ± 9	38 ± 8
4	6	134 ± 6	102 ± 8	67 ± 13	44 ± 7
5	6	141 ± 7	112 ± 9	72 ± 13	45 ± 8
6	6	145 ± 8	114 ± 11	71 ± 11	44 ± 5
Total organic C (g kg^{-1})					
1	8	204 ± 79	275 ± 75	225 ± 125	389 ± 90
2	8	221 ± 50	330 ± 74	330 ± 117	385 ± 165
3	6	255 ± 32	409 ± 30	436 ± 72	480 ± 147
4	6	276 ± 23	418 ± 22	455 ± 42	595 ± 47
5	6	261 ± 34	368 ± 40	405 ± 85	504 ± 79
6	6	295 ± 45	378 ± 37	436 ± 54	591 ± 120
Total N (g kg^{-1})					
1	8	9 ± 2	10 ± 3	8 ± 4	9 ± 3
2	8	10 ± 2	12 ± 2	10 ± 3	8 ± 3
3	6	11 ± 1	14 ± 2	13 ± 3	8 ± 3
4	6	12 ± 1	14 ± 1	12 ± 2	9 ± 2
5	6	11 ± 1	13 ± 1	12 ± 2	8 ± 3
6	6	12 ± 2	13 ± 1	13 ± 2	9 ± 3
C/N ratio					
1	8	22 ± 2	27 ± 2	29 ± 3	46 ± 13
2	8	21 ± 1	27 ± 2	32 ± 4	46 ± 12
3	6	23 ± 2	29 ± 3	35 ± 5	64 ± 25
4	6	24 ± 2	31 ± 2	39 ± 6	69 ± 16
5	6	23 ± 1	28 ± 2	33 ± 3	68 ± 19
6	6	25 ± 1	30 ± 2	35 ± 3	66 ± 22

The C/N ratio is an estimate of the degree of humification of soil organic matter, representing a good proxy for soil organic matter quality [46]. The mean C/N ratio in the study samples varied between 21 and 25 in the finest sediment size class (<0.2 mm) to 46–69 in the > 2 mm (Table 1). The statistically significant differences of the C/N ratio among sediment size classes ($H = 130.342$; $n = 158$; $p = 0.000$) may explain the different ability of sediments to capture and retain Hg depending on size. In fact, THg correlated negatively with the C/N ratio ($r_s = -0.732$; $p = 0.000$; $n = 158$), suggesting that Hg retention was favoured the more humified the organic matter was (lower C/N ratio). This is consistent with the greater Hg concentration observed in the finest sediments (<0.2 mm) which also showed the lowest C/N values (Fig. 2) and agrees with the findings of Méndez-López et al. [10], who found a close relationship between THg and the C/N ratio in soils affected by a prescribed fire. Our results also coincided with Amorim et al. [47], who evidenced that silt and clay-sized fractions of soils had systematically a lower C/N ratio compared to greater size fractions.

The close association between Hg and the C/N ratio in the finest sediment class may become an issue of environmental concern, as the risk of methyl-Hg production increases with the degree of organic matter humification [48]. Moreover, smaller sediments are more prone to be transported away from the burned area than coarser sediments. Both circumstances could endanger the ecological integrity of nearby freshwater ecosystems, since erosion and runoff from burned areas, particularly after intense rainfall events, may transport Hg-enriched sediments to freshwater courses [8,17,19]. Burton et al. [49] related peaks of Hg in river waters during stormflow events to the mobilization of soil particles due to runoff. In this regard, Dai et al. [50] stressed the need to provide a plant cover in burned areas as a tool to minimize the risk of soil erosion and the consequent mobilization of Hg.

3.2. *Temporal variation of Hg concentration in sediment classes*

During the 8-month study period, THg in sediments increased progressively as the collection date separates from the date of the prescribed fire (Table 1), being significantly different among rainfall events ($H = 20.092$; $p < 0.01$; $n = 158$). Thus, samples < 2 mm collected after the sixth rainfall event had between 1.50 and 1.86 times more Hg than those collected after the first event. For the different sediment size classes, percentages of Hg enrichment between initial and final collection events were 45 %, 58 %, 106 % and 66 % for < 0.2 mm, 0.2–0.5 mm, 0.5–2 mm, > 2 mm classes, respectively. A similar raise in THg during the first year following the fire was reported in surface soil and ash from burned areas [17,30,51], although such increase was also observed for trace elements (V, Ni, Cu, Zn, Co) in sediments collected in areas affected

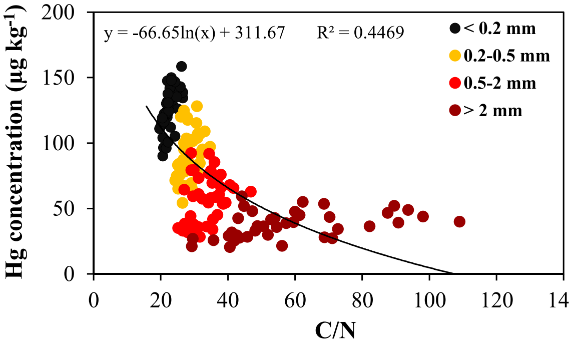

Fig. 2. Relationship between the concentration of Hg and the C/N ratio in different size classes of sediment.

by wildfires in Spain and Portugal [52,53]. These studies considered the accumulation of heavy metals because of the transport of fine particles and organic matter in burned areas.

In the present study, THg increased in all sediment size classes over the assessed period would be consistent with the wet and dry deposition, in the nearby burned area, of particulate-bound Hg (PBM) and gaseous elemental Hg (GEM). During the early stages after prescribed fire, the deposited Hg that contributed to the increase in Hg concentrations over time could be that released by biomass burning, as evidenced by previous studies [12,17,51,54]. At later post-fire stages, the Hg, wet or dry deposited, could have an additional origin not necessarily related to burning. Such an increase of THg in sediments was also found to account for a peak of particulate bound Hg in river waters crossing burned areas as a result of sediment mobilization [8]. However, besides local re-deposition of Hg, other processes like wetting-drying sediment conditions, mass loss and changes in soil organic matter quality or microbial activity, among others, could be simultaneously contributing to the accumulation of Hg in the sediments of the present study.

With the passage of time after fire, changes in the chemical composition of sediments may occur, which may affect their ability to retain Hg, especially those properties closely related to organic matter, since it has a strong biogeochemical affinity with Hg in terrestrial ecosystems [55]. All sediments size classes were enriched in total organic C with time post-fire, varying from 38 to 94 % enrichment when comparing the first and last rainfall events (Table 1). This trend agrees with previous studies in which the increase of C in sediments and soils from burned areas was attributed to partially burned plant debris [56,57] and the mobilization of C-rich ash [58]. In the present study, when sediment size fractions are individually evaluated, a positive correlation of THg and TOC was obtained for each of the four sediments size classes (for all cases $r_s > 0.405$ and $p < 0.01$), although the greatest Hg and C enrichment occurred in the class 0.5–2 mm.

Several explanations may justify the correlation of THg and C in sediments. Thus, the coating of organic matter on the sediment surface would increase its ability to adsorb Hg [50], whereas the mobilization of charred organic material could be carrying away legacy Hg [8]. Moreover, organic matter from surface soil layers become very recalcitrant and hydrophobic after fire [59], increasing its ability for Hg adsorption and hindering the desorption of Hg [60]. In this regard, two recent studies carried out in the same area could support the role of changing organic matter composition after fire in Hg accumulation. Thus, Fonseca et al. [28] revealed that new forms of C (pyrogenic C) can be transferred from burnt litter to mineral soil increasing its ability to adsorb Hg and being, at the same time, available for mobilization after rainfall events. On the other hand, Méndez-López et al. [10] reported that a prescribed fire had a significant impact on the quality of organic matter of soil samples, linking the occurrence of aromatic-like compounds after fire to Hg retention. However, further study of the organic matter composition of sediments would help to confirm how changes in organic matter quality may account for the temporal trend of THg concentrations in sediments mobilized after a prescribed fire.

3.3. Mobilization of Hg by sediment transport after rainfall

Soil loss by erosion is considered a direct consequence of prescribed fires and wildfires [15,22], being also the main responsible for the transference of Hg from terrestrial to freshwater ecosystems in remote areas [17,50,61–63]. The amount of Hg mobilized by sediment transport (Hg_{ST}) varied from 32 to 78 mg ha^{-1} (Fig. 3) after the different rainfall events, which is in the range of 30–100 mg Hg ha^{-1} year^{-1} estimated for Hg displaced by water erosion in topsoils in the NW of the Iberian Peninsula [64] and slightly higher than the range (10–13 mg Hg ha^{-1} year^{-1}) estimated for Hg export in burned catchments from USA [8,65]. However, several authors reported Hg export rates by erosion about three orders of magnitude higher in areas affected by wildfires [19,51,66].

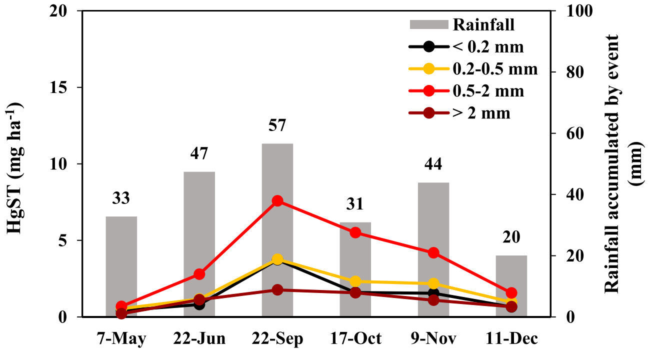

Fig. 3. Temporal variation of Hg mobilized (Hg_{ST}) by different sediment size classes (lines). Bars represent the amount of precipitation accumulated in each rainfall event (exact values indicated by numbers above the bars).

In NE Portugal, 5–10 mg Hg ha^{-1} year^{-1} could reach river basins by water erosion [64], therefore a significant amount of Hg mobilized through sediments in the present study might not reach nearby watercourses, but would be spread in the floodplains of aquatic ecosystems where the biogeochemical conditions would favour the conversion of Hg into highly toxic methyl-Hg [50,62,67–69]. The transformation of Hg mobilized by sediments in burned areas into methyl-Hg is still a matter of discussion. Sediments in which Hg is predominantly bound to organic matter, as it presumably occurs in the present study, often show a greater rate of methylation and bioavailability than other Hg forms [61,70]. On the contrary, Ku et al. [24] revealed that ashes originated by wildfires can sequester oxidized Hg species (HgII), transforming them in less reactive forms. More recently, Xu et al. [71] reported that the finest particle sizes of river sediments have a high potential for Hg adsorption due to their great specific surface area, making Hg less available for methylation.

Campos et al. [19] have shown the influence of slope in the mobilization of Hg in burned areas from Central Portugal. Similarly, Li et al. [63] identified landscape slope as a major driver of Hg transport to floodplain ecosystems and stream water, which was mainly due to slopes higher than 29 %, that were included in their study. In fact, they did not find significant differences among Hg in sediments from the experiments conducted in slopes of 13.2 % and 21.5 % [63], which are relatively similar to those of the current paper (6.9 % and 16.5 %, Fig. 1). Thus, Hg_{ST} in the study period did not differ statistically according to plot slope ($U = 190.0$; $p = 0.787$, $n = 40$), as the mean value from the lower slope plots (48.3 mg ha^{-1}) was similar to that from the steepest slope (48.6 mg ha^{-1}). It has been assumed that slight differences in the slope of the plots did not introduce statistically significant variations in Hg_{ST}, which agrees with Zema and Lucas-Borja [72], who showed that hillside slope is a key factor influencing surface runoff but not soil erosion [72].

The four size classes of sediment analysed contributed differently to Hg_{ST} during the study period ($H = 27.710$; $p = 0.000$; $n = 158$), with the 0.5–2 mm class accounting for 46 % of the total Hg transported (22.3 mg Hg ha^{-1}) after eight months since the prescribed fire (Fig. 3). The notable contribution of the 0.5–2 mm class to Hg_{ST} was to be expected, as it covers a wider size range than other size classes, and accounts for 50 % of the whole amount of sediment transported over the study period (Fig. S3). Finer size classes, such as 0.2–0.5 mm and < 0.2 mm, mobilized 10.9 and 8.8 mg Hg ha^{-1}, respectively, corresponding to 23 % and 18 % of total Hg_{ST}. The coarser size class (> 2 mm) mobilized 6.4 mg Hg ha^{-1} (13 % of Hg_{ST}). Considering all size classes, Hg_{ST} correlated positively with the load of sediments mobilized (ST; $r_s = 0.871$; $p = 0.000$; $n = 158$) but not with THg ($p > 0.05$), suggesting that Hg mobilization after fire was more dependent on erosion rates than on Hg concentration, a circumstance that has already been highlighted in previous studies [8,31,36,49,63]. Regarding sediment mobilization, special attention should be paid to the finer sediment classes because they are often enriched in heavy metals [37] and are more prone to reach streams than coarse fractions [73,74], even during low intensity rainfall events [75,76]. As an example of this issue, Jensen et al. [8] found a significant correlation between total suspended solids and particulate Hg in stream water from a burned catchment area.

The temporal variation of Hg_{ST} is highly dependent on erosion rates, which are expected to increase substantially in the absence of vegetation cover and with the occurrence of rainstorms after the fire [75,77]. The values of Hg_{ST} in each rainfall event correlated positively to the corresponding accumulated precipitation ($r_s = 0.430$; $p = 0.006$; $n = 40$), a correlation that also occured for all the individual sediments size classes ($p < 0.05$). The values of Hg_{ST} increased progressively during the first three rainfall events, following the patterns of accumulated precipitation and load of mobilized sediments (Fig. 3). This circumstance, which is also favoured by the absence of plant cover, coincides with a greater mobilization of Hg often observed during the first months after fire [8,31]. Moreover, in the same study area, Fonseca et al. [28] observed that the greater mobilization of sediments occurred during the first rainfall events. In addition to the lack of vegetation cover, these higher erosion rates during the first stages after burning would be favoured by the increased water repellency of soil particles, a common consequence of fire [78,79]. Soil repellency, combined with other factors (texture, structure, moisture, porosity, etc.) would explain higher erosion losses during the early stages after burning. The peak in $Hg_{ST,}$ with 16.8 mg ha^{-1} (35 % of the total Hg_{ST} in the whole study period), occurred during the third event (from June to September 2021), which is characterized by the highest accumulated precipitation (57 mm) as well as the higher load of mobilized sediments (239 kg ha^{-1}). During the following events, Hg_{ST} varied between 1.8 and 11.0 mg ha^{-1}, showing a decreasing trend over time after fire, even though the accumulated precipitation remained at similar levels as those of the first two events (20–44 mm; Fig. 3). This suggests that, in addition to accumulated precipitation, other factors can influence erosion rates, such as rainfall intensity, duration and frequency, and, consequently, they can also influence Hg mobilization [17,32,63]. Unfortunately, the lack of data on rainfall intensity in the present study precludes an assessment of its influence on Hg mobilization. However, previous research in the same study area noted a selective erosion of fine soil particles during the first rains after the fire and a progressive increase in the coverage of the soil surface by rock fragments, leading to a notable diminution in the soil loss rate [75,80]. After low severity fires, Béliveau et al. [31] also found a considerable loss of fine particles (< 0.063 mm) in areas affected by slash-and-burn. The decreasing pattern in Hg mobilization with time after fire agrees with the gradual decline in particulate Hg found by Jensen et al. [8], which was attributed to a diminution in the amount of precipitation received in the catchment. Although in the present study, the total rainfall accumulated over the eight months was lower than that reported by Jensen et al. [8], it could be enough to remove readily mobilizable Hg, resulting in legacy Hg mobilization even in the later post-fire stages.

The contribution of the individual sediment size classes to Hg_{ST} showed scarce variations throughout the events (Fig. S3), without a clear temporal trend (Fig. 3). The greatest contributions of the finer sediment classes (< 0.2 and 0.2–0.5 mm) to Hg_{ST} occurred during the first event with percentages of 22 % and 29 %, respectively, indicating the ability of these size classes to mobilize Hg at early post-fire stages. On the contrary, the smallest contribution of the 0.5–2 mm to the mobilization of Hg (38 %) occurred in the first event post-fire (Fig. S3).

4. Conclusions

The results suggest that a fraction of the Hg emitted during the prescribed fire is locally re-deposited, whereas ash and partially charred plant debris could also retain some of the Hg released during fire. This would justify the slightly higher levels of THg found in finer sediment fractions compared to those reported in a previous study for the organic horizons and the uppermost mineral soil layer (0–3 cm) of unburned soils located in the same site. The C/N ratio explained, to some extent, the trend of increasing THg as the size of sediment decreased. This may be due to the joint effect of a greater adsorption ability of the smaller particles and their increased availability of Hg binding sites as organic matter becomes more humified. The increasing trend of THg in sediments with time after fire would also be consistent with an increasing length of the exposure period to atmospheric Hg deposition. However, since total C showed a temporal trend parallel to that of THg, changes in the chemical composition of sediment organic matter during post-fire ageing, leading to a higher Hg retention capacity, cannot be ruled out.

The greater rates of Hg mobilization during the early post-fire events of precipitation imply the need to apply measures to mitigate soil erosion as soon as possible, which would reduce potential risk of toxicity from the arrival of Hg mobilized to aquatic ecosystems.

Finally, despite the low severity of prescribed fires, our results showed that they can generate a scenario in which Hg mobilization by sediments may occur, threatening the ecosystem services of terrestrial and aquatic areas in neighbouring burned zones. Therefore, much more attention should be paid to Hg mobilization in those areas where prescribed burning is often used as a tool to prevent the adverse effects of wildfires.

Environmental Implications

Mercury is a highly toxic pollutant of global concern. Forest ecosystems are sinks of mercury, but wildfires lead to the mobilization mercury to ecosystems vulnerable to its presence. This study aims to assess the mobilization of mercury with eroded sediments after a prescribed fire performed for forest management purposes in a scrubland from NE Portugal, a region highly threatened by wildfires. The results suggest that prescribed fires should be used with caution, as they result in the mobilization of mercury downslope. When mercury reaches water bodies, it could bioaccumulate in the food chain as methylmercury, becoming toxic for organisms.

CRediT authorship contribution statement

Melissa Méndez-López: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Manuel Arias-Estévez:** Visualization, Supervision, Resources, Project administration, Funding acquisition. **Tomás de Figueiredo:** Writing – review & editing, Visualization, Investigation, Data curation. **Juan Carlos Nóvoa-Muñoz:** Writing – review & editing, Writing – original draft, Visualization, Supervision, Resources, Project administration, Funding acquisition, Conceptualization. **Flora Alonso-Vega:** Writing – review & editing, Writing – original draft, Visualization, Supervision, Resources, Funding acquisition. **Lara Acemel-Míguez:** Writing – original draft, Visualization, Data curation. **Andrea Parente-Sendín:** Writing – original draft, Visualization, Methodology, Investigation, Data curation. **Israel Santos:** Visualization, Investigation, Data curation. **Felicia Fonseca:** Writing – review & editing, Visualization, Investigation, Data curation.

Declaration of Competing Interest

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

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2024.136657.

Data availability

Data will be made available on request.

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