Article

Elements' Content in Stream Sediment and Wildfire Ash of Suburban Areas in West Attica (Greece)

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Abstract: The composition of sediments and other materials occurring in streams, geochemical processes within the drainage basin, and various land uses are among the main factors influencing stream water composition. Stream sediment and wildfire ash samples were gathered from the area studied. The applied methodology consists of aqua regia and Diethylene-Triamine-Penta-Acetic acid (DTPA) chemical extraction; Cation Exchange Capacity (CEC), pH, and soil organic matter (OM) determination; a Geographic Information System (GIS) database; factor analysis; and determination of the contamination factor (CF) for the assessment of contamination degree. This study aimed to evaluate the elements' content in stream sediments of Kineta and Nea Peramos areas (West Attica, Greece) and investigate any relationship between elements (aqua regia and DTPA extracted) in stream sediment and ash in wildfire and flood-impacted areas. The stream sediments' properties, the bioavailable forms of copper (Cu), iron (Fe), manganese (Mn) and zinc (Zn), and the total content of other potentially toxic elements in wildfire ash samples, are discussed. This research estimated moderate contamination for Fe_{DTPA} , Mn_{DTPA} and Zn_{DTPA} in stream sediments of the study area. Contamination for Cu_{DTPA} and moderate contamination for Zn_{DTPA} in the stream sediments of the Nea Peramos area was recorded. Factor analysis results suggested that the contents of Fe_{DTPA} , Mn_{DTPA} , Cu_{DTPA} and Zn_{DTPA} in the study area's stream sediments may affect the chemistry of stream water.

Keywords: stream sediments; flood; wildfire; ash; DTPA-extraction; contamination factor

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1. Introduction

Stream and river sediments are an active part of the drainage basin, reflecting the geological and anthropogenic contamination sources [1–4]. The European Commission has recognized the importance of contaminated sediments as a water quality problem across the European countries. Trace elements may be added to an aquatic ecosystem either by natural (e.g., weathering of rocks and sediments) [5] or anthropogenic sources (wastewater, fossil fuel combustion, atmospheric pollutant deposition) [6,7]. Trace element contents originate from erosion, rock-water interaction and topsoil leaching [8,9], and riparian and floodplain interactions [10,11]. The concentration of trace elements accumulated in stream sediments may increase after a forest fire or a flooding event [12–16]. Swindle et al. [17] reported that water-soluble components of ash, which remains on the soil surface after fires, can leach into the soil and be transported through runoff, and particulate residues can be transported via erosion and runoff. Walling [18] and Glaser et al. [19] concluded that a strong interaction between stream sediment and suspended particles exists. Moreover, according to Walling [18] and Glaser et al. [19] the suspension load consequently represents a mixture of particles from different locations, mobilization history within catchment area, and anthropogenic or natural sources.

The study area is situated between latitudes $37^{\circ}57'$ and $38^{\circ}00'$ and longitudes $23^{\circ}12'$ and $23^{\circ}24'$. It includes Kineta and Nea Peramos areas, located about 55 and 35 km west of Athens, respectively (Figure 1).

The present research deals with stream sediment and geochemical properties in western Attica areas affected by wildfire (Kineta area) and flood (Nea Peramos area). The wildfire event on 23 July 2018 in the Kineta area burned houses and settlements and thousands of hectares of forestry, while the flooding event on 15 November 2017 in the Nea Peramos area resulted in significant damage to houses, stores, and the harbor (Figure 1). The primary geological formations in the areas studied include sandstones, dolomites, marls, marly formations with Mn-oxides, limestones, serpentinites, manganese ore deposits, peridotite bodies, schists, and volcanic rocks [20–26].

Figure 1. Location map of sampling sites of stream sediment in (a) Kineta area, and (b) Nea Peramos area. (Explanation for legend shown on Kineta area (1a)—1: Damaged buildings, 2: Possibly damaged buildings, 3: Wildfire impacted area, 4: Wildfire possibly impacted area). Modified from Google [27] and Copernicus EMS [28,29].

This study aimed to: (a) record the present elements' content in stream sediments in the area studied; and (b) evaluate the stream sediments' contamination in the study area.

2. Materials and Methods

2.1. Sampling

A total of twenty-six sediments (depth 0–10 cm) and five wildfire ash samples were gathered from streams located in the West Attica region (Figure 1). Twenty-two sediment samples and five ash samples were collected from the Kineta stream on 18, 24, and 26 September 2018 (Sampling sites S1–S22), and four sediment samples were collected from the Nea Peramos stream on 25 November 2017 (Sampling sites S23–S26) (Figure 1). Stream sediment samples were collected using a hammer and a polypropylene shovel. Ash samples were gathered in streams within the boundary of the wildfire-impacted area using plastic tools. All samples were stored in clean polyethylene (PE) bags.

2.2. Pretreatment of Sediment and Ash Samples

Pretreatment of stream sediment and wildfire ash samples was performed at the Laboratory of Geoenvironmental Science and Environmental Quality Assurance of the University of West Attica. All samples were dried indoors at room temperature ($<25\text{ }^{\circ}\text{C}$) for seven days and sieved first to $<2\text{ mm}$ and second to $<0.2\text{ mm}$ using a nylon sieve. Then, fine-grained sediment samples were stored in clean polyethylene bags for the determination of the extracted elements (Cu, Fe, Mn, and Zn) by Diethylene-Triamine-Penta-Acetic Acid (DTPA) and the measurement of sediment properties (pH, OM, and CEC). Contact with metallic components was avoided throughout laboratory procedures to prevent metal cross-contamination.

2.3. Grain Size Distribution

The size distribution of stream sediment particles (sand $< 0.02\text{--}2\text{ mm}$; silt $< 0.002\text{--}0.02\text{ mm}$; clay $< 0.002\text{ mm}$) was estimated using the method established by Bouyoucos [30] for eight stream sediment samples ($n = 8$).

2.4. Measurement of Physicochemical Stream Sediment Properties

CEC in stream sediment samples was determined by the barium chloride extraction method [31]. The detection limit of this method (LOD) is equal to 0.0214 mmol/L . According to this method [31], 2.5 g of each stream sediment sample was sieved at 0.2 mm , placed into 50 mL polyethylene tubes, and then 30 mL BaCl_2 was added. Then, the mixture of stream sediment and BaCl_2 was shaken for 60 min and inserted into a centrifuge for 10 min . After 10 min , the tubes were removed by centrifugation, and the supernatants were discarded. The concentrated BaCl_2 addition, agitation, centrifugation, and removal of supernatants were repeated two more times. After the third repetition, 30 mL of diluted BaCl_2 were added to tubes which were shaken overnight. A day later, the tubes were removed from the stirrer and inserted in the centrifuge for 10 min . Consequently, the supernatants were removed, and 30 mL MgSO_4 was added. The next day, centrifugation was carried out, and the supernatants were collected.

Stream sediment pH was determined in the water-saturated sample by adding distilled water using a Mettler Toledo FE20 Benchtop pH meter, which was calibrated using pH 4.01 and 7.01 buffer solutions (at $25\text{ }^{\circ}\text{C}$) provided by Merck® (Darmstadt, Germany). An in-house reference soil material (pH value 7.42) was applied to check the precision and accuracy of pH measurements found within international limits.

OM content (% wt) in stream sediment samples was measured by the dichromate oxidation method [32]. This method for the analysis of OM does not have a specific LOD. According to this method, 0.2 g of each fine-grained stream sediment sample was placed into 500 mL conical flasks, and 10 mL $\text{K}_2\text{Cr}_2\text{O}_7$ was added. After the mixture of sediment and $\text{K}_2\text{Cr}_2\text{O}_7$ was agitated, 20 mL concentrated H_2SO_4 was added, shaken for 1 min and left to stand for 30 min . Finally, approximately 200 mL of distilled water, 10 mL of concentrated H_3PO_4 , and 1 mL of diphenylamine indicator were added, and the excess of $\text{K}_2\text{Cr}_2\text{O}_7$ was titrated with FeSO_4 until the change of the indicator color from blue to green. During the OM measurement in sediments, a quality control sample was analyzed as reference material. This material is a soil sample for which the OM content was known (approximately $4\text{ wt } \%$) because it was measured many times at laboratory conditions.

The homogenized $< 0.2\text{ mm}$ stream sediment fraction was treated with the DTPA extraction method [33]. According to this method, 25 g of each stream sediment sample was placed in 250 mL conical flasks, and then 50 mL DTPA was added. After the mixture of sediment and DTPA was shaken for 120 min , the solution was filtered, and the supernatant was collected in polyethylene bottles. The supernatants were analyzed for Cu, Fe, Mn, and Zn by atomic absorption spectroscopy (AAS) using a Varian SpectrAA 220 model. The soil reference materials purchased from the NIST (National Institute of Standards and Technology), SRM 2710 and SRM 2711, and an in-house soil reference material were applied to monitor the procedure of chemical measurements. The element's content of soil reference

samples was routinely measured to determine analytical errors (accuracy < 10%). For every ten measurements of element content in samples, a duplicate measurement was performed to estimate the precision of chemical analysis (precision 2–3%). The LODs Cu_{DTPA} , Mn_{DTPA} , Fe_{DTPA} , and Zn_{DTPA} were 0.05, 0.125, 0.05 and 0.03 (mg L^{-1}), respectively.

2.5. Determination of Elements in Wildfire Ash

The wildfire ash samples were digested in a graphite heating block with an aliquot of the aqua-regia acid solution ($\text{HCl}:\text{HNO}_3$, 3:1 *v/v*). After cooling, the solution was diluted to 12.5 mL with deionized water, and mixed and analyzed for Ag, Al, As, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, Ga, Hg, K, La, Mg, Mn, Ni, P, S, Sb, Sc, Sr, Th, Ti, Tl, U, V, W, and Zn by ICP-AES (inductively coupled plasma-atomic emission spectrometry). The analytical results were corrected for inter-element spectral interferences. The laboratory measurements of elements in ash were conducted at Analytical Laboratories (ALS) OMAC Laboratories Limited in Galway, Ireland (ISO 17025:2005 Accredited) [34].

2.6. Data Treatment and Spatial Representation

The statistical treatment of the dataset was conducted using Microsoft Excel® (Redmond, Washington, DC, USA) and IBM SPSS® Statistics (International Business Machines Corporation; Statistical Product and Service Solutions; Armonk, NY, USA) for Windows. The univariate statistics of the geochemical (Cu_{DTPA} , Fe_{DTPA} , Mn_{DTPA} , and Zn_{DTPA}) and physicochemical properties (OM, CEC, and pH) datasets were calculated. The statistical analysis revealed that Cu_{DTPA} , Fe_{DTPA} , Mn_{DTPA} , and Zn_{DTPA} were positively skewed because the arithmetic means were more significant than the median. A logarithmic (base 10) transformation was applied to normalize this effect. The dataset of West Attica stream sediment was examined for outlying values using Tukey boxplots [35]. Outlying values were omitted, and the Cumulative Distribution Frequency (CDF) diagrams were constructed. Then, threshold values are calculated through CDF diagrams. The determination of the threshold values aims to investigate contamination due to anthropogenic and geological sources. The dataset of stream sediments was treated by the statistical techniques of R-mode factor analysis applying the varimax-raw rotational technique with Kaiser Normalization to determine the common origin of the elements. The multivariate statistics analysis was applied for CEC, OM, Cu_{DTPA} , Fe_{DTPA} , Mn_{DTPA} , and Zn_{DTPA} . The selection of the above-mentioned parameters was based on the anthropogenic activities, ore composition, and lithology of the West Attica region. All maps in this study were created using the ArcMap 10.4 GIS (ESRI®) software platform (Environmental Systems Research Institute; Redlands, CA, USA). The natural breaks procedure was applied to select the score classes. The statistical parameters of the aqua regia extracted elements in ash samples were evaluated by comparing their content to Sediment Quality Criteria.

3. Results and Discussion

3.1. Physicochemical Properties of Stream Sediments

The grain size analysis of the stream sediment samples showed that sand dominates in Kineta and Nea Peramos stream sediments, and silt and clay follow with lower percentages. Specifically, the median content values amount to 65.78% sand, 21.70% silt, and 11.38% clay in Kineta area stream sediments, and differ in percentage terms from the values of Nea Peramos area, which correspond to 13.63% clay, 21.43% silt and 64.94% sand. By plotting the above values on a triangular diagram, it was concluded that the stream sediments of the area studied can be classified as Sandy Loam (SL) [36]. The pH values in sediments of West Attica ranged from 7.30 to 8.28 and 7.17 to 7.68 for Kineta and Nea Peramos streams, respectively (Table 1).

Table 1. Descriptive statistics of the physicochemical dataset of stream sediments in the area studied.

Units	CEC	OM	pH	Fe _{DTPA}	Mn _{DTPA}	Cu _{DTPA}	Zn _{DTPA}
	cmol Mg ²⁺ Kg ^{−1}	wt%	-	mg Kg ^{−1}	mg Kg ^{−1}	mg Kg ^{−1}	mg Kg ^{−1}
West Attica stream							
<i>n</i>	26	26	26	26	26	26	26
min	4.04	1.10	7.17	10	11.7	0.4	0.8
max	38.7	13.3	8.28	92	93.7	8	53
mean	22.5 ± 8.2	7.73 ± 3.75	7.63 ± 0.22	50 ± 19	39.8 ± 22.1	2 ± 2	6 ± 11
median	22.2	7.27	7.63	47	32.7	0.8	3
Kineta area (burned sites)							
<i>n</i>	14	14	14	14	14	14	14
min	14.1	1.10	7.30	29	18.3	0.4	0.8
max	35.2	12.80	8.28	92	93.7	2	53
mean	24.8 ± 6.4	8.81 ± 3.69	7.67 ± 0.22	52 ± 15	42.7 ± 22.9	1 ± 0.6	8 ± 14
median	25.5	9.45	7.63	51	35.4	0.9	3
Kineta area (unburned sites)							
<i>n</i>	8	8	8	8	8	8	8
min	17.1	4.30	7.30	38	26.3	0.4	1
max	38.7	13.3	7.86	86	79.4	2	6
mean	25 ± 7.	8.00 ± 3.01	7.6 ± 0.2	52 ± 16	46.5 ± 19.5	0.7 ± 0.5	3 ± 2
median	22.8	7.10	7.67	46	41.0	0.6	2
Nea Peramos area (flooded sites)							
<i>n</i>	3	3	3	3	3	3	3
min	4.04	1.10	7.17	10	11.7	3	6
max	14.1	6.00	7.70	81	21.7	8	11
mean	9.23 ± 5.04	2.87 ± 2.72	7.45 ± 0.27	42 ± 36	16.5 ± 5.02	5 ± 3	8 ± 3
median	9.54	1.50	7.47	33	16.2	3	7
Nea Peramos area (non-flooded site)							
<i>n</i>	1	1	1	1	1	1	1
Measured Value	13.2	4.90	7.68	17	16.2	5	8

The results showed that sediment pH values were near neutral to slightly alkaline. The slightly alkaline pH value is due to the strong buffering capacity of the carbonates in the sediment fraction [37]. Values of pH recorded in stream sediments in wildfire-impacted areas did not show a significant statistical difference (p -value = 0.35 > 0.05). Specifically, a low pH increases from 7.63 to 7.67 was observed in sediment samples (Table 1). This increase is because the wildfire-impacted areas contained burned OM particles, either low or high quantities, which are transported in streams, increasing sediment pH. According to Faria et al. [38], the burned OM is derived from the oxidation of soil OM and the loss of hydroxyl groups (OH[−]), which is provoked by the wildfire event and the decomposition process of organic acids. The mean pH value in stream sediment of the non-flood-impacted area was 7.68 (Table 1). Moreover, Vincent et al. [39] reported a decrease in pH value in sediments collected from the flood-impacted area of Asaba (Nigeria), which was explained by acid production during sediment resuspension in floodwater, while alkaline sediment decreases pH, and acid sediment increases pH, during a flooding event.

Organic matter content in sediment ranged from 1.10 to 13.30 wt% and 1.10 to 6.04 wt% in Kineta and Nea Peramos streams, respectively (Figures 2 and 3). A comparison between the mean value of OM in the sediment of wildfire impacted (8.00 wt%) and non-impacted areas (8.81 wt%) of Kineta revealed a non-significant statistical increase (p -value = 0.29 > 0.05), which is explained by the presence of ash removed from forest soil after a wildfire and transported into streams. The OM content in stream sediment of flood-impacted areas of Nea Peramos was lower than the OM content in stream sediment of non-impacted areas because soil OM such as organic acids or humus is diluted in water, resulting in a decrease in its concentration in the particulate phase [39]. Moreover, De Marco et al. [40], who performed experimental fires in a Mediterranean maquis (south-western Italy), reported that burned soils presented higher concentrations of OM than control soils over the whole period of study. In Nea Peramos streams, the flood-impacted area showed high OM content in sediment (Figure 3).

Figure 2. Symbol plot of OM content in stream sediments of Kineta area compared with wildfire-impacted area (a) and geology (b). (Legend and explanations for 2a are shown on caption of Figure 1a; explanation for formations shown on the geological map of Kineta area (Figure 2b)—1: Sandstones, marls, marly formations, conglomerates of Upper Miocene; 2: Marls, sandstones of Miocene; 3: Sandstones, dolomites of Upper Triassic; 4: Urban area). Modified by Bezes [20,21], Copernicus EMS [28,29].

Cation Exchange Capacity (CEC) ranged from 14.1 to 38.7 cmol Mg²⁺ Kg^{−1} and from 4.04 to 14.1 cmol Mg²⁺ Kg^{−1} in stream sediments of Kineta and Nea Peramos areas, respectively (Table 1). According to the classification proposed by Hazelton and Murphy [41], the CEC values of stream sediments in the Kineta area are classified from moderate to high, whereas CEC values of stream sediments are classified as low for the Nea Peramos area. Moreover, the comparison of CEC mean values between non-wildfire-impacted (24.7 cmol Mg²⁺ Kg^{−1}) and impacted (24.8 cmol Mg²⁺ Kg^{−1}) areas showed an increase of 0.32%, which is explained by the increase in negative charges and consequently the retention of more cations [42,43], but this difference was not statistically significant (p -value = 0.49 > 0.05). However, an analysis of the sampling sites of the Kineta area

showed that some sites presented low CEC, whereas OM content in the sediment was high. This is attributed mainly to the clay content in sediment because the CEC value is controlled by, among other factors, the negative charges of clay minerals [44]. Moreover, concerning the Nea Peramos stream sediments, a significant decrease in CEC was observed in non-flood-impacted ($13.2 \text{ cmol Mg}^{2+} \text{ Kg}^{-1}$) compared to impacted areas ($9.23 \text{ cmol Mg}^{2+} \text{ Kg}^{-1}$), which can be attributed to the low OM content in sediments of flood-impacted areas [45].

Figure 3. Symbol plot of OM content in stream sediments of Nea Peramos area compared with satellite image (a) and geology (b). (Explanation for geological formations shown on the geological map of Nea Peramos (Figure 2b)—1: Sandstones, marls, marly formations, conglomerates of Upper Miocene; 2: Schists; 3: Limestones Middle-Upper Cretaceous; 4: Limestones, dolomites; 5: Urban area), Modified by Bezes [20,21], Google [27] and Copernicus EMS [28,29].

3.1.1. DTPA-Extractable Elements Content in Stream Sediments

The mean contents of Cu_{DTPA} and Zn_{DTPA} were higher in stream sediment in wildfire-impacted areas compared to non-impacted areas (Table 1). However, the statistical difference of these elements was not significant (p -values = 0.11 and 0.09 for Cu_{DTPA} and Zn_{DTPA} , respectively). The mean value of Mn_{DTPA} content was lower in the sediment of wildfire-impacted areas than areas that were non-impacted by wildfire, without statistical significance (p -value = $0.34 > 0.05$). The Mn_{DTPA} content in soils affected by wildfire can be attributed to the decomposition of forest species leaves, which releases Mn [46]. Moreover, the mean values of Fe_{DTPA} and Mn_{DTPA} content in stream sediment were higher in areas impacted by flooding compared to Fe_{DTPA} and Mn_{DTPA} content in sediment of area not impacted by flooding, whereas the mean values of Cu_{DTPA} and Zn_{DTPA} in stream sediment collected from flood-impacted areas were decreased compared to the corresponding values in stream sediments of non-impacted areas. According to the univariate statistical analysis, the threshold values of Fe_{DTPA} , Mn_{DTPA} , Cu_{DTPA} , and Zn_{DTPA} of the West Attica stream sediments were extracted from the Cumulative Frequency Diagrams (CDF) using criteria as breaks and inflection points (Figure 4). Threshold values for Cu_{DTPA} , Fe_{DTPA} , Mn_{DTPA} , and Zn_{DTPA} were 1, 38, 24.8, and 5 mg Kg^{-1} , respectively (Figure 4).

Figure 4. Cumulative probability plots (CDF diagrams) for (a) Cu_{DTPA} ; (b) Fe_{DTPA} ; (c) Mn_{DTPA} ; and (d) Zn_{DTPA} of the stream sediments of the study area ($n = 26$) (red dots demonstrate threshold values).

The contamination factor (CF) was estimated by the average and threshold value of each element according to the following mathematical equation proposed by Hakanson [47]:

$$\text{CF} = \frac{C_m}{C_n} \quad (1)$$

where C_m is the average content of trace metals, and C_n is the threshold concentration of each element.

The results showed that CF varied from 1 to 3 in Kineta stream sediments for Fe_{DTPA} , Mn_{DTPA} , and Zn_{DTPA} and $\text{CF} < 1$ in Nea Peramos stream sediments for Fe_{DTPA} and Mn_{DTPA} . Conversely, the CF of Cu_{DTPA} was classified as significant contamination ($3 < \text{CF} < 6$) in Nea Peramos stream sediments, whereas it was found to be low in Kineta sediments (Table 2).

Table 2. Stream sediment contamination degree (C_{deg}) of Kineta and Nea Peramos areas.

	Cu_{DTPA}	Fe_{DTPA}	Mn_{DTPA}	Zn_{DTPA}	C_{deg}
Mean of Kineta Streams	0.9	52	44.1	6	
Mean of Nea Peramos Streams	5	35	16.4	8	
Kineta CF	0.9	1.37	1.78	1.2	5.25
Nea Peramos CF	5	0.920	0.661	1.6	8.18

3.1.2. Elements Content in Ash

The most abundant element in ash collected in the streams of the area studied was Ca (17%), with Mg (4.03%), Fe (2.40%), and Al (1.21%) also present in elevated concentrations. In contrast, the elements found in lowest contents were Cd ($0.65 \mu\text{g Kg}^{-1}$), Be ($0.65 \mu\text{g Kg}^{-1}$), and Sb ($2 \mu\text{g Kg}^{-1}$) (Table 3).

The mean values in ash collected in the streams of the study area for Ca, Cd, Co, Cr, Cu, Fe, Ga, La, Mg, Ni, S, Sc, and Sr were higher than the median values in ash collected in the drainage basin of the area studied, as reported by Alexakis [12] (Table 3), highlighting the variation in element concentration within the ash. Wan et al. [48] reported that the median value of trace elements content in ash samples collected from the southern California Thomas fire were the following (in mg Kg^{-1}): As (5.39), Cd (0.77), Cr (50.7), Cu (74), Ni (33.3), and Zn (225), which are higher only for Cu and Zn compared to the corresponding mean values of the study area (Table 3). The oxide and hydroxides of Ca, Mg, and P were abundant in ash samples collected in the area studied. Similar findings were reported by Perreira and Ubeda [49] and Silva et al. [50], who studied wildfire ash

composition in the Lisbon region (Portugal) and District of Aveiro (Portugal), respectively. The effect of ash leaching on stream chemistry in the Kineta area may increase, among others, the K^+ content in seawater of Saronic Gulf. Despite the fact that any increase in the K^+ content in seawater would likely be transient, it may influence the biological activity along the coast of the Kineta area. Swindle et al. [17], who studied the effects of wildfires and ash leaching on stream chemistry in Southern California, reported that direct input of large amounts of stream sediments and ash that have an elevated content of extractable K^+ may increase the K^+ content in seawater. According to Swindle et al. [17], increases in the K^+ content in seawater of the California coast may influence marine organisms. Because the ash samples collected in the Kineta area were not chemically analyzed for the water-soluble concentration of major and trace elements, the solubility of elements in six composite ash samples provided by Harper et al. [51] was adopted. According to this convention, the following water-soluble concentrations of elements in ash of the Kineta area were estimated as follows: Al (<0.002%), As (0.09–0.67 mg Kg⁻¹), Ca (0.01–0.61%), Cd (<0.02 mg Kg⁻¹), Cu (0.16–4.13 mg Kg⁻¹), Fe (0.001%), Mg (<1.03%), Mn (0.05–3.46 mg Kg⁻¹), Ni (2.06–21.39 mg Kg⁻¹), and Zn (<8.64 mg Kg⁻¹). The calculated water-soluble elements suggest that a noticeable amount of potentially toxic elements is released by rainwater from wildfire ash and finally transported to Saronic Gulf.

Table 3. Descriptive statistics of elements concentration in the ash of Kineta area ($n = 5$) compared to the median content of wildland ash of Kineta area [12] and corresponding levels provided from the literature [52,53]; DL: Detection limit; SD: Standard deviation; n.d.: not detected.

Element	DL	Mean	SD	Wildland Ash [12]	Mean Sediment [52]	Sediment Quality Criteria [53]
Al (wt %)	0.01	1	0.3	1.52	10.2	-
As (mg Kg ⁻¹)	2	9	3	12	10.1	6
Be (mg Kg ⁻¹)	10	0.7	0.07	0.7	2.03	-
Ca (wt %)	10	17	5	14.25	5.81	-
Cd (mg Kg ⁻¹)	0.5	0.7	0.1	0.60	0.527	0.6
Co (mg Kg ⁻¹)	1	28	21	14	11.20	-
Cr (mg Kg ⁻¹)	1	131	77	72	92.80	26
Cu (mg Kg ⁻¹)	1	32	28	23.5	22.10	16
Fe (wt %)	0.01	2.4	1.1	2.24	2.25	2.12
Ga (mg Kg ⁻¹)	10	10	0	n.d.	12.1	-
K (wt %)	0.01	0.4	0.2	0.49	2.08	-
La (mg Kg ⁻¹)	10	12	4	10	41	-
Mg (wt %)	0.01	4	1	3.07	1.77	-
Mn (mg Kg ⁻¹)	5	532	139	612	716	460
Ni (mg Kg ⁻¹)	1	557	421	159	35.20	16
P (mg Kg ⁻¹)	10	2208	2307	1095	0.174	-
S (wt %)	0.01	0.09	0.08	0.10	0.092	-
Sb (mg Kg ⁻¹)	2	2	0	2	1.07	-
Sc (mg Kg ⁻¹)	1	4	1	3.5	-	-
Sr (mg Kg ⁻¹)	1	174	146	104	171	-
Ti (wt %)	0.01	0.02	0	0.03	0.685	-
V (mg Kg ⁻¹)	1	26	2	28.5	68.3	-
Zn (mg Kg ⁻¹)	2	108	78	118.5	98	120

The mean concentrations for Ca, Co, Cd, Cr, Cu, Fe, Mg, Ni, P, Sr, and Zn in the ash of the study area are higher than the corresponding values in the mean sediment reported by Salminen et al. [52] (Table 3). The mean contents for As, Cd, Cr, Cu, Fe, Mn, and Ni in wildfire ash in the Kineta area exceed the Sediment Quality Criteria [53] (Table 3). Therefore, wildfire ash constitutes a contamination source for the stream sediments of the area studied.

3.1.3. Evaluating Elements' Contamination Sources in Stream Sediments

Factor analysis (KMO = 0.678 and Barlett test <0.001) with principal component extraction and varimax rotation with Kaiser Normalization explained in terms of two factors the distribution of the element in the stream sediments of the study area ($n = 26$). The total cumulative variance of the studied dataset was 80.8% (Table 4).

Table 4. Varimax rotated component loadings of two factors and variance for West Attica stream sediments ($n = 26$).

Variable	Factor 1	Factor 2	Communality
Cu _{DTPA}	-	0.889	0.834
Fe _{DTPA}	0.833	-	0.694
Mn _{DTPA}	0.817	−0.337	0.781
Zn _{DTPA}	-	0.933	0.873
OM	0.901	-	0.832
CEC	0.884	-	0.836
Variance	3.00	1.85	4.85
% Variance	50.0	30.8	80.8

Factor 1 represents 50% of the total variance with high positive loadings (greater than +0.800) for Fe_{DTPA}, Mn_{DTPA}, OM, and CEC (Table 4). Factor 1 revealed the relation existing between OM and CEC. Alexakis et al. [54] conducted a soil geochemical survey in the wildfire-impacted area of Kineta and reported the same existing relation between OM and CEC. Furthermore, many researchers in other case studies [55,56] have also reported a similar relation. Iron and Mn are known to occur in the marl formations outcropping in the Kineta area [20,21,54]; therefore, Factor 1 is attributed mainly to the contribution of wildfire events to the remobilization of Mn oxides. Many researchers have reported similar findings [12,54,57–60] for Mn content in forest soil after wildfires. Gamvroula et al. [22] reported that the presence of Mn in soil and water at the Megara basin can be attributed to the Mn oxides present within Neogene deposits and the weathering products of Neogene deposits. Higher positive loadings (greater than +0.800) for Fe_{DTPA}, Mn_{DTPA}, OM, and CEC were recorded in the Kineta area (Figure 5). Lower positive loadings for Fe_{DTPA}, Mn_{DTPA}, OM, and CEC were observed in flood-impacted and non-impacted areas (Figure 6).

Factor 2 represents 30.8% of the total variance with high positive loadings (greater than +0.850) for Cu_{DTPA} and Zn_{DTPA} and low negative loading (−0.377) for Mn_{DTPA}. The high positive loadings of Cu_{DTPA} and Zn_{DTPA} showed that these elements play a principal role in explaining Factor 2. The higher positive loadings (greater than +0.370), which correspond to the high concentration of Cu_{DTPA} and Zn_{DTPA}, were observed in the wildfire-impacted area of Kineta (Figure 7). Similarly, stream sediment samples collected from Nea Peramos streams present higher positive loadings (greater than +0.370) (Figure 8). This means that the Cu_{DTPA} and Zn_{DTPA} contents recorded in the stream sediments of the study area can be attributed to the area's geogenic sources.

Figure 5. Symbol plot of Factor 1 scores ($\text{Fe}_{\text{DTPA}}\text{-Mn}_{\text{DTPA}}\text{-OM-CEC}$) for Kineta stream in comparison with soil condition (a) and geology (b). (Legend and explanations for 5a are shown in caption of Figure 1a; explanation for geological formations is shown in Figure 2b). Modified by Bezes [20,21] and Copernicus EMS [28,29].

Figure 6. Symbol plot of Factor 1 scores ($\text{Fe}_{\text{DTPA}}\text{-Mn}_{\text{DTPA}}\text{-OM-CEC}$) for Nea Peramos stream in comparison with satellite image (a) and geology (b). (Explanation for geological formations is shown in Figure 3b). Modified by Bezes [20,21], Google [27] and Copernicus EMS [28,29].

Figure 7. Symbol plot of Factor 2 scores ($\text{Cu}_{\text{DTPA}}\text{-Zn}_{\text{DTPA}}\text{-Mn}_{\text{DTPA}}$) for Kineta stream in comparison with soil condition (a) and geology (b). (Legend and explanations for 7a are shown in caption of Figure 1a; explanation for geological formations is shown in Figure 2b). Modified by Bezes [20,21] and Copernicus EMS [28,29].

Figure 8. Symbol plot of Factor 2 scores ($\text{Cu}_{\text{DTPA}}\text{-Zn}_{\text{DTPA}}\text{-Mn}_{\text{DTPA}}$) for Nea Peramos stream in comparison with satellite image (a) and geology (b). (Explanation for geological formations is shown on Figure 3b). Modified by Bezes [20,21], Google [27] and Copernicus EMS [28,29].

Factor analysis results revealed high positive loadings of Fe_{DTPA} , Mn_{DTPA} , Cu_{DTPA} , and Zn_{DTPA} , which suggested that these element contents in the study area's stream sediments may affect the chemistry of stream water. Swindle et al. [17], who studied the effects of wildfires and ash leaching on composition of stream water in the Santa Ynez Mountains (Southern California, USA), reported that, after the wildfire, the concentration of all major cations (K^+ , Na^+ , Ca^{2+} , Mg^{2+}) increased in stream water collected near the fire perimeter following the first storm of the season.

4. Conclusions

The Kineta streams presented a low degree of contamination, whereas Nea Peramos streams were moderately contaminated. The C_{deg} of Kineta streams ($C_{\text{deg}} = 5.18$) was lower than C_{deg} of Nea Peramos streams ($C_{\text{deg}} = 7.84$). A GIS elaboration showed that the contamination in Kineta streams is due to both wildfire and geogenic sources such as OM and Fe-Mn oxides for Mn_{DTPA} and Fe_{DTPA} , whereas Cu_{DTPA} and Zn_{DTPA} contamination in Kineta streams is mainly attributed to wildfire. Moreover, Cu_{DTPA} and Zn_{DTPA} content in stream sediment can be attributed to the decomposition of OM and the dissolution of Mn oxides. Wildfire ash in the study area contains potentially toxic elements (As, Cd, Cr, Cu, Fe, Mn, and Ni), which show content higher than the Sediment Quality Criteria. Moreover, the mean ash concentrations for Ca, Co, Cd, Cr, Cu, Fe, Mg, Ni, P, Sr, and Zn exceed the corresponding values of the mean sediment. The geochemical composition of stream sediments collected in the area studied is controlled mainly by events of wildfire and flood, in addition to the geogenic sources of the West Attica region, resulting in the variation in the elements' distribution in stream sediments. Ash in the Kineta streams may increase the potentially toxic elements' content in stream sediment and, subsequently, contaminated material may be transported to the Saronic Gulf.

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