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Accumulation patterns and health risk assessment of potentially toxic elements in the topsoil of two sloping vineyards (Tokaj-Hegyalja, Hungary)

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Abstract

Purpose In agricultural soils, accumulation and bioavailability patterns of potentially toxic elements (PTEs) are key ecological and health risk issues, especially in metal-based crop protection systems such as those applied in vineyards. However, PTE levels in the topsoils of historical wine-growing regions of Hungary have been scarcely studied so far. The specific goals of this research were to assess the accumulation tendencies and bioavailability of PTEs complemented with human health risk assessment in two sloping vineyards with pH-contrasted soils in Tokaj-Hegyalja, Hungary, and under different farming practices.

Methods Composite topsoil (< 20 cm) and borehole samples were collected from two vineyards and local forests. The total and bioavailable PTE contents (Zn, Pb, Co, Ni, Cr, Cu) were analyzed following digestion in aqua regia and extraction with a strong chelating agent (0.05 M Na_2-EDTA). Enrichment factors (EFs) were calculated based on Fe as a reference element and the local uncultivated soil. The hazard indexes (HIs) for outdoor workers, children, and adults living in residential areas near the vineyards were calculated to assess the health risks associated with the target PTEs.

Results Higher PTE contents were observed in the organic vineyard (near Tokaj) compared to the conventional one (near Tállya), except for Cu and Pb. The EFs confirmed that the duration of Cu-fungicide applications mainly determines the soil-bound Cu levels, with an average of 2.6 in the 28-year-old organic vineyard and 9.6 in the more than 100-year-old conventional vineyard. The PTEs predominantly accumulated at the top of the hillslope in Tállya, while in Tokaj, a general trend of downslope accumulation of PTEs can be noticed. Bioavailable Cu reached a maximum of 50% of total Cu at the top of the hillslope (Tállya) and positively correlated with soil organic matter content. Iron/Mn oxides, total Ca content, and soil pH show a significant correlation with the PTE total contents (other than Cu); meanwhile, their bioavailability is mainly influenced by Mn oxides. The calculated HIs are less than 1, indicating no elevated health risk. Total Cr is the major contributor to the HI in both vineyards, reaching 79.0% (Tokaj) and 49.7% (Tállya).

Conclusion Overall, the accumulated Cu contents mainly depended on the vineyard age, while farming practices and terrain morphology play a minor role in its spatial distribution. The further accumulation of PTEs, especially in high-metal hotspots within the vineyards, may ultimately cause toxicity to re-planted grapevines, soil biota, and, in the longer term, farmers and residents.

Keywords Accumulation tendency · Bioavailability · Enrichment factor · Health risk · Toxic metal · Vineyard soil

1 Introduction

Viticulture is an important agricultural practice, especially in Europe, that still encompasses the world's largest and oldest vineyard area (Fraga et al. 2012). Although most of the agricultural land in Europe can be considered adequately safe for food production (Tóth et al. 2016), one of mankind's

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greatest challenges is providing sustainable crop yields without damaging and polluting the environment. In vineyards, the soil degradation via contamination with potentially toxic elements (PTEs) and nutrient loss through water erosion can reduce the quality of soil and pose important environmental and toxicological concerns (Komárek et al. 2010; Preston et al. 2016. Therefore, with the long-term use of chemical fertilizers, fungicides, pesticides, and organic byproducts such as animal manure, viticulture can induce a considerable risk of soil contamination, particularly by PTEs.

Contamination of soil with PTEs is a major environmental issue at a global scale (Doumett et al. 2008; Rai et al. 2019). Potentially toxic elements, unlike organic chemicals (Pempkowiak 1991), can accumulate and contaminate the soil environment, affecting agricultural production in the long term. Their enrichment in vineyard topsoils (Farsang and Barta 2004; Farsang et al. 2012) is of particular concern due to the repeated use of metal-based pesticides and fertilizers, especially through the accumulation of Cu and Zn (Brunetto et al. 2014, 2016; Komárek et al. 2010; Patinha et al. 2018). The contamination by PTEs of neighboring ecosystems and residential areas is often inevitable due to leaching and wash-off (Brunetto et al. 2014; Mirlean et al. 2007; Patinha et al. 2018). Indeed, vineyards are primarily cultivated on steep slopes, and high Cu fluxes can be transported downslope and eventually off-site by surface runoff, in which metals are predominantly bound to soil particles (El Azzi et al. 2013; Babcsányi et al. 2016). Examining the spatial distribution of the soil-bound PTE contents can allow for the identification of contamination hotspots within sloping vineyards, potentially displaying hazardous concentration levels (Liu et al. 2005; Sipos 2004). Additionally, by calculating metal enrichment factors, the prevailing natural or anthropogenic character of PTEs can be unveiled (Bora et al. 2015; Fernández-Calviño et al. 2012; Liao et al. 2017; Loska et al. 2004; Preston et al. 2016; Szolnoki and Farsang 2013).

On the other hand, vineyards located close to residential areas can exert a considerable health risk when the soils display high PTE contents. The health risk quotient associated with soil contaminants by estimating the average daily intake (ADI, mg element/kg bodyweight/day) has been widely used in investigating the risk associated with PTEs (Farsang et al. 2009; Mirzaei et al. 2020; Rinklebe et al. 2019). Furthermore, evaluating the bioavailability of PTEs is a common method for assessing environmental risk and toxicity to crops (Borgese et al. 2013; Nunes et al. 2014; Violante et al. 2010). Several studies suggested that PTE availability depends mainly on the soil environment, such as the soil pH, soil texture, and soil organic matter (SOM) content (Alibrahim and Williams 2016; Takáč et al. 2009; Violante et al. 2010). Meanwhile, farming practices (conventional and organic agriculture), the age of the vineyard, and terrain morphology can also be considered dominant factors in determining soil quality and the PTE contamination status of the soil. Therefore, further investigation of the spatial distribution, accumulation patterns, and bioavailability of PTEs in vineyard soils and the associated human health risk is necessary to supply information on the sustainability of current agricultural practices. Based on the initial analysis of some PTE concentrations (e.g., Cu, Zn, Pb, Cd, Co, Ni, and Cr) and previous research, the selected PTEs of our study were Zn, Pb, Co, Ni, Cr, and Cu. In some previous works (e.g., Farsang and Barta 2004; Farsang et al. 2009; Liang et al. 2015; Milićević et al. 2018, 2020; Mirlean et al. 2007; Mirzaei et al. 2020; Romíc et al. 2004; Szolnoki and Farsang 2013; Szolnoki et al. 2013), these PTEs were reported as potential soil pollutants with health risk concerns.

Tokaj-Hegyalja is one of the historical wine-producing regions in northeastern Hungary, part of the UNESCO World Cultural Heritage List (Nyizsalovszki and Fórián 2007). The vineyards are situated at the hillslopes of the Tokaj Mountains, a member of the inner Carpathian volcanic range. Lately, innovative processes, changes in cultivation techniques, and socio-economic transformations have significantly transformed the landscape that also affected the soil environment in Tokaj-Hegyalja (Nyizsalovszki and Fórián 2007; Novák et al. 2014). In addition to conventional farming, organic and ecological farming practices gain ground in the vineyards of the region. These practices can exert different effects on the soil environment via impacting the physico-chemical soil properties and soil erosion dynamics and can also influence PTE accumulation patterns. Contrasting soils characterize the foothills of the Tokaj Mountains. Mostly acid soils have developed on the magmatic rock basement of the hills, but slightly alkaline soils are also found on the loess envelope of the magmatic rocks. Our previous studies investigated the enrichment and spatial distribution of PTEs in the topsoil of the vineyards in Tokaj-Hegyalja (near Tállya and Tokaj) using point samples (Manaljav et al. 2021; Pham et al. 2021). Meanwhile, this study focused on the accumulation patterns, and bioavailability of PTEs complemented with human health risk assessment by applying a hillslope position–based composite sampling strategy. In addition, we assessed the impact of contrasting soil characteristics and different viticultural practices on the soil environment. The specific goals of this research were as follows: (1) assessing the accumulation patterns and bioavailability of target PTEs (Zn, Pb, Co, Ni, Cr, and Cu) in two sloping vineyards with pH-contrasted soils and under different farming practices; (2) using enrichment factors to determine whether natural or anthropogenic input sources of the target PTEs are dominant in the vineyard soils; and (3) assessing human health risk of PTEs for workers, children, and adult residents.

2 Materials and methods

2.1 Study area

The vineyards, a 0.4-ha plot near Tállya and a 1.8-ha plot in the outskirts of Tokaj, are both located in the Tokaj-Hegyalja wine region (northeastern Hungary) (Fig. 1). The vineyards are situated at the hillslopes of the Tokaj Mountains. The interplay between the volcanic base rocks, the loess cover of variable thickness of the Tokaj Hill, and the surrounding alluvial plains, as well as the landscape topography create the diversity of soils in the region (Lóczy, 2015; Nyizsalovszki and Fórián 2007; Zelenka et al. 2012; Szepesi et al. 2018). Although the vineyards of Tállya and Tokaj belong both to the Tokaj-Hegyalja region, the soil-forming processes, soil characteristics, and cultural practices of the two sites considerably differ.

In Tállya, vineyard soils developed on rhyolit and rhyolite tuff, fine-grained, extrusive igneous rocks. The andesite of the subvolcanic body in Tállya was intruded into loose pumiceous rhyolite tuff in two phases based on the observations of two kinds of pyroxene andesite (Zelenka et al. 2012). The vineyard's slope shape displays a slight curving at the upper part and sharply increasing steepness at the middle (Fig. 1). The mean slope of the plot is 18°, and a marked inflection point is situated at the middle of the 146-m-long slope. The soil type slightly varies along the transect. At the top of the hillslope, it belongs to the Skeletic Regosol (Loamic, Ochric) type, while at the backslope it shows Skeletic Leptosol (Loamic, Ochric) patterns, and at the footslope the soil is Skeletic Colluvic Regosol (Loamic, Ochric) according to the World Reference Base for Soil Resources 2014 (FAO 2015). Conventional grape growing was set up a long time ago (supposedly more than 100 years ago) in Tállya. The Bordeaux mixture was applied in 2019 (the year of study) in a dose of 2.5–3 kg/ha/year (in May, June, and July), representing a yearly Cu dose of 0.5–0.6 kg/ha. The Bordeaux mixture was complemented with a foliar micronutrient fertilizer application in a dose of 4–5 l/ha, containing Fe (3.2 m/*V*%), Mn (0.32 m/*V*%), Cu (0.15 m/*V*%), B (0.31 m/*V*%), and Mo (0.003 m/*V*%). A synthetic insecticide (deltamethrin)

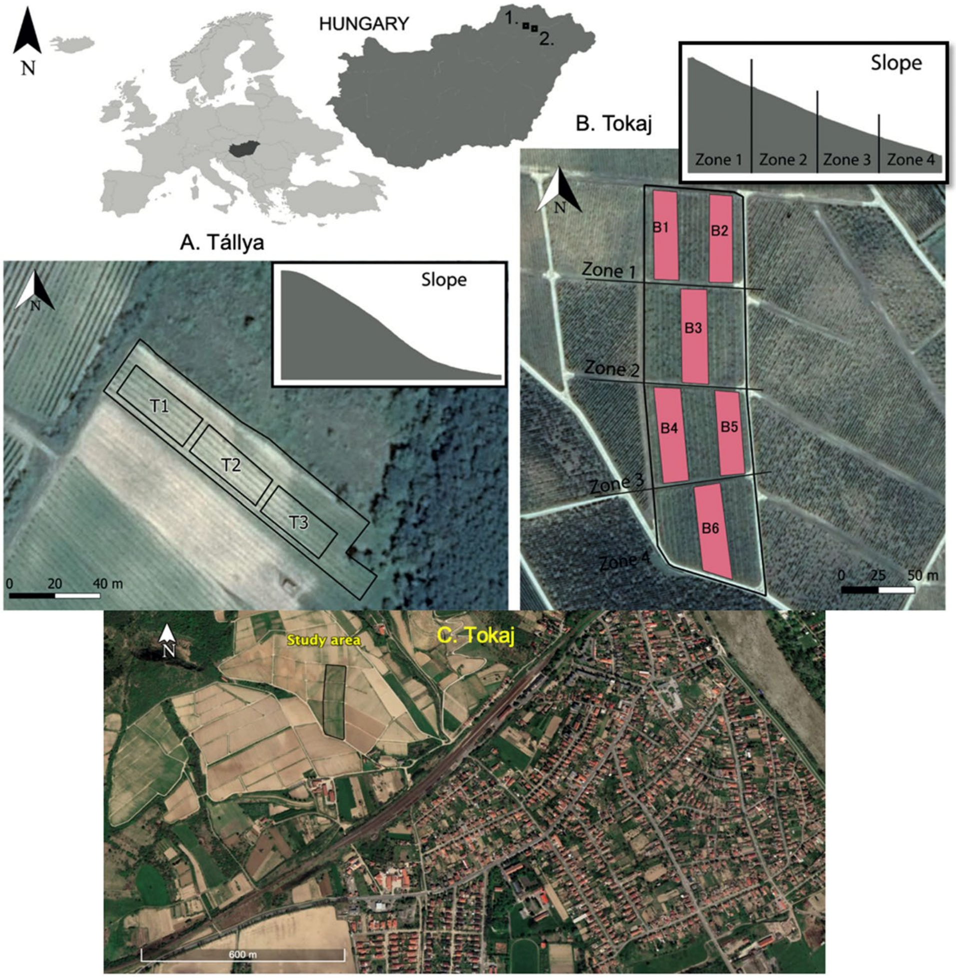

Fig. 1 Location of the two studied vineyards: (**A**) Tállya, (**B**) Tokaj, both situated in the winegrowing region of Tokaj-Hegyalja (NE Hungary), the slope profiles of each site are also displayed. Sampling zones for composite sampling are marked with rectangles and are named T1-T3 for Tállya and B1-B6 for Tokaj; (**C**) Tokaj: the location of the study plot in Tokaj with the nearby residential areas

was also sprayed in the vineyard. The soil is regularly ploughed, and no cover crops are sown in the vine inter-rows for soil protection.

Our second study site is situated at the southeastern foothill of the Tokaj Nagy Hill, the easternmost part of Hungary's inner Carpathian volcanic range (Lóczy 2015). The late Miocene pyroxene dacite lava flows and subordinate pyroxene dacite tuffs, mainly represented by the Amadévár Andesite and Tarcal Dacite Formations (Novák et al. 2014), constitute the primary parent materials with geochemical characteristics typical of subduction-related magmas (Harangi and Lenkey 2007). The Tokaj Nagy Hill is surrounded by alluvial plains formed from the deposits of the Bodrog, Tisza, and Takta rivers. Its andesite cone is mantled by loess deposits with a downslope increasing thickness, except for the summit zone (Kerényi 1994; Lóczy 2015; Novák et al. 2014). The vineyards surrounding Tokaj are situated mainly on these loess deposits. The soil in the vineyard is a Calcaric Regosol (Siltic, Ochric) according to the World Reference Base for Soil Resources 2014 (FAO 2015). The mean slope of the plot is 8°, and the investigated slope section is of 270 m in length. The study area is part of an organic vineyard. Before its replantation in 1993, no viticulture was practiced at the site from the 1950s. The local farmers used in the past fresh cattle manure to fertilize vineyards at a dose of 0.3t/ha every 3–4 years. However, for the last 10 years, cattle manure pellets have replaced fresh manure. To protect grape plants, Cu-based fungicides are repeatedly used in a typical dose of 4 kg/ha/year (in terms of Cu metal), supplemented with sulfur-containing pesticides and foliar fertilizers containing macro- and micronutrients. The vine inter-rows are covered by a mixture of grass and plants from the *Fabaceae* family that are regularly resown (every 4 years approximately). Every second year, the inter-rows are tilled (0–20 cm) to reduce soil compaction and increase their infiltration capacity, while tillage is more frequently practiced (two times a year) underneath the vine rows. In addition, there is a nearby open-pit quarry (Binét quarry) in the research area situated at ~ 500 m from the experimental site. The dacite stone quarry was abandoned in the 1980s.

2.2 Soil sampling and sample marking

Soil samples (including 18 composite topsoil samples (< 20 cm: 0–10 cm and 10–20 cm) and eight borehole samples) were collected from the studied vineyards in March 2019. Composite sampling was performed by mixing topsoil samples from the 0–10-cm and 10–20-cm layers taken from five points based on a two-way diagonal method conducted in each sampling zone (Fig. 1(A) and (B)). Hence, one 0–10-cm layer composite sample and one 10–20-cm layer composite sample were taken in each sampling zone. Soil samples were taken from the middle of grape inter-rows. Subsoil samples were collected from the center of each sampling zone (Fig. 1(A) and (B)) with the help of a hand auger. The samples were stored in clean polyethylene bags for transport to the laboratory, then disaggregated by hand, air-dried at room temperature for 7 days, and sieved at 2 mm.

The study plot in Tállya consists of nine grapevine rows. Composite samples (subsamples) were taken from three sampling zones (T1, T2, and T3) (of ~ 1500 m^2) (Fig. 1(A)). Two subsoil samples were collected from the 120–130-cm depth and the 180–200-cm depth considered respectively as the reference soils for sampling zones T1–T2 and T3. Pathways separate the study plot in Tokaj into four zones perpendicular to the main slope (Fig. 1(B)), whose primary role is surface drainage and erosion control. Twelve composite soil samples were collected from the six sampling zones chosen by a zig-zag method. Each zone includes ten rows of vine plants and has an approximate area of 1200 m^2. Subsoil samples collected from boreholes at 180–200-cm depth are considered reference soils, suggesting that negligible quantities of agrochemical-derived PTEs reach those layers.

Composite topsoil samples were collected from local forests as reference soils (not being impacted by vine-growing) at both study areas from the 0–10-cm and 10–20-cm soil layers.

2.3 Soil analyses

The soil samples were air-dried at room temperature, disaggregated by hand in a mortar, and then sieved through a 2-mm sieve. Basic soil parameters such as pH (d.w), carbonate content, soil texture, total salt content, and soil organic content (SOM) were analyzed following Hungarian Standards (MSZ 21470–52 1983; MSZ-08–0206-2 1978; MSZ-08-0205 1978). The $pH_{d.w}$ was measured in deionized water with a soil/water ratio of 1:2.5 using a digital pH meter (Inolab pH 720) (MSZ-08–0206-2 1978) (± 0.05). The carbonate content (in percentage of dry matter weight, ± 8%) was determined with a calcimeter according to the Scheibler method using 10% HCl solution for the reaction (Bojar et al. 2020). The determination of the soil texture based on the plasticity index values according to Arany (Arany plasticity index) was performed in accordance with the Hungarian Standard (MSZ-08–0205, 1978) (Table 1). Briefly, this index is calculated from the amount (cm^3) of deionized water added to an air-dry soil sample (100 g) until reaching the upper limit of its plasticity (Pham et al. 2021; Szolnoki and Farsang 2013). The total salt content was analyzed by a conductivity meter type Orion 3-Star (Thermo Electron Corporation) in saturated soil samples (± 10). The SOM content was determined by a UV–VIS spectrophotometer (a type Spectronic Helios-γ, Thermo Fisher Scientific), following H_2SO_4 (95%)-aided oxidation of the organic matter with 0.33 M $K_2Cr_2O_7$ (overnight) (MSZ-21470–52 1983) (± 2%).

Table 1 Texture categories according to the Arany plasticity index, a measure of soil texture according to a standard method (MSZ-08–0205 1978)

Soil texture class	Arany plasticity index (ml d.w./100 g soil)
Coarse sand	< 25
Sand	25–30
Sandy loam	30–38
Loam	38–42
Clayey loam	42–50
Clay	50–60
Heavy clay	> 60

For determining the PTE and major element contents (Zn, Pb, Co, Ni, Cr, Cu, and Al, Fe, Mn, Mg, K, Ca), approximately 0.5 g of oven-dried (at 105 °C) soil was precisely weighed into a PFA vessel, and 7 ml aqua regia (HNO_3/HCl = 1:3) was added. Before sample digestion, soil samples were ground in an agate ball mill to pass through a 250-µm sieve. All sample containers (tubes, vessels, volumetric flasks, etc.) were acid-washed and clean acids were applied (Normatom® for trace metal analysis, VWR Chemicals) for sample digestion and multi-element standard dilution. Soil samples were digested in a microwave oven (Anton Paar Multiwave 3000), as described elsewhere (Manaljav et al. 2021). Element concentrations in digested samples were determined by an inductively coupled plasma optical emission spectrometer (ICP-OES) (Optima 7000 DV, PerkinElmer) (± 10% uncertainty), using yttrium as an internal standard. For quality control purposes, a commercially available certified reference material (ERM®-CC141, a loam soil) was digested following the aforementioned method. The recoveries were consistent with the certified values, with the recovery percentages of 92 ± 2% (Zn), 87 ± 1% (Pb), 88 ± 0.2% (Co), 117 ± 2% (Ni), 102 ± 2% (Mn), 116 ± 2% (Cr), and 104 ± 3% (Cu). The procedural blank and laboratory reference materials for soils were regularly checked. To estimate the percentage of Mn and Fe in the form of oxyhydroxides, the reducible soil fraction was determined in the studied soils. The reducible fraction was extracted according to the BCR method with a 0.5 mol/l hydroxylamine hydrochloride solution (acidified to pH 1.5 with HNO_3) (Rauret et al. 1999). The extraction was performed by shaking on an end-over-end shaker (Stuart SP3 Rotator), at a speed of 30 rpm and a room temperature of 22 ± 5 °C (for 16 h). Finally, the Mn and Fe contents in the reducible fraction were analyzed by ICP-OES as described above and the oxide-bound Mn and Fe contents were compared to their respective aqua regia extracted pseudototal contents.

Single extraction procedures are recommended for studying the mobile and bioavailable proportions of elements in soils (Milićević et al. 2018, 2020). The chelating agent Na_2EDTA has proven to be effective in isolating the elements with which it usually builds very stable complexes (Milićević et al. 2018). The 0.05 M Na_2-EDTA was presented in studies as an agent that simulates the uptake of available elements in various soils (Alibrahim and Williams 2016). In the present study, the bioavailability of target PTEs (Zn, Pb, Co, Ni, Cr, Cu) was also determined by that agent (0.05 M Na_2-EDTA partially neutralized with NH_4^+) as described by Carter and Gregorich 2007. For that, 1.0 g of soil was weighed in a 50 ml centrifuge tube and 25 ml of purified 0.05 M Na_2-EDTA was added. All centrifuge tubes and labware were acid-washed and rinsed with purified 0.05 M Na_2-EDTA followed by a complete ultrapure water rinse. Samples were shaken on an end-over-end shaker (Stuart SP3 Rotator) (at 15 rpm for 1 h), centrifuged (at 2500 g for 20 min), filtered (at 0.45 µm), and then kept at 4 °C until ICP analysis. Prior to determining bioavailable PTE concentrations by ICP-OES, all samples were diluted 20 times with ultrapure water. Calibration standards were prepared in the same matrix as the diluted extract solution.

2.4 Enrichment factor

The enrichment factors (EFs) have been widely used to assess the enrichment of metals in soils and sediments, decipher their prevailing sources, and prove the anthropogenic interferences with natural element cycles (Cheng et al. 2018; Farsang et al. 2009; Preston et al. 2016; Reimann and Caritat 2005; Szolnoki et al. 2013). In this research, EF was applied to explore PTE accumulation and determine whether the natural or anthropogenic sources are dominant in the studied vineyard topsoils. The EF was calculated based on a reference element assumed to have negligible human sources. The most common reference elements used in similar studies are Fe, Al, Ti, Li, Sc, and Zr (Chatterjee et al. 2007; Duplay et al. 2014; Liu et al. 2005; Loska et al. 2004; Preston et al. 2016; Reimann and Caritat 2005; Rezaee et al. 2010; Szolnoki et al. 2013). Iron was appointed the reference element in our research, with the subsoil horizon as the reference soil. Enrichment factor was determined as follows:

$$EF = \frac{[E]_{SH}/[Fe]_{SH}}{[E]_{RH}/[Fe]_{RH}}$$

where $[E]$ is the concentration of PTE in the topsoil (SH) (0–10 cm) and the reference horizon (RH) (the subsoil).

Potentially toxic elements are not considered enriched in the topsoil when EF values are around 1 or slightly below 1. In contrast, EFs greater than 1 indicate that PTEs are enriched in the soil surface either from geogenic or anthropogenic sources. Enrichment factor values higher than 2 usually imply non-negligible anthropogenic input of PTEs into the soil (Facchinelli et al. 2001; Szolnoki et al. 2013).

2.5 Human health risk assessment

Our research evaluated the health risk associated with the total PTE contents primarily for outdoor workers in the two studied vineyards. In Tokaj, residential areas are located nearby the vineyards (Fig. 1(C)). Therefore, a resident scenario was also included for human health risk assessment considering children and adults. Accordingly, the hazard quotient (HQ) was determined as the ratio of the average daily intake (ADI) from ingestion of soil and a specific reference dose (RfD) of each PTE. Reference doses of PTEs were taken from the literature (Chen et al. 2015; Kamunda et al. 2016; Mirzaei et al. 2020; Rinklebe et al. 2019). The RfD is the potentially toxic element dose, defined as the maximum allowable level of an element that will not pose any harmful effects on human health. Average daily intake (mg element/kg bodyweight/day) was calculated as follows (USEPA 2001):

$$ADI = \frac{C_n \times SIR \times EF \times ED}{BW \times AT} \times 10^{-6}$$

where C_n is the concentration of a PTE in the 0–10-cm soil layer (mg/kg);

SIR is the soil ingestion rate: workers: 100; children: 200; adults: 100 mg/day;

EF is the exposure frequency: (workers: 250; children: 350; adults: 350 days/year);

ED is the exposure duration (workers: 25; children: 6; adults: 30 years);

BW is the bodyweight (workers: 70; children: 15; adults: 70 kg);

and AT is the averaging time (workers: 25 × 365 day/year = 9125 days, children: 2190 days; adults: 10,950 days).

Then, HQ (unitless) was calculated with the following formula:

$$HQ = ADI/RfD$$

where RfD is the oral reference dose of a PTE (unit is the same as for ADI): for Cr = 0.003, Cu = 0.04, Ni = 0.02, Pb = 0.0035, Zn = 0.3 (Chen et al. 2015; Mirzaei et al. 2020; Rinklebe et al. 2019), and Co = 0.02 (Kamunda et al. 2016).

The sum of all HQ values of each target PTE is the hazard index (HI):

$$HI = \Sigma HQ$$

When the values of HQ and HI are higher than 1, an apparent probability of the occurrence of adverse health effects is indicated (USEPA 2001), and HI < 1 shows that exposed persons are unlikely to experience dangerous health effects (Liang et al. 2015; Mirzaei et al. 2020).

2.6 Data analysis

Spearman rank-order correlation analysis determined the relationships between PTEs, relief, and soil properties. The significance level was considered at $p < 0.05$ and $p < 0.01$. In addition, one-way analysis of variance (ANOVA) was used to compare means of soil parameters between top- and subsoil layers at the level of $p < 0.05$. The statistical analyses were performed with the SPSS version 20.

3 Results and discussion

3.1 Vineyard soil properties

Table 2 summarizes data about the measured soil properties, such as soil pH (d.w), soil texture, salt content, carbonate, and soil organic matter (SOM) contents. According to the Arany plasticity index, topsoils in Tállya display a loam to sandy loam texture without any marked difference between the 0–10-cm and the 10–20-cm layers. The predominant soil texture in Tokaj is also sandy loam, only with a slightly lighter character compared to Tállya. Stone fragments are one of Tállya's soils' significant components and appear in a higher proportion at the middle section of the main slope. The top 0–10-cm soil layer contains an average of 48% of coarse fraction (> 2 mm), and the underlying 10–20-cm layer displays 53%. Vineyard topsoils in Tállya are slightly acid, with $pH_{d.w}$ ranging from 6.12 to 6.67, while, in Tokaj, the soil $pH_{d.w}$ is moderately alkaline (ranging from 7.96 to 8.10). In Tállya, the soil is poor in carbonates ($CaCO_3$). The generally higher carbonate contents characterizing the soil in Tokaj (varying from 1.65 to 6.26%) explain their moderately alkaline character and originate from the loess parent material. The highest SOM content in Tállya is detected at the upper part of the hillslope (19.5 g/kg), which then decreases to 10.4 g/kg at the backslope zone. The re-raising SOM at the footslope suggests that the organic matter is transported downslope from the backslope zone. Signs of intense erosion at the backslope, such as the high coarse fraction and low SOM, clearly explain the uneven distribution of soil components. Likewise, in Tokaj, the redeposition of SOM-rich material is observed from zone B2 (12.6 g/kg) to zone B4 (14.6 g/kg), while the highest SOM content is found at the top zone (zone B1) (16.7 g/kg). Generally, the downslope accumulation of organic matter can be indicated for both sites. There is no marked difference in the soil texture between topsoil samples along the hillslope in Tokaj. The main erosion-impacted part of the hill is situated at zone B2, where the slope increases significantly.

Table 2 Basic soil parameters: Arany plasticity index (API) (-), total salt content (%), $\text{pH}_{\text{d.w.}}$(-), $CaCO_3$ (%), soil organic matter (SOM) content (g/kg), and major element contents determined in the soil samples collected from the studied vineyards in Tállya and Tokaj

		Depth (cm)	API	Salt (%)	pH (d.w)	$CaCO_3$ (%)	SOM (g/kg)	Al*	Fe*	Mn***	Mg**	K**	Ca**
Tállya	T1.1	0–10	39	0.02	6.14	2.94	19.5	32	21	427	371	572	329
	T2.1	0–10	38	0.03	6.28	2.94	10.4	36	19	212	386	564	357
	T3.1	0–10	36	0.02	6.67	3.36	14.6	20	11	285	214	418	264
	T1.2	10–20	36	0.02	6.12	3.36	11.0	32	22	471	378	531	295
	T2.2	10–20	39	0.03	6.30	3.36	8.8	36	20	199	392	497	352
	T3.2	10–20	36	0.02	6.65	2.94	11.1	19	11	264	216	349	265
	Mean		37	0.02	6.36	3.15	12.6	29	17	310	326	489	310
	Median		37	0.02	6.29	3.15	11.1	32	20	275	375	514	312
	Min		36	0.02	6.12	2.94	8.8	19	11	199	214	349	264
	Max		39	0.03	6.67	3.36	19.5	36	22	471	392	572	357
	SD		2	0.01	0.24	0.23	3.9	8	5	113	86	88	42
	Tp1	120–130	41	0.04	6.28	3.36	8.3	22	10	470	259	397	226
	Tp2	180–200	27	0.01	6.58	2.10	6.6	6	8	862	121	105	168
Tokaj	B1.1	0–10	34	0.02	7.97	2.48	15.9	25	24	507	692	552	1549
	B2.1	0–10	37	0.02	8.01	4.95	15.5	22	24	503	678	491	1585
	B3.1	0–10	35	0.02	8.01	5.37	12.6	23	24	498	790	503	1906
	B4.1	0–10	34	0.01	8.10	1.65	14.4	21	24	510	793	460	1931
	B5.1	0–10	34	0.02	8.02	5.00	14.6	20	23	498	772	410	1908
	B6.1	0–10	35	0.02	8.00	5.42	14.2	22	25	538	745	438	1892
	B1.2	10–20	34	0.02	7.96	3.72	16.7	23	25	509	680	471	1446
	B2.2	10–20	35	0.02	8.02	5.37	13.7	21	24	499	682	441	1662
	B3.2	10–20	34	0.02	8.08	4.13	12.5	20	23	476	762	397	1855
	B4.2	10–20	35	0.02	8.07	2.48	10.0	21	24	502	760	421	190
	B5.2	10–20	34	0.02	8.05	6.26	10.5	19	24	497	754	405	1828
	B6.2	10–20	37	0.02	8.01	5.42	13.1	18	24	516	671	294	2097
	Mean		35	0.02	8.03	4.35	13.6	21	24	504	732	440	1654
	Median		35	0.02	8.02	4.98	14.0	21	24	503	750	440	1842
	Min		34	0.01	7.96	1.65	10.0	18	23	476	671	294	190
	Max		37	0.02	8.10	6.26	16.7	25	25	538	793	552	2097
	SD		1	0.00	0.04	1.46	2.0	2	1	14	47	65	498
	Bp1	180–200	41	0.01	8.64	5.86	5.0	19	23	500	902	410	2612
	Bp2	180–200	34	0.02	8.26	6.70	3.7	19	23	494	859	361	2349
	Bp3	180–200	34	0.01	8.58	7.53	4.7	18	22	476	864	374	2432
	Bp4	180–200	39	0.05	8.15	4.19	6.5	22	25	532	781	412	1443
	Bp5	180–200	35	0.07	7.85	3.77	13.2	29	27	585	666	654	1117
	Bp6	180–200	36	0.02	8.14	5.02	8.2	19	24	527	6615	313	1563

*Units are in g/kg

**Units are in mg/100 g

***Units are in mg/kg

3.2 Total and bioavailable contents of PTEs in the vineyard soils

The soil-bound PTE contents at the vineyards in Tállya and Tokaj are summarized in Fig. 2. In Tállya, the PTEs apart from Pb showed higher mean contents in the vineyard than the local forest soil. In Tokaj, the mean contents of Zn, Ni, Cr, and Cu in the 0–10-cm layer were adequately higher than in the local forest soil, while Pb and Co showed opposite trends. Although the observed Pb contents in both control sites were higher than in the vineyards, evaluating Pb contents is necessary due to health risk concerns (especially for children) (Rinklebe et al. 2019; Tirima et al. 2016). So far, Co has received little attention and it has rarely been assessed in vineyards, especially for evaluating

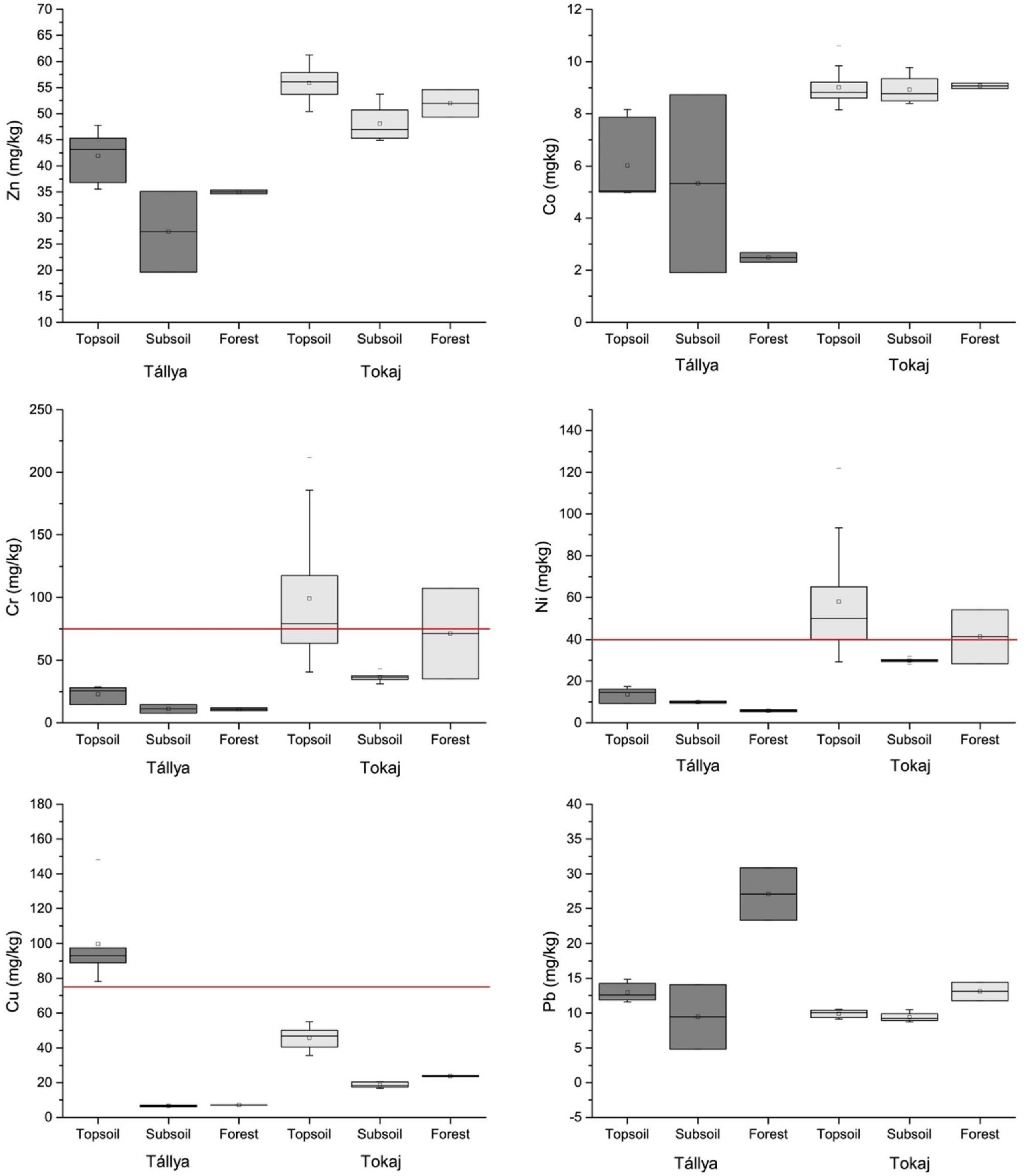

Fig. 2 Concentrations (mg/kg) of PTEs measured in the top- (0–10-cm and 10–20-cm) and subsoil samples in the vineyards at Tállya and Tokaj and the local forest soils. The pollution limit values (B) of the Hungarian standards (Joint Decree No. 6/2009 2009) for the PTEs are indicated by the horizontal red line (Zn: 200 mg/kg; Pb: 100 mg/kg; Co: 30 mg/kg; Ni: 40 mg/kg; Cr: 75 mg/kg; and Cu: 75.3 mg/kg)

Co-related environmental and health risks. Therefore, Co is also included in the present study.

The mean content of Cu in Tállya (111.4 mg/kg) exceeded the pollution limit value (B) (75 mg/kg) for soils and sediments figuring in Hungarian standards (Joint Decree No. 6/2009 2009). Due to the long-term and repeated use of Cu-based fungicides, Cu contamination can be considered a major environmental concern in vineyards. Old vineyards (such as the vineyard in Tállya) are particularly affected. Even though the background content of Cu in Tállya was lower than that in Tokaj, twice as high concentrations were observed in the vineyard topsoil (Cu-fungicides have been used for more than 100 years in Europe). The shorter period of Cu-fungicide applications in Tokaj can be a plausible reason for the lower Cu contents as most of the agrochemical derived Cu took place during the past 28 years, starting from the re-planting of the vineyard in 1993. Copper contamination in vineyard soils is a common challenge in conventional and organic farming as vine growers hardly have any efficient alternative to Cu substances for protecting vine plants against the infection by *Plasmopara viticola* (Brunetto et al. 2016). High anthropogenic Cu contents in the soil may raise ecotoxicological concerns; therefore, determining the easily soluble proportions of the total Cu is a prerequisite for assessing the environmental impact of Cu-based fungicide use (Komárek et al. 2010).

Although the observed Zn contents did not exceed the Hungarian standards (Joint Decree No. 6/2009 2009), assessing its accumulation tendencies is important as Zn is an essential micronutrient for plants. Zinc is often used in fertilizers, in particular in those, spread directly on the plants during the growing season (30 g/ha/year used in Tokaj). In both vineyards, the higher Zn contents in the vineyard topsoil compared to the subsoil and the local forest soils imply a moderate enrichment of Zn from fertilizer applications. While in Tállya, the vineyard topsoil overall contains low levels of Ni and Cr, the markedly higher Ni and Cr levels observed in Tokaj exceed the pollution limit values (B) (Joint Decree No. 6/2009 2009). These similarly high contents of Ni and Cr in both the forest and vineyard topsoils suggest that additional local sources (such as the dacite base rock and the abandoned quarry), other than pesticides and manure treatments, should be considered. In addition, a strong correlation was found for Ni and Cr ($R = 0.99$), indicating their common origins and behavior in the investigated soils at Tokaj. Indeed, the soil samples from the local forest have been taken near the edges of the abandoned dacite stone quarry, supporting the idea of the bedrock's influence and the elevated local geochemical background for Ni and Cr. In addition, the repeated applications of fresh manure, manure compost, and, more recently, manure pellets can also exert impact on Ni and Cr enrichment in the vineyard topsoil. Indeed, it has been previously suggested that such organic amendments can add significant amounts of PTEs, such as Zn, Cr, Ni, and Cu (Barakat et al. 2016; Gong et al. 2019; Wang et al. 2018; Wuana and Okieimen 2011). High concentrations of Cr and Ni in vineyard topsoils may eventually pose a potential risk of toxicity, especially when replanting young vine plants (Romić et al. 2004; Lago-Vila et al. 2015). Hence, their bioavailable proportions should be evaluated.

On the other hand, besides the vineyard's age and farming practices, the relief and soil properties such as soil texture, $pH_{d.w}$, SOM content, and other soil constituents may impact total and bioavailable PTE contents in the vineyard soils. The significant correlation between SOM and total Cu ($R = 0.88$) in Tállya pointed out the role of organic matter in Cu binding. The high molecular weight and insoluble fractions of organic matter can retain significant Cu in the soil (Brunetto et al. 2016). Meanwhile, Zn, Ni, and Cr showed a significant positive relationship with total Al and Fe, and the Arany plasticity index (API) (Table 3), revealing their association with fine soil fractions and the inorganic colloids, such as clay minerals and Fe/Mn oxyhydroxides, contained in those fine-grained fractions (Fernández-Calviño et al. 2012; Scheniost 2005). The aqua regia extractant is ideal to release elements trapped in Mn and Fe oxides and oxyhydroxides. This solution is also suitable to release PTEs from their strong bonds, probably by total or partial dissolution of Mn-oxides (due to its better ability to dissolve Mn-oxides than Fe/Al oxides) (Pavlíčková et al. 2003). Meanwhile, aqua regia is unable to dissolve silicates and Al oxides (Niskavaara et al. 1997; Lymperopoulou et al. 2017). Also in Tállya, the negative correlation between the soil $pH_{d.w.}$ and Zn, Ni, and Cr contents, may indicate a higher environmental risk prevailing at lower pH.

In Tokaj, the relationship between Zn and Pb contents and total Mn also indicates their association with oxides of Mn (bounded and/or occluded). Our extraction results of Mn recovered in the reducible fraction accounting for more than 50% of the total Mn in both studied vineyards, indicate that Mn oxides are the predominant form of the soil-bound Mn (data not shown). Iron/Mn oxides in the soil may exhibit a high surface area, with reactive sites which strongly bind and tightly sorb metals such as Cu, Pb, Zn, Ni, Co, and Cd (Scheniost 2005). Conversely, a negative correlation was observed between total Al and Co, Ni, and Cr contents suggesting that low Al- and high Ca-containing (due to a positive relationship with Ca) soil constituents (such as pyroxene, amphibole, and apatite) may carry the soil-bound Ni and Cr in Tokaj (Table 3). Indeed, in Tokaj, the Ca content is anticorrelated with Al contents. In addition, negative relationships of Co, Ni, and Cr with the slope steepness imply that terrain morphology plays an essential role in their spatial distribution, showing the possible impacts

Table 3 Spearman's rank correlation matrix between soil properties, slope, and total PTE data in vineyard soils in Tállya and Tokaj

	Al	Fe	Mn	Ca	SOM	API	$CaCO_3$	Mean slope (degree)	$pH_{d.w}$
Tállya									
Zn	0.88*	0.91*	0.24	0.84*	0.18	0.93**	−0.17	−0.23	−0.90*
Pb	0.10	0.45	0.91*	−0.08	0.59	0.10	−0.31	−0.93**	−0.59
Co	0.28	0.64	0.95**	0.03	0.48	0.19	0.05	−0.93**	−0.74
Ni	0.84*	0.98**	0.51	0.64	0.04	0.67	0.14	−0.44	−0.98**
Cr	0.89*	0.99**	0.43	0.73	0.04	0.74	0.06	−0.36	−0.99**
Cu	0.05	0.22	0.50	0.11	0.88*	0.37	−0.63	−0.59	−0.34
Al	1.00	0.92*	0.00	0.94**	−0.20	0.84*	0.00	0.08	−0.85*
Tokaj									
Zn	0.28	0.43	0.76**	−0.10	0.47	−0.03	−0.05	−0.18	0.47
Pb	0.19	0.70*	0.61*	−0.37	0.39	0.38	0.32	0.14	0.39
Co	−0.60*	0.42	0.35	0.58*	−0.37	0.27	0.05	−0.69*	−0.37
Ni	−0.69*	0.26	0.19	0.62*	−0.44	0.35	0.10	−0.63*	−0.44
Cr	−0.67*	0.25	0.19	0.64*	−0.49	0.27	0.04	−0.68*	−0.49
Cu	−0.20	0.05	0.42	0.05	0.26	0.19	0.36	−0.18	0.26
Al	1.00	0.21	0.19	−0.75**	0.57	−0.19	−0.36	0.66*	0.57

API Arany plasticity index, *SOM*, soil organic matter

*Significant at the level of $p < 0.05$

**Significant at the level of $p < 0.01$

of soil erosion and redeposition processes. Generally, Cu showed a less significant correlation with soil properties in the vineyard at Tokaj.

Based on the EDTA extracted PTE contents (marked with X_{BIO}), Zn, Ni, and Cr showed an overall low mobility in both vineyards (Fig. 3).

The contents of Ni_{BIO} and Cr_{BIO} are not considerably tailored by their total concentrations. Even though the total contents of Ni and Cr in Tokaj are markedly higher compared to Tállya (Fig. 3), there is no significant difference between their bioavailable proportions. Bioavailability ratios of Cr in both studied sites are less than 1%, indicating its almost immobile character and a strong binding to the soil (Table 4). Despite differences in the total Cu contents between the two vineyards, the bioavailability ratios in Tállya (48–49%) are only slightly higher than those in Tokaj (39–49%) (Fig. 3). Similarly to Cu, the high bioavailable proportions of Pb and Co in both vineyard soils and uncultivated soils indicate their lability in the soils (Table 4). Indeed, bioavailable proportions of Pb reached almost 50% of its total contents both in the vineyard and the forest topsoils, with slightly higher Pb_{BIO} in Tállya. This can be explained by the high retention capacity of Mn oxides for Pb and the high affinity of Pb for Fe oxides in the soil (Arenas-Lago et al. 2014), which can be extracted with the applied EDTA (Lo and Yang 1999). In the current study, Pb shows the greatest affinity for Mn oxides in both vineyards. The Zn_{BIO} contents are less than 10 mg/kg (Fig. 3). However, its slight increase in the uppermost soil layer may indicate the input of some soluble Zn via foliar fertilizers' use. There is a significant difference between the bioavailable proportions of Zn and Cu in the vineyard soil and the local forest soil. In both vineyards, the markedly higher percentages of bioavailable Cu in the topsoil compared to the local forest soils suggest that the anthropogenic sources can be a factor of variation for the bioavailability of Cu. As expected, with a strong accumulation of Cu in the vineyard topsoil, Cu can be sorbed in the soil solid phases through mechanisms such as ion exchange (non-specific adsorption), specific adsorption, and complexation with soil organic matter (Brunetto et al. 2016; Kabata-Pendias 2004). The weak and unstable non-specific adsorption directly affects the availability of Cu (Brunetto et al. 2016). Although there is no noticeable difference between the total content of Zn in the vineyard soil and the local forest soil, significantly high proportions of bioavailable Zn are presented in the vineyard topsoil in Tokaj. Meanwhile, higher bioavailable ratio of Zn in the local forest soil in Tállya with lower total contents of Zn suggests that Zn bioavailability probably depends on the local soil properties, while anthropogenic inputs play a minor role.

In Tállya, the SOM content strongly influenced the EDTA extracted contents of Cu showing a positive correlation (Table 5). The quantity of PTEs extracted by EDTA can be highly dependent on mineral phases and their respective total contents (Couto et al. 2014; Brunetto et al. 2014, 2018; Duplay et al. 2014; Fernández-Calviño et al. 2012). Significant correlation between total Mn contents and PTEs (except for Cu) may indicate their association with the reactive surface sites of Mn oxides (Table 5). Indeed, the extractant Na_2-EDTA has proven to be effective in removing

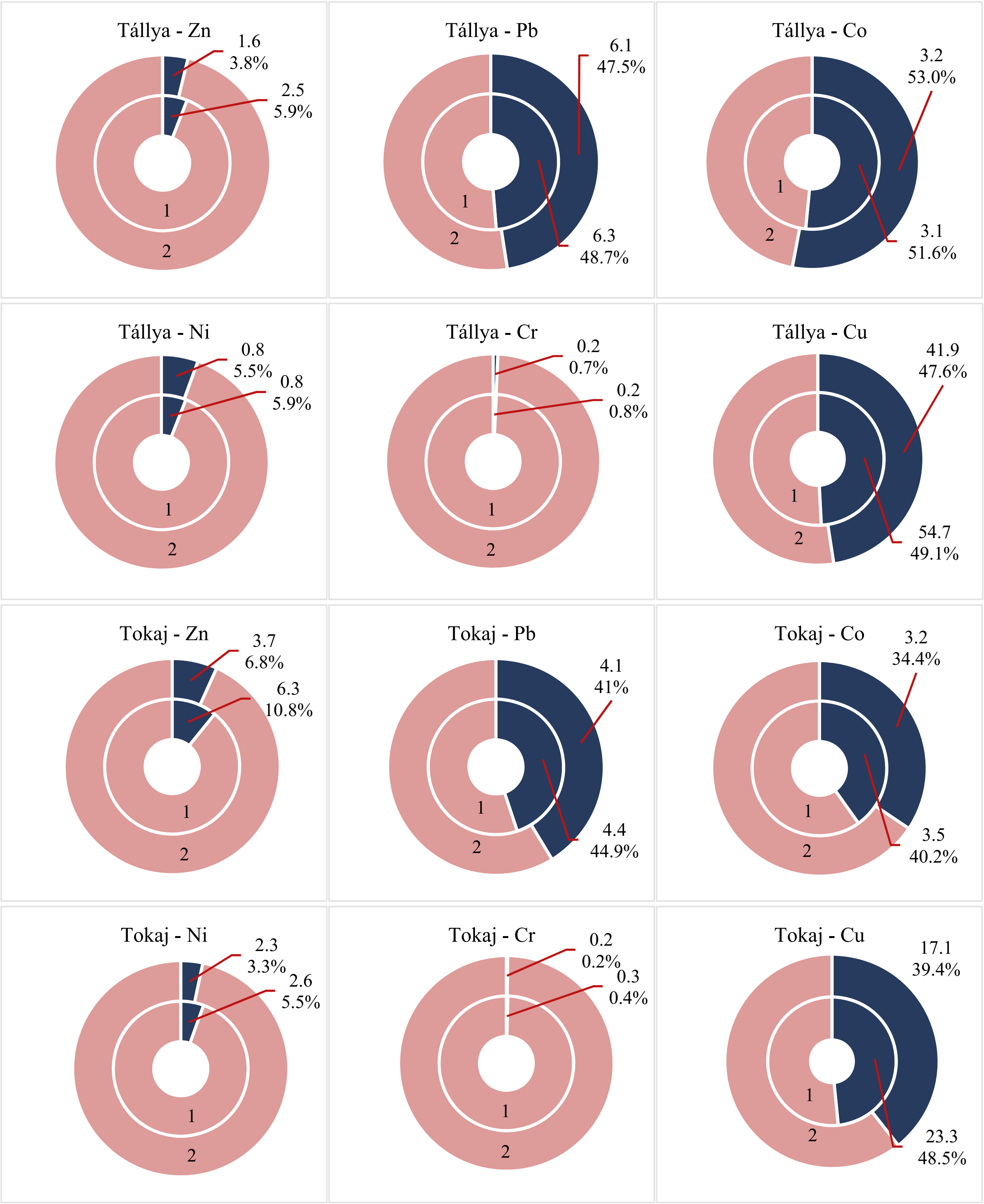

Fig. 3 Average concentrations of bioavailable PTEs (mg/kg) and bioavailable ratio (%) in vineyard soils (diagram 1: 0–10 cm; diagram 2: 10–20 cm) in Tállya and Tokaj

Table 4 Average of bioavailable ratio* (%) of target PTEs in the topsoils in Tállya and Tokaj

Sampling site	Depth (cm)	Zn	Pb	Co	Ni	Cr	Cu
Tállya ($n=3$)	0–10	5.9	48.7	51.6	5.9	0.8	49.1
	10 – 20	3.8	47.5	53.0	5.5	0.7	47.6
The local forest in Tállya ($n=1$)	0 – 10	11.5	50.7	33.8	0.0	2.7	13.7
	10 – 20	9.3	65.0	28.1	0.0	1.8	20.5
Tokaj ($n=6$)	0 – 10	10.8	44.9	40.2	5.5	0.4	48.5
	10 – 20	6.8	41.3	34.4	3.3	0.2	39.4
The local forest in Tokaj ($n=1$)	0 – 10	2.6	48.8	33.6	7.1	0.0	23.2
	10 – 20	0.0	51.6	34.7	4.3	0.2	20.1

*Average of bioavailable ratio (%) = average of (bioavailable PTE fraction/total PTE concentration in soil) × 100 (Alibrahim and Williams 2016; Kashem and Singh 2001)

metals bound to Fe/Mn oxide surfaces (Lo and Yang 1999). On the other hand, soil properties, such as the Arany plasticity index (texture), carbonate content, and soil $\mathrm{pH_{d.w}}$ seem to play a minor role in the bioavailability of the studied PTEs.

3.3 The impact of the topography on the enrichment factor of the PTEs in the vineyard soils

Calculating EF based on Fe as a reference element may also allow deciphering the predominant natural or anthropogenic character of PTEs and their accumulation trends in the sloping landscape.

In Tállya, the mean EFs for Zn and Ni were below 1, while EFs for Pb, Co, and Cr were slightly higher than unity (Table 6). The mean Cu EF was 9.7 (Table 6), indicating significant Cu enrichment in the surface soil layer. In Tokaj, the mean EFs for Zn, Pb, Co, and Ni were around 1, while Cr (2.2) and Cu (2.6) EFs (Table 6) indicate their moderate enrichment in the topsoil. The anthropogenic origin of Cu is predominant in the vineyards. The higher EFs of Cu observed in Tállya (Fig. 4) show the effect of its long-term use as Cu-fungicides. In contrast, the prevailing geogenic origin of Pb, Co, and Cr can be concluded in Tállya and no significant enrichment of Ni and Zn can be noticed. Likewise, Zn, Pb, and Co are mainly of geogenic origin in Tokaj, while a moderate enrichment of Ni and Cr cannot be excluded due to local sources, such as abandoned open pit quarry and inputs through organic amendments.

In Tállya, Pb, Co, and Cu tend to get enriched at the top of the hillslope at the summit zone, while in Tokaj a pronounced downslope enrichment can be highlighted (Fig. 4). Due to the complex slope shape of the vineyard in Tállya, EF (Cu) reaches the highest value at the top (the summit) and decreases to its lower value at the steepest backslope area (T2), before re-raising at the footslope

Table 5 Spearman's correlation matrix between soil properties, relief, and bioavailable PTE contents in the vineyard topsoil in Tállya and Tokaj

	$\mathrm{Fe_{BIO}}$	$\mathrm{Mn_{BIO}}$	SOM	API	$\mathrm{CaCO_3}$	Mean slope (degree)	$\mathrm{pH_{d.w}}$
Tállya							
$\mathrm{Zn_{BIO}}$	0.20	0.36	0.70	−0.20	−0.50	−0.16	0.52
$\mathrm{Pb_{BIO}}$	0.46	0.81	0.73	−0.27	−0.53	−0.68	0.12
$\mathrm{Co_{BIO}}$	0.53	0.99^{**}	0.64	−0.30	−0.02	-0.95^{**}	−0.18
$\mathrm{Ni_{BIO}}$	0.38	0.91^{*}	0.50	−0.21	−0.12	-0.96^{**}	−0.43
$\mathrm{Cr_{BIO}}$	0.30	0.94^{**}	0.59	−0.61	0.00	−0.77	0.22
$\mathrm{Cu_{BIO}}$	0.39	0.57	0.89^{*}	0.04	−0.62	−0.50	0.07
Tokaj							
$\mathrm{Zn_{BIO}}$	0.02	0.72^{**}	0.57	−0.02	−0.24	0.28	−0.23
$\mathrm{Pb_{BIO}}$	−0.04	0.70^{*}	0.51	0.54	−0.11	0.42	−0.57
$\mathrm{Co_{BIO}}$	0.38	0.95^{**}	0.46	0.31	−0.29	0.35	−0.33
$\mathrm{Ni_{BIO}}$	0.63^{*}	0.79^{**}	0.34	0.22	−0.12	−0.09	−0.33
$\mathrm{Cr_{BIO}}$	0.37	0.78^{**}	0.33	0.46	−0.10	−0.10	−0.20
$\mathrm{Cu_{BIO}}$	−0.11	0.51	0.53	0.22	0.12	0.25	−0.27

API Arany plasticity index, *SOM* soil organic matter

*Significant at the level of $p < 0.05$

**Significant at the level of $p < 0.01$

Table 6 The enrichment factor values calculated using Fe as a reference element for PTEs based on composite samples in Tállya and Tokaj

	Sampling zone—Tállya			Mean value	Sampling zone—Tokaj						Mean value
	T1	T2	T3		B1	B2	B3	B4	B5	B6	
Zn	0.7	0.6	1.3	0.9	1.2	1.2	1.1	1.2	1.2	1.2	1.2
Pb	1.5	1.3	0.6	1.1	1.1	1.1	1.0	1.0	1.1	1.0	1.1
Co	2.0	1.4	0.4	1.3	1.0	1.0	0.9	1.0	1.1	1.0	1.0
Ni	0.7	0.7	0.7	0.7	1.3	1.3	0.9	1.6	2.5	1.9	1.6
Cr	0.9	0.9	1.4	1.1	1.7	1.6	1.1	2.3	3.4	3.1	2.2
Cu	11.8	8.2	9.0	9.7	2.4	2.7	2.1	2.4	3.1	2.9	2.6

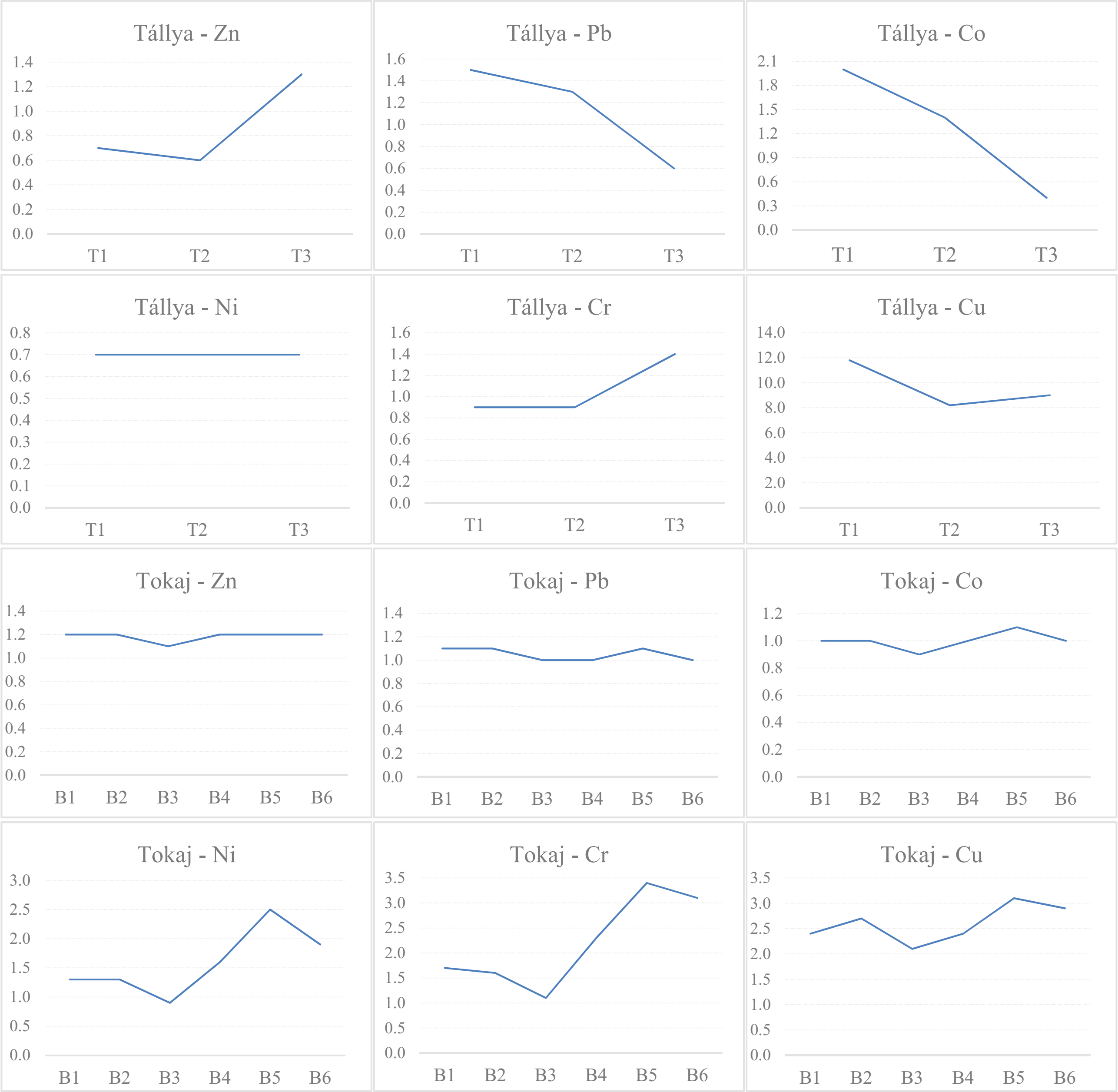

Fig. 4 The spatial variations in the enrichment factors of the examined PTEs calculated using Fe as a reference element for the different sampling zones along the hillslope

(Fig. 4). The higher EFs for Zn and Cr at the bottom of the slope also indicate their erosion-impacted spatial distribution pattern. Indeed, along with the complex slope shape of the vineyard in Tállya, the low slope gradients (0–5°) and the high contents of the coarse fraction probably protect the soil from excessive erosion-induced losses at the summit zone (Manaljav et al. 2021). In contrast, the EFs of Cu in Tokaj reach their highest values at the footslope (Fig. 4). In line with the negative correlations between PTEs and the mean slope gradients of the sampling zones, a general trend of downslope accumulation can be noticed, especially for Co, Ni, Cr, and Cu (Fig. 4). Hence, the terrain morphology plays a significant role in the accumulation and enrichment patterns of PTEs in the topsoil, showing the impact of erosion in their redistribution within the sloping vineyards.

3.4 Human health risk assessment

The calculated hazard quotient (HQ) accounting for the health risk associated with the "soil-to-human" exposure of the studied PTEs was either negligible or moderate. Only the mean HQ for total Cr of 0.34 (unitless) for children based on the topsoil Cr content of the vineyard in Tokaj shows a moderate risk. There was a remarkable difference between HQ values for children, workers, and adults. Generally, the HQs for workers, adults, and children were in the order of Cr > Pb > Ni > Cu > Co > Zn in Tokaj; and Cr > Pb > Cu > Ni > Co > Zn for workers in Tállya (Fig. 5). The HQ assessment results in Tállya are in good agreement with the findings of Mirzaei et al. (2020), who estimated similar HQ for Cr, Pb, Cu, and Zn in vineyard soils. In Tokaj, due to the high total concentration of Ni in the topsoil, the HQ (Ni) exceeds the health risks associated with Cu (Fig. 5). No significant non-carcinogenic risk was revealed in our study, similarly to anterior research on potential human health risks related to PTEs in vineyard soils by Mirzaei et al. (2020) and Liang et al. (2015) for adults and children, and Milićević et al. (2018) for workers.

Indeed, the calculated hazard index (HI) accounting for the overall health risk related to all studied PTEs for all groups considered (children, adults, and workers) and throughout the study areas was less than 1 (Fig. 6). Therefore, no elevated health risks were detected. However, in Tokaj, considering overall health risks, the mean HI for children (0.43) was more than nine times higher than for adults (0.046). Despite the substantially higher contents of Cu observed in Tállya, the mean HI for workers in Tállya

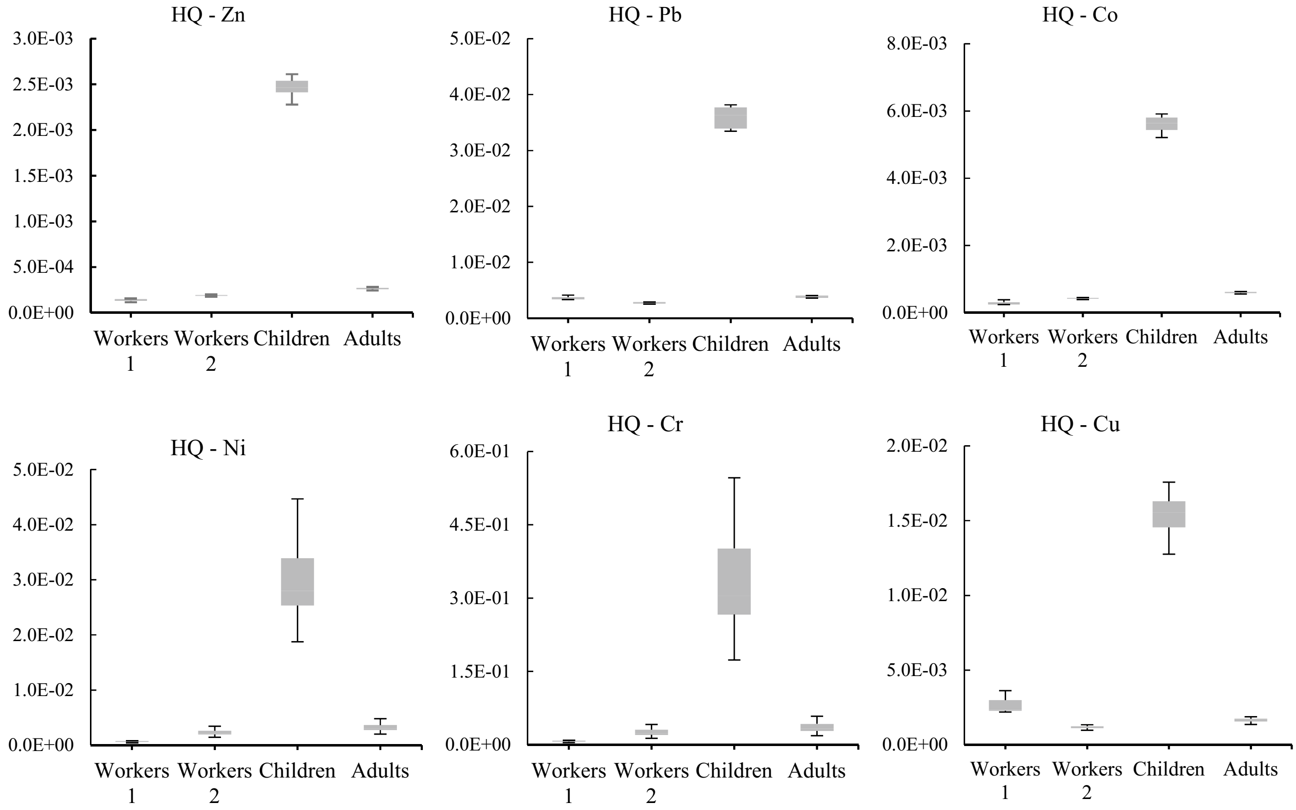

Fig. 5 Hazard quotient (HQ) based on the individual PTE contents in the studied vineyards for workers (1—in Tállya; 2—in Tokaj) in both vineyards; for children and adult residents only in Tokaj

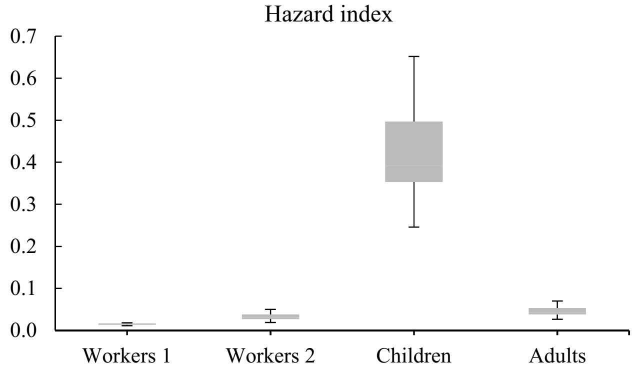

Fig. 6 Hazard index (HI) accounting for the health risk related to the total PTE contents in the two studied vineyards for workers (1—in Tállya; 2—in Tokaj); children and adult residents in Tokaj

(0.015) was two times lower than that in Tokaj (0.033) (Fig. 6), showing the minor role of Cu in causing health risk issues. Higher contents of PTEs can explain the latter observed in Tokaj compared to Tállya (apart from Pb and Cu). In a previous study, Mirzaei and co-workers (2020) reported a maximum HI of 0.062 in a vineyard topsoil (0–20 cm) for Cu, Cd, Cr, Pb, and Zn, a score inferior to the highest HI of 0.65 in Tokaj (Fig. 6). The difference is likely due to the significantly higher concentration of Cr in the Tokaj vineyard due to a probably higher geochemical background. Hence, the high Cr content observed in Tokaj originating from local sources and to a smaller extent manure applications considerably increased the overall human health risks of the soil-bound PTEs included in the study.

As expected from the individual HQs in Tokaj, the highest contribution percentage to the total HI is associated with Cr, reaching up to 79.0%, followed by Pb (8.4%), Ni (7.0%), and Cu (3.6%). The relative contribution of the rest merely accounted for the remaining 2.0% of the total risk (Fig. 7). Similarly to Tokaj, Cr displayed the highest contribution percentage (with 49.7%) in the vineyard topsoil of Tállya, followed by Pb (24.6%) and Cu (18.5%). The higher total Cu content due to the longer term use of Cu pesticides in Tállya resulted in a higher contribution of Cu to the total HI (18.5%), against 3.6% in Tokaj. Even though Cu is the main pollutant in the vineyards showing the highest enrichment levels, still, Cr and Pb are reported to represent the highest risks in similar studies discussing PTE-related health risks in vineyard soils (Mirzaei et al. 2020; Milićević et al. 2018).

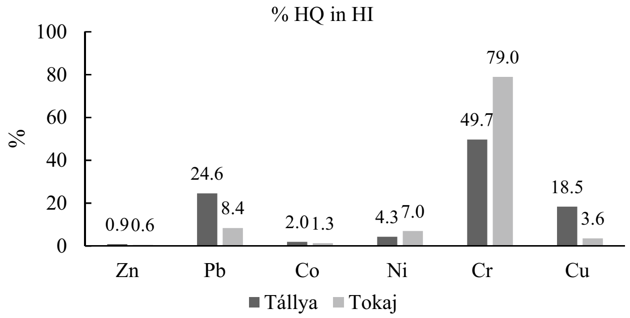

Fig. 7 Contribution percentage of the individual PTEs to the hazard index (HI), the latter representing the overall human health risk of the six PTEs in the vineyard soils. Note that percentages are equal among the various studied groups (children, adults, and workers alike)

4 Conclusions

Viticulture exerts a significant impact on the soil environment. It changes specific soil properties and results in the accumulation of potentially toxic elements (PTEs) through farming practices. The soil management also induces enhanced soil erosion in the vineyards, which in turn impacts PTE accumulation patterns. Copper was the major pollutant in the older conventional and the younger organic vineyards too. In the more than 100-year-old vineyard in Tállya, the Cu content exceeded the Hungarian environmental quality standards for soils and sediments, while in the more recently re-planted vineyard in Tokaj, Cu was observed below that limit with only a moderate enrichment of Cu in the topsoil. Apart from Ni and Cr, the considered PTEs accumulated principally in the top 10 cm in both vineyards. The higher Zn content in the vineyard topsoil compared to the subsoil and the local forest soils implies a moderate enrichment of Zn from regular foliar fertilizer applications. In accord with the total contents, the proportions of bioavailable Cu were highest among the target PTEs, reaching 50% at the top of the hillslope in Tállya. Consequently, Cu represents a plausible risk of toxicity to grapevines and soil biota. Despite the substantially higher contents of Ni and Cr observed in Tokaj, their bioavailable proportions were generally below 1% in the two study sites.

The studied PTE accumulation and bioavailability can present very different behavior in the uppermost soil layers depending on the different soil conditions. Data revealed that mineral phases, total Ca content, and soil pH (especially in Tállya) show a significant correlation with the total contents of the target PTEs; meanwhile, the bioavailability of PTEs was largely influenced by their binding to Mn oxides, except for Cu. Soil organic matter content positively correlates with the total and bioavailable Cu content showing that Cu has a high affinity for organic matter. Total Cu content mainly depends on the vineyard age. In addition, terrain morphology played an essential role in the enrichment patterns of PTEs in the topsoil (due to erosion-impacted spatial distribution patterns). In Tállya, an inversed pattern was observed with predominant enrichment of PTEs at the top of the hillslope. Conversely, the target PTEs showed higher contents at the footslope in Tokaj, confirming the downslope relocation of PTE-rich sediments due to soil erosion processes.

The PTE-related health risk assessment showed no elevated health risk (hazard index, HI < 1) for children, adult residents and workers. The overall health risk related to all target PTEs in Tokaj (0.43) was more than nine times higher for children than adults (0.046). The health risk related to Cr contents was the predominant risk factor in both vineyards, accounting for 49.7% (in Tállya) and 79.0% (in Tokaj) of the calculated HIs.

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Declarations

Conflict of interest The authors declare no competing interests.

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