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# Evaluation of Soil Heavy Metal Contamination and Potential Human Health Risk inside Forests, Wildfire Forests and Urban Areas

**Speranza Claudia Panico [1,2], Lucia Santorufo [3,4], Valeria Memoli [3,*], Francesco Esposito [3], Giorgia Santini [3], Gabriella Di Natale [5], Marco Trifuoggi [6], Rossella Barile [7] and Giulia Maisto [3,4]**

1 Dipartimento di Scienze Agroalimentari, Ambientali e Animali, Università degli Studi di Udine, Via delle Scienze 206, 33100 Udine, Italy; speranza.panico@uniud.it
2 National Biodiversity Future Center, NBFC, Piazza Marina 61, 90133 Palermo, Italy
3 Dipartimento di Biologia, Università degli Studi di Napoli Federico II, Via Cinthia, 80126 Napoli, Italy; lucia.santorufo@unina.it (L.S.); francesco.esposito1991@live.it (F.E.); giorgia.santini@unina.it (G.S.); gmaisto@unina.it (G.M.)
4 BAT Center—Interuniversity Center for Studies on Bioinspired Agro-Environmental Technology, University of Naples Federico II, 80055 Portici, Italy
5 CeSMA-Centro Servizi Metrologici e Tecnologici Avanzati, Università degli Studi di Napoli Federico II, Corso Nicolangelo Protopisani, 80146 San Giovanni a Teduccio, Italy; gabriella.dinatale@unina.it
6 Dipartimento di Scienze Chimiche, Università degli Studi di Napoli Federico II, Via Cinthia, 80126 Napoli, Italy; marco.trifuoggi@unina.it
7 Parco Nazionale del Vesuvio, Via Palazzo del Principe c/o Castello Mediceo, 80044 Ottaviano, Italy; rbarile@epnv.it
* Correspondence: valeria.memoli@unina.it; Tel.: +39-081679111

**Abstract:** Recently, due to rapid industrialization and urbanization, many studies have focused on the assessment of soil metal contamination. The present research aimed to investigate the concentration of potentially hazardous elements (Al, Cr, Cu, Fe, Mg, Mn, Ni, Pb, V and Zn) in three different areas (forest, wildfire forest and urban) of the Mediterranean region. Contamination levels were further assessed calculating different indices: contamination factor (CF), geo-accumulation index (GI) and ecological risk (E). The results showed high concentrations of V, Mg and Mn in forest and wildfire forest soils, as well as high concentrations of Al, Fe, Ni, Pb and Zn in urban soils. According to the contamination indices, Pb exceeded the background level in both wildfire forest and urban soils. According to human risk assessment, inhalation appeared the main route of exposure of metals in soils, especially for children. The overall cancer risk was higher than the safe level, especially for Pb. Different relationships were found between the contamination indices and the potential risk of carcinogenic effects according to the diverse metal concentrations. Particularly, wildfire soils showed human health risks mainly linked to Pb, Cu and Cr contaminations, due to human activities, and Ni, due to both anthropogenic and pedogenetic input. Instead, the urban soils showed that Zn contamination, mainly related to urban traffic, influenced the potential carcinogenetic risk in this area. The carcinogenic risk was higher than acceptable values for all the metals assessed. These findings highlighted the need to develop further management practices to protect soils from metal pollution and reduce human health risks.

**Keywords:** Mediterranean region; carcinogenic risk; soil contamination indices; potential toxic elements; land uses

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## 1. Introduction

Recently, soil metal pollution has been widely recognized as a serious environmental problem due to rapid urban expansion, intensive agricultural practices and natural processes. In fact, soils have become the receivers of various pollutants by nearby industrialized and densely populated areas [1–3]. This may lead to soil metal accumulation and

consequently to a decline in soil quality with consequences on the entire ecosystem and on human health [4–6]. Among the contaminants that can potentially be found in soils, the most relevant hazardous metals are Cr, Cu, Ni, Zn and Pb because they can be transferred from soil directly in aquatic and terrestrial food chains/webs, causing important implications for wildlife and human health [7,8]. Particularly, the human risks associated with metals in soil may be due to ingestion, inhalation or dermal contact and their long-term toxicity effect; exposure to even low levels of metals can result in neurological and physical degenerative processes and cancer over time [9,10].

Moreover, the widespread conversion of forests into human-made ecosystems that causes soil degradation [11] has strongly influenced soil's physico-chemical properties, enhancing soil metal contamination and, in turns, triggering its ability to preserve environmental quality and human health [12,13]. Above all, the Mediterranean regions are the most vulnerable due to their semi-arid conditions which, together with frequent fires and rapid urbanization, can transport metals in the soil by atmospheric deposition and cause accumulation even far from the source of metal contamination [14,15]. However, the number of elements in soils depends on pedogenesis and parent material quality, as well as on further input due to the deposition of the burnt epigeal [16,17].

Soil contamination degree and human health risk have been assessed using several indices, such as the contamination factor (CF), the ecological risk (E) and the geo-accumulation index (GI), which are also widely used to evaluate the contamination source of soil metals pollution [18] and their potential ecological risk, in association with the hazard quotient index (HQI) and the aggregate carcinogenic risk (ACR), known to evaluate the potential human health risk deriving from exposure to metals [19].

As the numerous intersections between soil metal content and human health occur, the main goal of the present research was to contribute to filling in the current gap in knowledge about the potential human health risk deriving from exposure to contaminated soils. Particularly, the research evaluated the following: (i) the concentrations of different metals (Al, Cr, Cu, Fe, Mg, Mn, Ni, Pb, V and Zn) in soils of forests, forests after a wildfire and urban areas; (ii) the soil metal pollution using three indices, namely the contamination factor, ecological risk and geo-accumulation index (CF, E and GI, respectively); (iii) the human health risk (non-carcinogenic and carcinogenic) linked to five metals (Cu, Cr, Ni, Pb and Zn) through the use of the hazard quotient index and the aggregate carcinogenic risk (HQI and ACR, respectively); and (iv) the relationships among soil contamination indices (CF, E and GI) and human health risk indices (HQI and ACR).

The present research was performed in a Mediterranean area, which is a mosaic of different site typologies (forest, wildfire forest and urban) and usually is densely populated; moreover, the characteristic peculiar climatic conditions and frequent wildfires act as multiplying factors of soil contamination and degradation.

Therefore, the novelty of the present research is that it represents a first attempt to simultaneously investigate the impact of metal contamination on soil quality and on human health risk inside three land patches (forest, wildfire forest and urban). The findings can be used as a starting point for developing models to monitor and mitigate human health risk in a large range of different widespread environments subjected to the same source of soil contamination.

## 2. Materials and Methods

### *2.1. Description of the Study Area*

The study was established in the surroundings of Naples (Southern Italy), characterized by the typical Mediterranean climate. The metropolitan area of Naples has about 4 million inhabitants, one of the most densely populated areas in Europe, and has a population density of 2632 in hab/km$^2$ (https://worldpopulationreview.com/, accessed on 30 June 2022). The soils were collected at three areas in the surroundings of Naples: forest (F), forest after a wildfire (FW) and urban (U) areas. Both forest sites (F and FW) are located inside the Vesuvius National Park (8482 ha, Campania, Italy) at 12 km SE far from the city of

Naples. The Vesuvius National Park is characterized by Mediterranean vegetation, mainly dominated by trees such as holm oaks, pines, maples and alders. The urban sites (U) were located in a densely populated area (ca. 72,500 inhabitants), comprehending four municipalities (Pomigliano d'Arco, Castello di Cisterna, Brusciano and Mariglianella) of Naples. The investigated urban sites are covered by holm oak trees, and they are located along urban roads with highly intense vehicular traffic and are surrounded by small metallurgical industries.

### 2.2. Soil Sampling

The sampling design is shown in Figure 1. Soil sampling was performed in Spring 2018, a year after a huge wildfire that interested part of the sampling area, within two consecutive days and after seven days without rainfall to minimize variability due to the climatic conditions.


**Figure 1.** Map of the study area.

The surface (0–10 cm) soils were sampled at 6 sites for each area: forest, forest after a wildfire and urban (namely F, WF and U, respectively). At each of the 18 sites, 8 subsamples of surface soil, after litter (for F and U) or ash removal (for WF) were collected and mixed together in order to obtain a composite and homogeneous sample. The fresh soil samples were put in sterile flasks and transported on ice to the laboratory, where they were sieved through a mesh (<2 mm) and prepared for analyses. All the investigated soils are classified as leptic–vitric Andosols [20].

### 2.3. Sample Preparation and Analyses

The sieved soil samples were analyzed for pH, water content (WC), organic matter (OM) and total C and N concentrations. Soil pH was measured in a soil: distilled water suspension (1:2.5 = *w*:*w*) by an electrometric method, whereas WC was determined gravimetrically by drying fresh soil at 105 °C until a constant weight was reached. The element concentrations were evaluated on oven-dried (75 °C) and pulverized samples (Fritsch Analysette Spartan 3 Pulverisette 0). Particularly, C and N concentrations were measured by an elemental analyzer (Thermo Finnigan, Mundelein, IL, USA, CNS Analyzer); the other elements (Al, Cr, Cu, Fe, Mg, Mn, Ni, Pb, V and Zn) were measured on previously digested samples by hydrofluoric acid (50%) and nitric acid (65%) at a ratio of 1:2 (*v*:*v*) in a microwave oven (Milestone-Digestion/Drying Module mls 1200) by inductively coupled plasma mass spectrometry (ICP-MS Aurora M90, Bruker). Accuracy of the element measurements was checked by concurrent analysis of standard reference material (BCR CRM

142R—Commission of the European Communities 1994). The overall element recovery ranged from 80 to 120% for all the investigated soil samples.

All laboratory analyses were performed in triplicate.

### *2.4. Soil Metal Pollution Assessment*

In order to assess the soil metals pollution and the degree of anthropogenic influence in each area, different contamination indices were calculated. These indexes of potential contamination were calculated by the normalization of one metal concentration in the topsoil with respect to the concentration of a reference element. The metal contents of forest sites (F) were used as a geochemical background since this area was located far from any contamination sources, and the soil showed similar pedogenetic properties to the other two (WF and U) considered in this study (leptic–vitric Andosols).

#### 2.4.1. Contamination Factor

The contamination factor (CF) is a quantification of the degree of contamination relative to either the average crustal composition of a respective metal or to the measured background values from geologically similar and uncontaminated area. The CF value of each element was calculated according to Buat-Menard and Chesselet [21], using Equation (1):

$$CF = Cn/Cb \quad (1)$$

where Cn represents the concentration of the element in the soil samples collected at WF and U sites, and Cb (background concentration) represents the concentration of the same element in soils collected at F sites (Table 1). The metal concentrations in forest soils (F) were used as background in order to refer to the level of contamination of the investigated human-impacted areas (WF and U) for the same kind of soil (leptic–vitric Andosols). The CF < 1 values indicate low contamination, 3 ≤ CF ≤6 signifies considerable contamination, while CF > 6 is evidence of high contamination [21].

**Table 1.** Mean values (±s.e.) of metals (Al, Cr, Cu, Fe, Mg, Mn, Ni, Pb, V and Zn) expressed as mg $kg^{-1}$ d.w. of the soils collected at forest (F), burned forest (WF) and urban (U) areas. Different letters indicate statistically significant differences among metal concentrations under different soils (ANOVA test, at least, $p < 0.05$).

| | Sites | | |
|---|---|---|---|
| **Metals** | **F** | **WF** | **U** |
| **Al** | 19,969 $^{B}$ | 12,508 $^{B}$ | 28,390 $^{A}$ |
| (mg $kg^{-1}$ d.w.) | ±2553 | ±1910 | ±3793 |
| **Cr** | 53.0 $^{A}$ | 40 $^{B}$ | 38 $^{B}$ |
| (mg $kg^{-1}$ d.w.) | ±0.4 | ±0.4 | ±0.3 |
| **Cu** | 70.6 $^{A}$ | 64.3 $^{B}$ | 63.8 $^{B}$ |
| (mg $kg^{-1}$ d.w.) | ±3.2 | ±2.5 | ±2.1 |
| **Fe** | 26,967 $^{A}$ | 19,053 $^{B}$ | 702 $^{C}$ |
| (mg $kg^{-1}$ d.w.) | ±2157 | ±468 | ±43.5 |
| **Mg** | 17,007 $^{A}$ | 9422 $^{B}$ | 12,261 $^{B}$ |
| (mg $kg^{-1}$ d.w.) | ±1912. | ±1042 | ±928 |
| **Mn** | 911.3 $^{A}$ | 838 $^{A}$ | 421 $^{B}$ |
| (mg $kg^{-1}$ d.w.) | ±23.8 | ±35.6 | ±0.0 |
| **Ni** | 52 $^{A}$ | 45 $^{A}$ | 21 $^{B}$ |
| (mg $kg^{-1}$ d.w.) | ±0.2 | ±0.2 | ±0.8 |
| **Pb** | 48.5 $^{B}$ | 35.8 $^{B}$ | 83.8 $^{A}$ |
| (mg $kg^{-1}$ d.w.) | ±4.1 $^{B}$ | ±2.6 | ±14.9 |
| **V** | 197.2 $^{A}$ | 189.8 $^{A}$ | 111.6 $^{B}$ |
| (mg $kg^{-1}$ d.w.) | ±7.7 | ±9.0 | ±7.6 |
| **Zn** | 29 $^{B}$ | 23 $^{B}$ | 100 $^{A}$ |
| (mg $kg^{-1}$ d.w.) | ±0.4 | ±0.2 | ±4.6 |

2.4.2. Geo-Accumulation Index

The geo-accumulation index (GI) has been widely used in the assessment of pollution status of soils since the late 1960s [22–24]. The GI was useful in assessing the pollution levels of each heavy metal in the surface soil to account for their background value and to produce a more robust contamination degree analysis [25]. This index considers small variations in the background value using a 1.5 factor (factor K).

The GI was calculated according to Müller [23] and using Equation (2):

$$GI = \mathbf{Log2}\frac{\mathbf{Cn}}{1.5 \times \boldsymbol{Cb}} \quad (2)$$

where Cn represents the concentration of the element in soils collected at WF and U sites, and Cb represents the concentration of the same element (Table 1) in soils collected at F sites. According to Müller [23], the GI was divided into seven classes: unpolluted level for $GI \leq 0$; unpolluted to moderately polluted level for $0 < GI \leq 1$; moderately polluted level for $1 < GI \leq 2$; moderately to heavily polluted level for $2 < GI \leq 3$; heavily polluted level for $3 < GI \leq 4$; heavily to extremely polluted level for $4 < GI \leq 5$; and extremely polluted level for $GI > 5$.

2.4.3. Potential Ecological Risk Index

The potential ecological risk index (E), used to assess the ecological risk of soil subjected to anthropogenic contamination, was calculated as suggested by Hakanson [22], according to Equation (3):

$$\boldsymbol{E = Tf \times CF} \quad (3)$$

where CF is the contamination factor, and Tf is the toxic response factor of heavy metals and has value of 2 for Cr; a value of 5 for Cu, Ni and Pb; and a value of 1 for Zn [23]. The E was distinguished into five classes: low ecological risk for $E < 40$; moderate ecological risk for $40 \leq E < 80$; considerable ecological risk for $80 \leq E < 160$; high ecological risk for $160 \leq E < 320$; and very high ecological risk for $E \geq 320$.

*2.5. Human Exposure and Health Risk Assessment*

2.5.1. Ingestion and Dermal Contact Indices

The human health risks (non-carcinogenic and carcinogenic risks) arising from exposure to metals in soil sampled at each area (F, WF and U) were evaluated by taking into account two main exposure pathways: daily intake from ingestion (ADIi) of soil and dermal contact (ADId) of metals through contact of exposed skin with soil, both in adults and in children [26].

The ADIi and ADId of each element were calculated according to USEPA [27–29] and using Equations (4) and (5) as follows:

$$\boldsymbol{ADIi} = \left(\boldsymbol{Cn \times SIR \times EF \times \frac{ED}{BW} \times AT}\right) \times \mathbf{10^{-6}} \quad (4)$$

$$\boldsymbol{ADId} = \left(\boldsymbol{Cn \times SA \times AF \times ABS \times EF \times \frac{ED}{BW} \times AT}\right) \times \mathbf{10^{-6}} \quad (5)$$

where Cn represents the concentration of the element in soils; SIR is the soil ingestion rate estimated at 100 and 200 mg $day^{-1}$ for adults and children, respectively [30]; EF is the exposure frequency considered as 350 days $a^{-1}$ [30]; ED is the exposure duration estimated at 24 and 6 a for adults and children, respectively [31]; SA is the exposed skin surface area considered as 0.153 and 0.086 m2 for adults and children, respectively [22,25]; AF is the adherence factor estimated in 0.49 and 0.65 mg $cm^{-2}$ $day^{-1}$ for adults and children, respectively [32]; ABS is the dermal absorption factor calculated as 0.04 for Cr, 0.006 for Pb, 0.1 for Cu, 0.02 for Zn and 0.35 for Ni [33]; BW is the average body weight estimated at 56.8

and 15.9 kg for adults and children, respectively [31]; and AT is the average time of 26,280 and 2190 days for carcinogenic and non-carcinogenic risk, respectively [31].

Non-carcinogenic risk assessment of a single element in metal-contaminated soils is usually characterized using the hazard quotient (HQ). The HQ of element n received from the ingestion (HQin) and dermal contact (HQdn) exposure routes were calculated using Equations (6) and (7), respectively [28]. In order to assess the overall non-carcinogenic health risk posed by heavy metals through all means of exposure, the hazard index (HI) was recommended by the United States Environmental Protection Agency, and its calculation formula is Equation (8) [5,28]. If the HI is more than 1, there will be obvious potential risk for exposed population health and vice versa.

$$\boldsymbol{HQin} = \left(\frac{\boldsymbol{ADIin}}{\boldsymbol{RfDin}}\right) \tag{6}$$

$$\boldsymbol{HQdn} = \left(\frac{\boldsymbol{ADIdn}}{\boldsymbol{RfDdn}}\right) \tag{7}$$

$$\boldsymbol{HQI} = \boldsymbol{\Sigma HQin} + \boldsymbol{\Sigma HQdn} \tag{8}$$

where $RfD_{in}$ is the reference dose [34,35] of the element n obtained from ingestion (0.003 mg $kg^{-1}$ $day^{-1}$ for Cr; 0.0035 mg $kg^{-1}$ $day^{-1}$ for Pb; 0.04 mg $kg^{-1}$ $day^{-1}$ for Cu; 0.3 mg $kg^{-1}$ $day^{-1}$ for Zn; and 0.02 mg $kg^{-1}$ $day^{-1}$ for Ni), and $RfD_{dn}$ is the reference dose [22,25] of the element n obtained from dermal contact ($6.00 \times 10^{-5}$ mg $kg^{-1}$ $day^{-1}$ for Cr; $5.25 \times 10^{-4}$ mg $kg^{-1}$ day$^{-}$1 for Pb; $1.20 \times 10^{-2}$ mg $kg^{-1}$ $day^{-1}$ for Cu; $6.00 \times 10^{-2}$ mg $kg^{-1}$ $day^{-1}$ for Zn; and $5.40 \times 10^{-3}$ mg $kg^{-1}$ $day^{-1}$ for Ni).

#### 2.5.2. Carcinogenic and Non-Carcinogenic (ACR) Risk Assessment

Carcinogenic and non-carcinogenic risk (ACR) refer to the probability of an individual to develop any type of cancer in the course of his or her entire lifetime due to exposure to carcinogenic hazards [22,28]. According to Chen et al. [25], the ACR is calculated by summing the individual cancer risks across ingestion and dermal contact exposure ways and using Equation (9):

$$\boldsymbol{ACR} = \boldsymbol{ADIin} \times \boldsymbol{SFin} + \boldsymbol{ADIdn} \times \boldsymbol{SFdn} \tag{9}$$

where SFin and SFdn represent the cancer slope factor of element n via ingestion and dermal contact of soil particles ($mg^{-1}$ $kg^{-1}$ $day^{-1}$), respectively. However, cancer slope factors (CSFs) for other heavy metals were available for neither the ingestion nor the dermal contact means of exposure. Therefore, only the ACR of Pb, Ni and Cr was estimated according to SFin (0.0085 for Pb, 0.5 for Cr and 0.4 for Ni) [36], as in the study in which the ADIin was null for all the elements assessed and land use considered.

Generally, ACR values surpassing $1 \times 10^{-4}$ are viewed as unacceptable, whereas ACR values below $1 \times 10^{-6}$ are not considered to pose significant health effects, and ACR values lying in the range from $10^{-6}$ to $10^{-4}$ are generally regarded to be at a tolerable level [37,38].

### *2.6. Statistical Analyses*

The Shapiro–Wilk test was performed to test the normality of the data distribution.

The ANOVA test was performed to compare the significance of the differences among the investigated abiotic properties of soils collected in areas at different sites (forest, F; wildfire forest, WF; urban, U).

Principal component analysis (PCA) was carried out on the dataset of the soil metal concentrations to evaluate the site distributions, and the confidence ellipses (for $\alpha = 0.05$) for the land uses were superimposed to PCA (addEllipses function). In addition, permutational multivariate analysis of variance was used to evaluate the significance of the distance matrices (ADONIS, Montreal, QC, Canada). Redundancy analysis (RDA) was performed to

assess the impact of soil contamination indices (CF, E and GI), considered as independent variables, on human health risk indices calculated for adults (HQI_A and ACR), considered as dependent variables, calculated for FW and U soils. Due to the high correlation between human health risk indices calculated for children and adults, only the latter were considered for RDA. Forward selection (Ordistep function) of all the investigated independent variables was performed to select those that significantly affected the dependent variables.

All the statistical analyses and visualizations were performed using the R 4.3.0 programming environment with the ade4ˆ, vegan and Factoextra packages.

## 3. Results

### *3.1. Soil Abiotic Properties*

The mean values of the abiotic properties of soils collected in three different areas (forest, F; wildfire forest, WF; urban, U) are reported in Supplementary Table S1. The soils showed slightly alkaline pH, and WC ranged from 4.98 to 18.5% d.w., with values significantly higher in F and U than WF (Supplementary Table S1). Soil C concentrations ranged from 5.78 and 7.11% d.w., with no statistically significant differences among F, WF and U (Supplementary Table S1); meanwhile, soil N concentrations ranged from 0.35 to 0.89% d.w. and were significantly higher in WF (Supplementary Table S1). C/N and OM contents ranged from 9.42 to 15.5 and from 8.58 to 10.1% d.w., respectively, and there were no statistically significant differences among F, WF and U (Supplementary Table S1).

The total concentrations of the investigated metals in soils and the descriptive statistics among the three different sites (F, WF and U) of the study area are given in Table 1. The trends in the metal concentrations were as follows:

- Fe > Al > Mg > Mn >V > Cu> Cr > Ni > Pb > Zn at F sites;
- Fe > Al > Mg > Mn >V > Cu> Ni > Cr > Pb > Zn at WF sites;
- Al > Mg > Fe >Mn >V >Zn > Pb > Cu > Cr > Ni, at U sites.

According to ANOVA, the concentrations of the metals statistically varied among soils collected at different sites. In detail, the concentration of Al, Pb and Zn was significantly higher in U than F and WF (Table 1); the concentrations of V, Mn and Ni were significantly higher in F and WF than in U (Table 1); and finally, the concentrations of Cr, Cu, Fe and Mg were significantly higher in F than in U and WF (Table 1).

### *3.2. PCA of Metals and Site Distributions*

The first two PCA axes accounted for 64.6% of the total variance (PC1 explained the 47.9% and PC2 explained the 16.7%). The first axis clearly separated the three different sites in the PC space, which was confirmed by the PERMANOVA test ($p < 0.05$) that showed statistically differences between F, WF and U soils. In detail, the F and FW sites are distributed positively and negatively, respectively, along PC1, while U is mainly distributed positively along PC2 (Figure 2).

The total concentrations of Al, Cr, Fe and Ni were positively correlated to PC1, whereas the total concentrations of Mn and V were negatively correlated to PC1 (Figure 2). The total concentrations of Cu, Mg, Pb and Zn were positively correlated to PC2 (Figure 2).

### *3.3. Soil Metal Contamination Assessment*

The CF, GI and E indices for soil metal contamination and ecological risk calculated for the WF and U sites are reported in Table 2. The metal concentrations of F soils are used as background values. Based on CF classification, soils at FW did not show contamination for heavy metals, whereas soils at U sites showed considerable contamination for Zn, as well as moderate contamination for Pb and Al (Table 2).

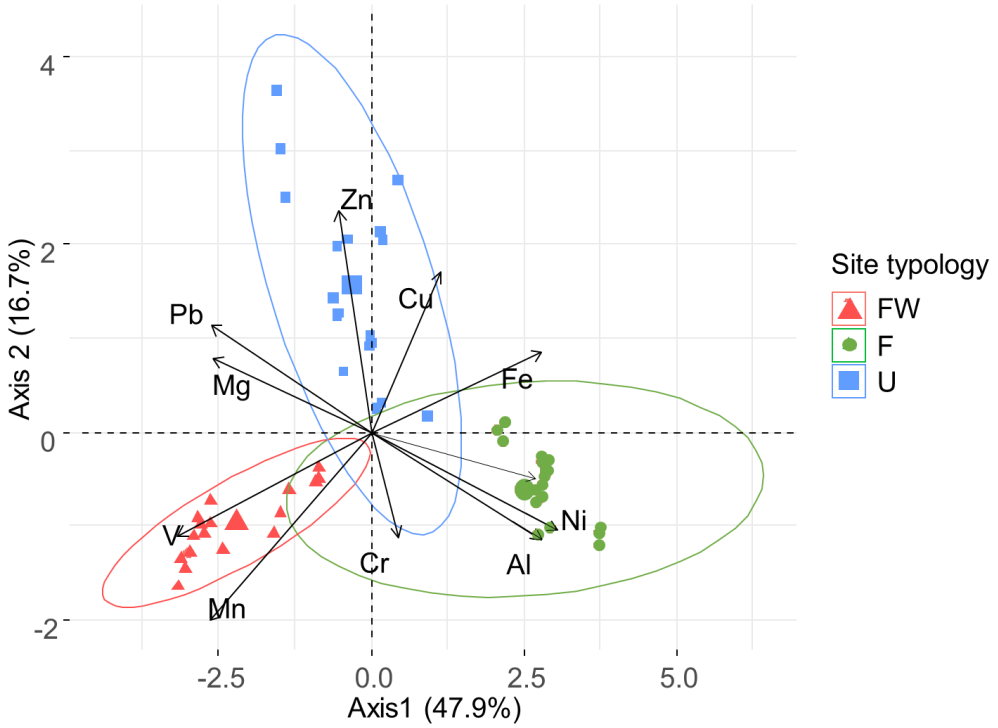


**Figure 2.** Principal component analysis (PCA) plots of metal concentrations (Al, Cr, Cu, Fe, Mg, Mn, Ni, Pb, V and Zn) and their distribution in space for the soils collected at forest (F), burned forest (FW) and urban (U) areas. Circle lines in PCA plots are superimposed to show the sampling sites in the same area. F: soil sampled at forest; WF: soil sampled at burned forest; and U: soil sampled at urban.

The trends in the metal contamination factor (CF) were as follows:

- V > Mn > Cu > Ni > Zn > Cr > Pb > Fe > Al > Mg at WF sites;
- Zn > Pb > Al > Cu > Cr > Mg > V>Mn > Ni > Fe at U sites.

Based on the GI classification, soils at WF did not show contamination with the metals assessed, whereas soils at U were contaminated for Pb and Zn (Table 2). The trends in the geo-accumulation index (GI) were as follows:

- Zn > Pb at U sites.
- The potential ecological risk (E) trends were as follows:
- Cu > Ni > Pb > Cr> Zn> at FW sites;
- Pb > Cu > Zn > Cr> Ni at U sites.

**Table 2.** Contamination factor (CF), geo-accumulation index (GI) and ecological risk (E) values for soils collected at burned forest (FW) and urban (U) areas for all metals assessed (Al, Cr, Cu, Fe, Mg, Mn, Ni, Pb, V and Zn). The highest values for each index are reported in bold.

| | CF | | GI | | E | |
|---|---|---|---|---|---|---|
| | **FW** | **U** | **FW** | **U** | **FW** | **U** |
| **Al** | 0.62 | 1.42 | −1.25 | −0.1 | 0.00 | 0.00 |
| **Cr** | 0.75 | 0.72 | −1.0 | −1.1 | 1.51 | 1.43 |
| **Cu** | 0.91 | 0.90 | −0.72 | −0.73 | **4.55** | 4.51 |
| **Fe** | 0.71 | 0.03 | −1.10 | −5.85 | 0.00 | 0.00 |
| **Mg** | 0.55 | 0.72 | −1.43 | −1.05 | 0.00 | 0.00 |
| **Mn** | 0.92 | 0.46 | −0.70 | −1.70 | 0.00 | 0.00 |
| **Ni** | 0.86 | 0.40 | −0.80 | −1.90 | 4.32 | 2.10 |
| **Pb** | 0.73 | 1.73 | −1.0 | 0.20 | 3.70 | **8.64** |
| **V** | **0.96** | 0.56 | **−0.64** | −1.41 | 0.00 | 0.00 |
| **Zn** | 0.79 | **3.45** | −0.92 | **1.20** | 0.79 | 3.45 |

### *3.4. Human Health Risk Assessment*

The average daily intake (ADIi) calculated for Cu, Cr, Ni, Pb and Zn is reported in Table 3. The ADIi calculated for both adults (A) and children (C) decreases as follows: Pb > Ni > Cu > Cr > Zn at FW areas; meanwhile, at U areas, it decreases as follows: Pb> Cu > Ni> Cr > Zn (Table 3).

**Table 3.** Daily intake from ingestion (ADIi) for adults (A) and children (C), hazard quotient index (HQI) and aggregate carcinogenic risk (ACR) values for soils collected at burned forest (FW) and urban (U) areas for Cr, Cu, Ni, Pb and Zn. The highest values for each index are reported in bold.

| | **ADIi HQI** | | | | **ACR** | | | |
|---|---|---|---|---|---|---|---|---|
| | **FW** | | **U** | | **FW** | | **U** | |
| | **A** | **C** | **A** | **C** | **A** | **C** | **A** | **C** |
| **Cr** | 0.02 | 0.04 | 0.03 | 0.04 | 0.01 | 0.02 | 0.01 | **0.02** |
| **Cu** | **0.04** | 0.06 | 0.04 | 0.06 | - | - | - | - |
| **Ni** | 0.02 | 0.04 | 0.01 | 0.02 | 0.01 | **0.03** | **0.04** | 0.01 |
| **Pb** | 0.02 | **0.08** | 0.05 | **0.08** | 0.0002 | 0.0007 | 0.0004 | 0.0007 |
| **Zn** | 0.01 | 0.02 | **0.06** | **0.1** | - | - | - | - |

The average daily dermal contact (ADId) was null for adults and children at both FW and U.

The hazard quotient index (HQI) values calculated for Cu, Cr, Ni, Pb and Zn are reported in Table 3. The HQI values calculated decreases as follows:

- Cu> Cr > Ni> Pb> Zn for adults (A) and Pb> Cu> Cr > Ni> Zn for children (C) at FW;
- Zn > Pb> Cu> Cr > Ni for adults (A) and Pb> Cu> Cr > Ni> Zn for children (C) at U.

The aggregate carcinogenic risk (ACR) for Cr, Ni and Pb showed the highest values for both children and adults for Ni at FW and at U, and for Cr at U (Table 3).

### *3.5. RDA of Soil Contamination and Human Health Risk Indices*

The first two axes of the redundancy analysis (RDA) explained 72% (Figure 3) of the total variance (PC1 explained 69% and PC2 explained 3%). The ordination of the plots based on the soil contamination indices and ecological risk index (CF, GI and E) is related to the human health risk indices (HQI and ACR) for each metal (Cu, Cr, Ni and Pb) considered (Figure 3).

The soil CFs for Cu, Cr, Ni and Pb and Es and GIs for Cr, Ni and Pb showed significant positive correlations with human health risk indices among FW soils (Figure 3), particularly regarding the HQI values for Cu, Cr and Ni and ACRs for Ni and Pb. Meanwhile, the CF, E and GI indices for Zn and Cu exhibited a significant positive correlation with HQI for Zn among U soils (Figure 3).

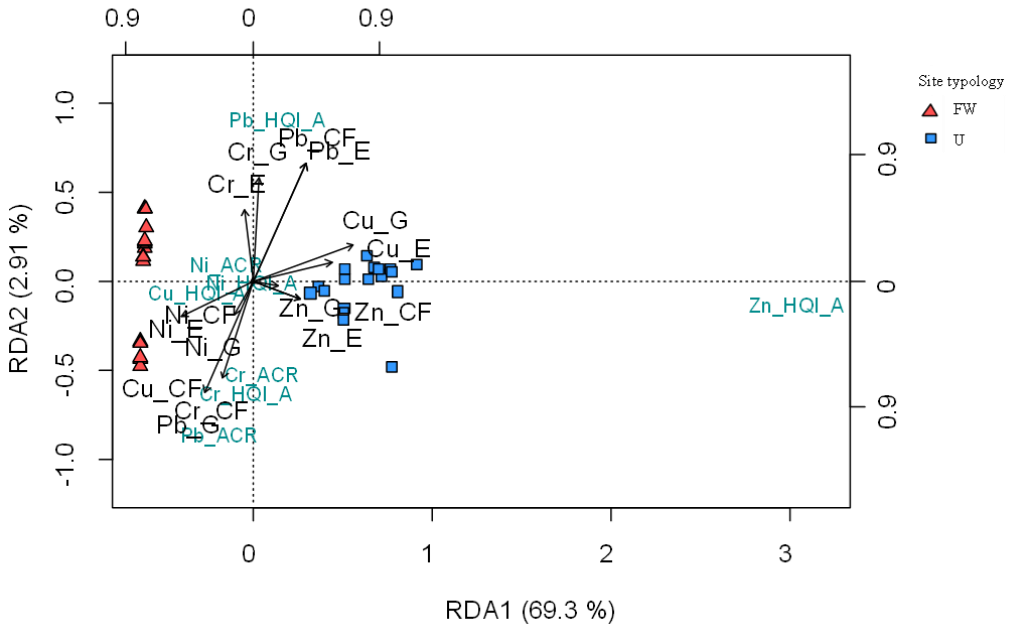


**Figure 3.** Redundancy analysis (RDA) of the soil contamination indices (CF, E and GI) and the human health risk indices (HQI_A and ACR) for each metal (Cu, Cr, Ni and Pb) in burned forest (WF) and urban (U) areas.

## 4. Discussion

Soil metal concentrations, together with the PCA results, highlighted different metal sources and pollution degrees. In fact, in F soils, Al, Fe, Mn, Ni and V seem to derive from parent materials through lithogenic and pedogenic processes, according to previous research performed in the investigated area [39]. Conversely, FW soils appeared moderately contaminated by Mg and V and highly contaminated by Mn. However, according to the GI, natural sources (substrate weathering) of Mg and V cannot be excluded in both FW and U soils. This supposition agrees with the findings reported by Memoli et al. [17], who found pedogenetic derivation of these metals in volcanic soils. The high input of Mn, which adds to the natural source, in FW soils could be due to the intense and brief emission sources from the numerous fires that occurred during the summer of 2016 in the investigated area [17]. The moderate degree of Pb and Zn pollution found in U soils highlighted by the CF and E index values suggests the derivation of this metal by human activities, more so than by substrate weathering or natural sources [40,41]. In fact, among metals related to road traffic, it is well known that Pb is a marker of vehicular emissions.

The moderate level of Pb pollution in U soils could derive from air particulate deposition coming from the nearby urban areas. This is in agreement with many studies that addressed the concentration of metals in soils and showed it to be higher near roads where vehicular traffic and emissions are higher [42–44].

According to previous findings by Santorufo et al. [45], the main sources of Cu, Mn and Zn also seem to be linked to human activities, such as agriculture, urbanization and industrialization, which highly impact the investigated areas, especially the U sites. In fact, the long-term use of excessive amounts of fertilizers, animal manures, pesticides and fungicides resulted in huge Cu, Mn and Zn accumulation in soils [46,47]. Furthermore, soil Zn contamination could be also caused by the presence of commercial areas, truck parks, welding factories and fabrication workshops, frequently characterizing peri-urban areas [48].

The findings show that the main route of human exposure to metals appears to be ingestion rather than dermal contact, as the average daily dermal contact (ADId) was null for both adults and children. Moreover, children appear to be exposed to a higher risk than adults as the average daily intake for ingestion (ADIi) was on average 2- to 4-fold higher for

children than for adults for the investigated metals (Cu, Cr, Ni, Pb and Zn). These findings agree with those reported in a recent study of Chonokhuu et al. [1] who found that risks for children were from 2- to 9-fold higher than that for adults in a population living in a zone characterized by values of soil metal accumulation similar to those measured in the area of Southern Italy.

However, among the investigated metals, only Pb appeared particularly dangerous, especially for children living in U areas. This is corroborated by the hazardous quotient index (HQI) for Pb, which was higher for children than adults. These results are relevant not only because children are highly exposed to soils during playtime [49,50], but also because they are particularly vulnerable to the adverse impacts of Pb [51]. In fact, Pb resuspension from soils has been shown to be an important contributor to the burden of blood Pb in children [52,53], which can also cause neurological and behavioral damages [54].

The cancer risk due to soil metals, as the aggregate carcinogenic risk (ACR) suggests, exceeded the safe level [27] for Cr, Ni, and Pb concentrations in both FW and U areas. If ACR values exceed the threshold set at $1 \times 10^{-4}$, it represents a lifetime carcinogenic risk to the human body. In fact, metals may accumulate in the fatty tissues of human bodies, presenting middle- and long-term health risks, adversely affecting physiological functions, disrupting the normal functioning of internal organs or acting as cofactors in other diseases [55].

The findings highlight strong relationships between soil contamination indices, the potential ecological risk index (CF, GI and E) and human health risk indices (HQI and ACR) in both areas (FW and U), suggesting positive relationships between each investigated metals and the area-specific risk for human health following the following decrescent order: Ni > Cr > Pb. Those areas (FW and U) are both typical of Mediterranean regions and urban areas, which are characterized by different human-induced pressures/impacts due to expanding growing cities and high industrial and agricultural activities, as well as natural impacts, such as frequent fires, especially during the drought season. This has been one of the most serious consequences of soil contamination and degradation due to metals. Also, previous studies carried out at the same area reported high concentrations of Cr and Pb due to fire and intense human activities [2,17,40]. Nevertheless, cancer risk cannot be excluded due to the geogenic Pb and Ni concentrations, as positive correlations were found between GI and ACR for these metals. In the urban area (U), the contamination of soil by Zn appeared to be the main potential risk for human health. By contrast, at forests sites (F), located further away from highly densely populated areas, showed high Al, Fe, Mn, Ni and V soil concentrations that appeared to be related to lithogenic and pedogenic processes [15] and scarcely related to any risk for human health. However, as FW and U sites surround F sites, they could receive metals for both brief and long-term periods. In fact, fires could become more frequent, especially during the drought period, and human activities could become more intensive as urban areas are expanding. As such, they cannot be excluded in F sites in the future as a risk for soil contamination and for human health.

## 5. Conclusions

As numerous intersections between soil metal contamination and human health occurred, the present research gives an important contribution to the current gap of knowledge about the extent of soil metal accumulation in human-impacted environments, as well as its potential risk for human health.

Firstly, metal contamination of soils of different site typologies varied according to the extent of both natural and human emission sources. In fact, V and Mn were higher in forest and wildfire forest soils, Mg in wildfire forest soils, and Al, Fe, Ni, Pb and Zn in urban soils. The contamination indices showed that Pb and Zn exceeded the background level in both wildfire forest and urban soils, and Mn did so in wildfire forest soils.

According to the human risk assessment, inhalation appeared to be the main route of exposure to metals in soils, and children were more exposed than adults to soil metal contamination. The overall cancer risk was higher than the safe level for Cr, Ni and Pb.

The findings highlight strong relationships between soil contamination indices (CF, E and GI) and human health risk indices, namely the hazard quotient index and aggregate carcinogenic risk (HQI and ACR, respectively), for each investigated metal.

Finally, management practices in forests also aiming to monitor soil pollution can benefit human health and can be helpful in developing further measures to preserve these ecosystems over time. This could be particularly useful in vulnerable ecosystems such as Mediterranean ones, where multiplying factors/pressures act to worsen the overall environmental value that, in turn, can reduce soil quality and increase human health risk.

**Supplementary Materials:** The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/environments10080146/s1, Table S1: Mean values (±st.dv.) of pH, water (WC), carbon (C), nitrogen (N) and organic matter contents (expressed as % d.w.) and C/N ratio of the soils collected at forest (F), burned forest (WF) and urban (U) areas.

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