



## Heavy metal contamination in recorded and unrecorded spirits. Should we worry?

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### ABSTRACT

Heavy metals can be released into all alcoholic beverages during production and storage. However, there is at least a theoretical risk that they could be present in higher, and potentially toxic, concentrations in those produced in the household and small-scale stills common in Central and Eastern Europe, which lack quality control and whose products are unrecorded by authorities. Yet, so far, few studies comparing concentrations of heavy metals in recorded and unrecorded spirits have been published. In this study we ask whether there is any difference between heavy metal concentrations in recorded and unrecorded spirits and, thus, the related health risk. The levels of heavy metals were determined in recorded ( $n = 97$ ) and unrecorded ( $n = 100$ ) spirits using inductively coupled plasma optical emission spectrometric analysis and applied to population-based risk assessments, considering average, regular and chronic heavy drinkers. Concentrations of Cu, Zn, and Sn were significantly higher in unrecorded spirits than those in their recorded counterparts and recorded spirits contained significantly higher levels of Fe, Mn, and Ni than unrecorded spirits. Combined exposure to heavy metals posed a potential health risk in chronic heavy drinkers consuming recorded spirits. However, when compared to the health risk arising from drinking large volumes of ethanol, the risk is negligible. Consequently, there are no grounds to worry about the adverse effects of heavy metals from spirits.

### 1. Introduction

Excessive alcohol intake has been identified as one of the leading risk factors contributing to disease burden, associated with more than 60 acute and chronic diseases (Griswold et al., 2018). Epidemiological studies have shown that the burden of alcohol-related disease depends on both the volume of alcohol consumed and the drinking pattern (Horvat et al., 2018; Rehm et al., 2017) but there is growing evidence that, while ethanol is by far the most important component of alcoholic beverages, other biologically active components should also be taken into account (Bujdosó et al., 2019).

Hundreds of such chemicals have been identified in beer, wine and spirits (IARC, 1988; Jellesen et al., 2006). Some chemicals such as acetic

acid, ethyl acetate, and tannins act as flavouring agents at the concentrations usually found (IARC, 1988) but there may also be many toxic and, in some cases, carcinogenic compounds, including methanol, acetaldehyde, ethyl carbamate, and heavy metals (Bujdosó et al., 2019; Rehm et al., 2014; Tariba, 2011; Weber and Sharypov, 2009). The last of these can be released into alcoholic drinks from raw materials and equipment used during brewing, distillation, aging, bottling, and storage (Ibanez et al., 2008; Jellesen et al., 2006). As a result, a variety of metalloids and metals have been detected in alcoholic beverages such as arsenic (As), cadmium (Cd), chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), tin (Sn), lead (Pb), and zinc (Zn) (Ibanez et al., 2008; Rehm et al., 2014; Tariba, 2011). Although some of these, such as Fe, Mn, and Zn, are essential elements, acting as cofactors

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for several enzymes (WHO, 1996). In contrast, other heavy metals consumed in alcoholic drinks can cause toxic effects even at low concentrations (IARC, 2012; Tatarková et al., 2019). For example, chronic exposure to Pb or Cd has been shown to induce anaemia, kidney damage and neurological disorders (Nordberg et al., 2007; Wani et al., 2015). Although less toxic heavy metals including Cu, Cr (III), Fe, Mn, Sn, Zn have been found in all types of alcoholic beverages, the concentration of Pb and Cd may be higher in unrecorded alcohols, also known as non-commercial alcohols, which comprise home-made, informally produced, and smuggled alcoholic products as well as industrial and medicinal alcohols that are not intended for human consumption (WHO, 2018) and which may be produced using sub-standard metal equipment (Lachenmeier et al., 2011a; Tatarková et al., 2019).

This is especially important in Central-Eastern European (CEE) countries, where these products are widely consumed (WHO, 2018), almost exclusively as home-made spirits (Popova et al., 2007), in conditions lacking any meaningful quality control (Bujdosó et al., 2019; Lachenmeier et al., 2011a; Tatarková et al., 2019). Consequently, consumers, and especially heavy drinkers (Lopez et al., 2002), may be exposed to higher levels of heavy metals than if they only drank recorded spirits. Given the scale of consumption, if heavy metals are present in these beverages there is cause for concern about health beyond any effects of ethanol (Lachenmeier et al., 2011a; Tatarková et al., 2019).

There is, however, only limited data on concentrations of heavy metals in unrecorded spirits, unlike their recorded counterparts (Ibanez et al., 2008; Lachenmeier et al., 2011a), so we do not know how they compare with the toxicological threshold values proposed by the Alcohol Measures for Public Health Research Alliance (AMPHORA) project (Lachenmeier et al., 2011a). We are aware of a few studies that examined the health risk arising from drinking recorded and unrecorded spirits (Iwegbue et al., 2014a, Iwegbue et al., 2014b; Lachenmeier et al., 2012, Lachenmeier et al., 2013; Otim et al., 2019). One used the margin of exposure approach to compare the health risk from consumption of recorded and unrecorded alcoholic beverages containing As, Cd and Pb (Lachenmeier et al., 2012). However, it did not include other heavy metals such as Cr, Co, Cu, Fe, Mn, Ni, Sn, and Zn. We are unaware of any published studies that compare health risks arising from drinking recorded and unrecorded spirits in CEE countries. Consequently, we seek to fill this gap by determining and comparing the concentration of heavy metals in recorded and unrecorded spirits. We then used these data to estimate the daily intake of heavy metals attributable to alcohol consumption in the Hungarian population, using a probabilistic Monte Carlo Simulation, looking separately at men and women and with different consumption levels. Finally, the target hazard quotient (THQ) method was used to estimate the health risk posed by consumption of recorded and unrecorded spirits containing heavy metals.

## 2. Materials and methods

### 2.1. Sample collection

The sampling methodology was described in our previous publication (Bujdosó et al., 2019). In summary, the samples were collected by the members of our research team using snowball sampling (Piauiense et al., 2020; Sfodera et al., 2020). Spirits manufactured industrially were categorised as recorded, while spirits produced in small scale distilleries or in private homes were considered to be unrecorded. Recorded spirit samples (n = 97), including Hungarian fruit spirits (pálinka, n = 25), whiskey (n = 21), vodka (n = 16), brandy (n = 18), rum (n = 6), artificially flavoured spirits (n = 5), gin (n = 3), tequila (n = 2), and absinth (n = 1) were purchased from Hungarian supermarkets. All commercial spirits bore tax stamps. Unrecorded spirits, lacking tax stamps (n = 100), were bought informally from 31 persons (on average 2–3 samples/person from different batches) in 19 settlements in Eastern Hungary who ferment fruits at home and either distil the mash in their own stills or send it to small local distilleries. The inclusion criteria were that the

**Table 1**

Limit of detection and limit of quantitation of heavy metals

Nr.	heavy metal	limit of detection [ $\mu\text{g}/\text{litre}$ ]	limit of quantitation [ $\mu\text{g}/\text{litre}$ ]
1	cadmium	0.1	0.5
2	chromium (III)	0.2	1.0
3	cobalt	0.3	1.5
4	copper	0.1	0.5
5	iron	0.2	1.0
6	lead	2.2	11.0
7	manganese	0.02	0.1
8	nickel	0.5	2.5
9	zinc	0.1	0.5
10	tin	1.0	5.0

unrecorded spirits should be distilled from fruits fermented at private homes and they were not taxed. Following collection, each spirit was labelled with an identification number to prevent mismatches, decanted into glass bottles and kept in the dark at 4 °C until inductively coupled plasma optical emission spectrometry analysis (ICP OES) could be performed.

### 2.2. Inductively coupled plasma optical emission spectrometric analysis

Analyses of spirit samples were carried out using an Agilent ICP OES system (5100 SVDV model, Agilent Technologies, Santa Clara, USA) as described previously (Baranyai et al., 2015; Simon et al., 2013). Briefly, spirit samples were diluted 10 fold with 0.1 M nitric acid (Merck, Darmstadt, Germany) prepared in ultrapure water prior to the analysis. Calibration series were diluted from a multielement standard solution containing Ag, Al, B, Ba, Bi, Ca, Cd, Co, Cr, Cu, Fe, Ga, In, K, Li, Mg, Mn, Na, Ni, Pb, Sr, Tl, Zn at a concentration of 1000 mg/L (ICP, IV, Merck, Darmstadt, Germany). A double pass spray chamber and concentric (Meinhard type) nebulizer was used to introduce samples. Argon gas was used to supply the plasma and nitrogen gas was applied for sample introduction and optical purge. To validate the method, blank samples were used to check the purity of water and glassware applied. To determine the influence of ethanol concentration on the detectability of heavy metals, solutions containing ethanol (Merck, Darmstadt, Germany) at concentrations of 0.0, 5.0, 10.0, 25.0, and 40.0% were spiked with the multielement standard solution. The final concentration of the elements was 0.1 mg/L. The accuracy of the measurements was more than 95%. The limit of detection and quantitation for each heavy metal analysed is shown in Table 1.

### 2.3. Compliance of heavy metal concentrations with threshold values

The AMPHORA project has established threshold values for contaminants of spirits, including heavy metals (Lachenmeier et al., 2011a). The heavy metal concentrations measured in our spirit samples were compared with the AMPHORA threshold values. The results of this comparison are shown in Tables 4 and 5.

### 2.4. Population-based comparative risk assessment

To estimate the health risk associated with the consumption of recorded and unrecorded spirits we used a probabilistic risk assessment approach with different scenarios, as described in detail in our previous paper (Bujdosó et al., 2019 - Supplement 1). First, the distribution of estimated daily intakes (EDI) of heavy metals were calculated as follows (Christophoridis et al., 2019; Harmanescu et al., 2011; US EPA, 2019):

$$EDI = \frac{MDI \times MCS}{BW}$$

where, the MDI is the mass of daily alcohol intake in g/day, MCS is the

**Table 2**

Volumes of per capita alcohol consumption and daily ethanol intake from recorded and unrecorded spirits on average consumption on a population level, consumption by regular drinkers only, and consumption by chronic heavy drinkers<sup>a</sup>

sex	average consumption on a population level [litres of absolute ethanol per capita]		regular drinkers only [litres of absolute ethanol per capita]		chronic heavy drinkers, version A [litres of absolute ethanol per capita]		chronic heavy drinkers version B [litres of absolute ethanol per capita]	
	recorded spirits	unrecorded spirits	recorded spirits	unrecorded spirits	recorded spirits	unrecorded spirits	recorded spirits	unrecorded spirits
men	5.35 (11.5 g/day)	2.52 (5.4 g/day)	6.74 (14.5 g/day)	3.16 (6.8 g/day)	7.77 (16.8 g/day)	3.66 (7.9 g/day)	27.75 (60 g/day)	27.75 (60 g/day)
women	1.26 (2.7 g/day)	0.59 (1.2 g/day)	2.33 (5.0 g/day)	1.09 (2.3 g/day)	5.18 (11.2 g/day)	2.44 (5.2 g/day)	18.50 (40 g/day)	18.50 (40 g/day)
both sexes	3.2 (6.9 g/day)	1.5 (3.2 g/day)	4.80 (10.3 g/day)	2.25 (4.8 g/day)	–	–	–	–

<sup>a</sup> For detailed methodology see supplement 1.

**Table 3**

Oral reference doses of the detected heavy metals

Nr.	heavy metal	oral reference dose [mg/kg/day]
1	chromium (III)	1.5
2	cobalt	0.0003
3	copper	0.04
4	iron	0.7
5	manganese	0.024
6	nickel	0.011
7	zinc	0.3
8	tin	0.6

concentration of different heavy metals in spirits in mg/g, and BW is the average body weight, here taken as  $73.9 \pm 14.9$  kg for both sexes and separately for men ( $82.0 \pm 13.1$  kg) and women ( $67.2 \pm 12.8$  kg) supposing a normal distribution (EFSA, 2012; Marmet et al., 2014). Probabilistic Monte Carlo simulations were carried out with @Risk for Excel software, version 7.6 (Palisade Corporation, Ithaca, NY, USA) using 10,000 iterations, Latin Hypercube sampling, and Mersenne Twister random number generator (Bujdosó et al., 2019). To obtain the probability density functions for the concentrations of heavy metals (mg/g), the best fit distributions were selected using the Akaike information criterion test with a lower limit fixed at zero. These probability density functions and the mass of alcohol intake (gram/day) were included in our model. Then the probabilities were combined using Monte Carlo simulation to get the distribution of EDI at population level.

Three exposure scenarios were employed. The first was termed “average”, using data on per capita consumption averaged across the entire population aged 15+. The second, termed “regular”, uses data on all drinkers (defined as total population minus abstainers) aged 15+. The third was divided into subcategories termed “chronic heavy drinkers, version A” (defined as consuming 60 g/day by men and 40 g/day by women taking into consideration the share of recorded and

unrecorded alcohol consumption) and “chronic heavy drinkers, version B” (defined as consuming 60 g/day and 40 g/day for men and women, respectively). The corresponding figures for alcohol consumption are shown in Table 2 (with data sources described in detail in Supplement 1). For each scenario, daily intake was expressed in mg/kg body weight/day for each metal.

The next step was to calculate THQ values. THQ is a method developed to estimate the non-carcinogenic health risk associated with long term exposure to chemicals (Christophoridis et al., 2019; Harmanescu et al., 2011; US EPA, 1989). Here, the THQ is the ratio of the oral dose of a heavy metal to its reference level (Christophoridis et al., 2019; Harmanescu et al., 2011; US EPA, 1989). If the ratio is less than or equal to 1.0 the hazard is considered to be negligible, while values above 1.0 indicate an increased health risk (Christophoridis et al., 2019; Harmanescu et al., 2011; US EPA, 1989). When calculating THQ values, the distribution of EDI at population level, single values of the exposure frequency (EF, days/year), the exposure duration (ED, years), the reference dose of the heavy metal (RfD, mg/kg/day), and the average exposure time (AET) for non-carcinogens (365 days/year x ED) are taken into consideration (Christophoridis et al., 2019; Harmanescu et al., 2011; US EPA, 1989). In our study the EF was considered to be 365 days per year. The ED was defined as the average life expectancy of the Hungarian population at age 15, for both sexes (61.8 years), and separately for males (58.1 years), and females (65.2 years) (WHO, 2019). The following formula was used (Christophoridis et al., 2019; Harmanescu et al., 2011; US EPA, 1989):

$$THQ = \frac{MCS \times MDI \times EF \times ED}{RfD \times BW \times AET}$$

Then the distribution of EDI at population level and the single values described above were combined using Monte Carlo simulation to get the distribution of THQ values in the exposed population.

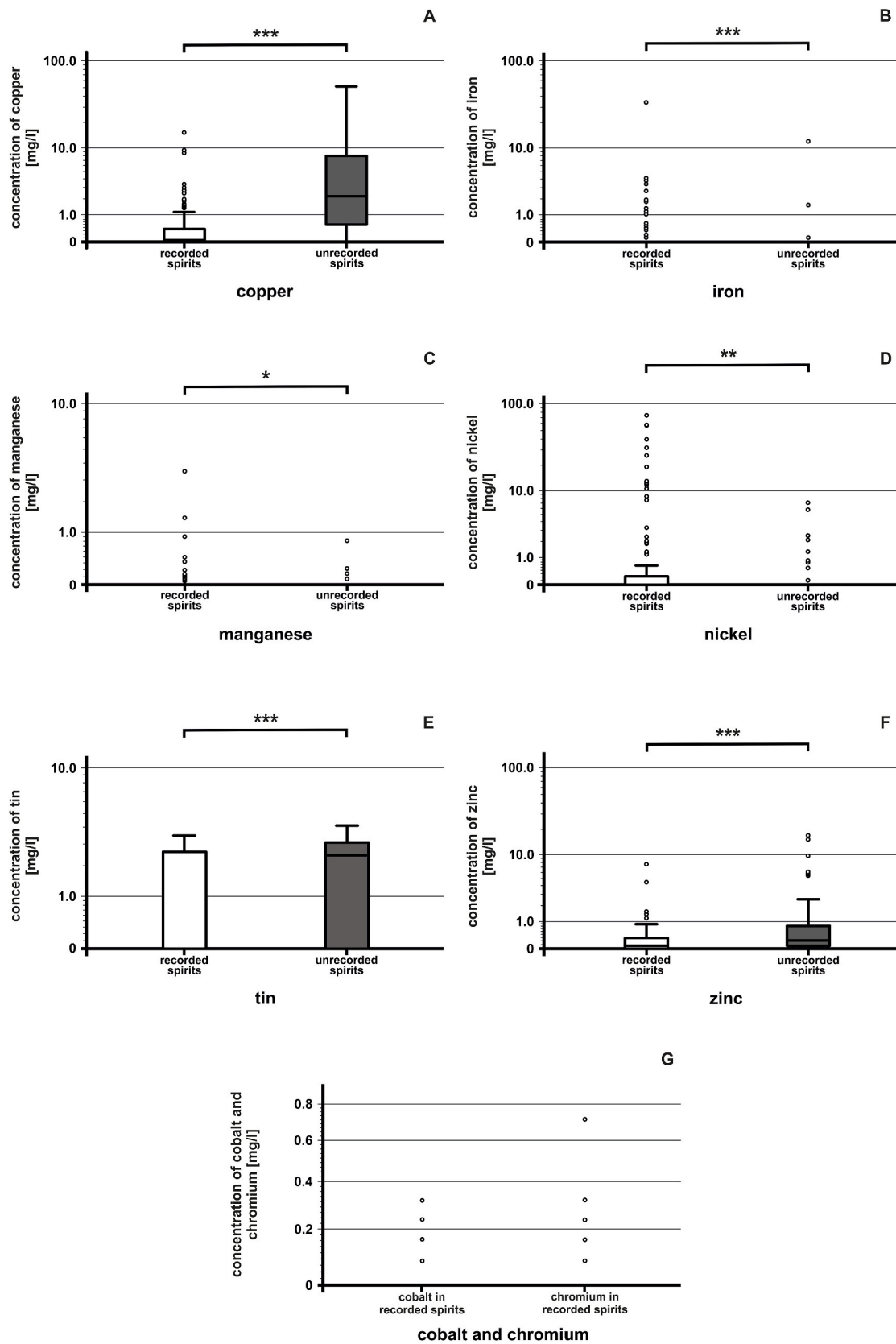
To estimate the total health risk from combined exposure to heavy metals detected in recorded and unrecorded spirits, the distribution of

**Table 4**

Average concentrations of heavy metals in recorded spirit samples and compliance with AMPHORA<sup>a</sup> threshold values

spirit category	AMPHORA threshold values of heavy metals [mg/litre]	heavy metals detected in recorded spirit samples							
		copper	cobalt	chromium	iron	manganese	nickel	zinc	tin
recorded spirits (n = 97)	average concentration of heavy metals $\pm$ SD [mg/litre]	0.71 $\pm$ 2.05	0.01 $\pm$ 0.05	0.03 $\pm$ 0.13	0.65 $\pm$ 3.57	0.07 $\pm$ 0.37	4.29 $\pm$ 13.04	0.33 $\pm$ 0.90	0.93 $\pm$ 1.34
	minimum – maximum concentration of heavy metals [mg/litre]	0.0–15.3	0.0–0.31	0.0–0.71	0.0–34.63	0.0–3.38	0.0–77.16	0.0–7.49	0.0–3.47
	proportion of samples in which heavy metals were detected [%]	62	6	12	23	18	26	76	33
	proportion of samples in which level of heavy metals were above the AMPHORA threshold values [%]	7	–	3	6	3	26	1	33

<sup>a</sup> AMPHORA: Alcohol Measures for Public Health Research Alliance.



**Fig. 1.** Concentration of heavy metals in recorded and unrecorded spirits. Concentration of copper (A), iron (B), manganese (C), nickel (D), tin (E), and zinc (F) in recorded and unrecorded spirits. Concentration of cobalt and chromium (G) in recorded spirits. Median concentrations of heavy metals, their interquartile ranges, and 1.5 times of interquartile ranges as whiskers are shown. Outlier values are indicated by open circles. \* $p < 0.05$ , \*\* $p < 0.01$ , \*\*\* $p < 0.001$ .

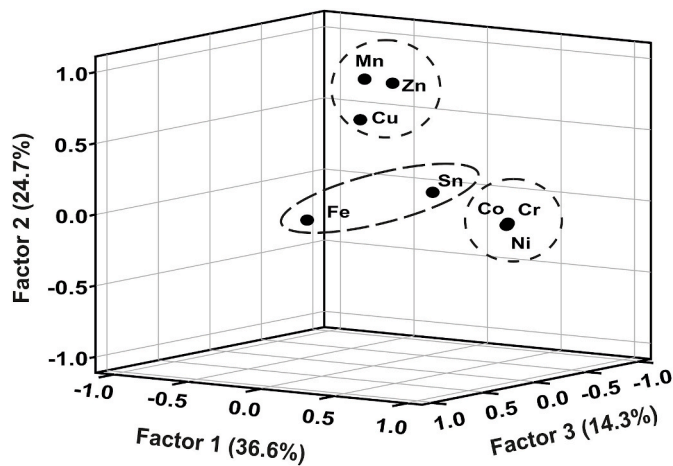


Fig. 2. Groups of related metals in recorded spirits. Groups of related heavy metals (factors) identified in recorded spirits by factor analysis are presented. The loading plot shows the influence of individual heavy metal on the groups of related heavy metals and variance accounted by each factor. Factor 1: Mn, Zn, Cu; Factor 2: Co, Cr, Ni; Factor 3: Sn, Fe.

THQ values of each heavy metal were summed to calculate the distribution of combined THQ (THQ<sub>c</sub>), also known as the hazard index at population level (Christophoridis et al., 2019; Harmanescu et al., 2011; US EPA, 1989). The RfD values of heavy metals used in this study are presented in Table 3 (US EPA, 2019).

2.5. Statistical analysis

Statistical analysis was carried out as described previously (Bujdosó et al., 2019). In summary, levels of heavy metals were expressed in mg/litre of pure ethanol to ensure comparability of the measured concentrations in spirit samples containing different amounts of ethanol. The concentrations of heavy metals were considered to be zero when their levels in spirits were below the limit of quantitation of the ICP OES analysis. For statistical analyses, the samples were divided into two groups, recorded (n = 97) and unrecorded (n = 100) spirits. The recorded spirits were divided further into four subgroups comprising pálinka (n = 25), whiskey (n = 21), vodka (n = 16), and brandy (n = 18). All unrecorded spirits (n = 100) were pálinka so were not subcategorized. Rum (n = 6), artificially flavoured spirits (n = 5), gin (n = 3), tequila (n = 2), and absinth (n = 1) were not subcategorized due to small sample sizes. The Shapiro-Wilk test showed that the data were non-normally distributed so differences in concentrations of heavy metals between recorded and unrecorded spirits were determined using Mann-Whitney U tests.

Descriptive statistics, including average ± standard deviation (SD), minimum and maximum values, percentage of samples in which heavy metals were detected, and the proportion of samples in which the level of heavy metals was above the AMPHORA threshold values are shown in Tables 4 and 5

The distributions of THQ<sub>c</sub> values were compared using Kruskal-Wallis test with Dunn-Bonferroni post hoc method. To identify clusters of heavy metals (factors), a factor analysis (FA) was performed using principal component analysis and varimax with Kaiser normalization as

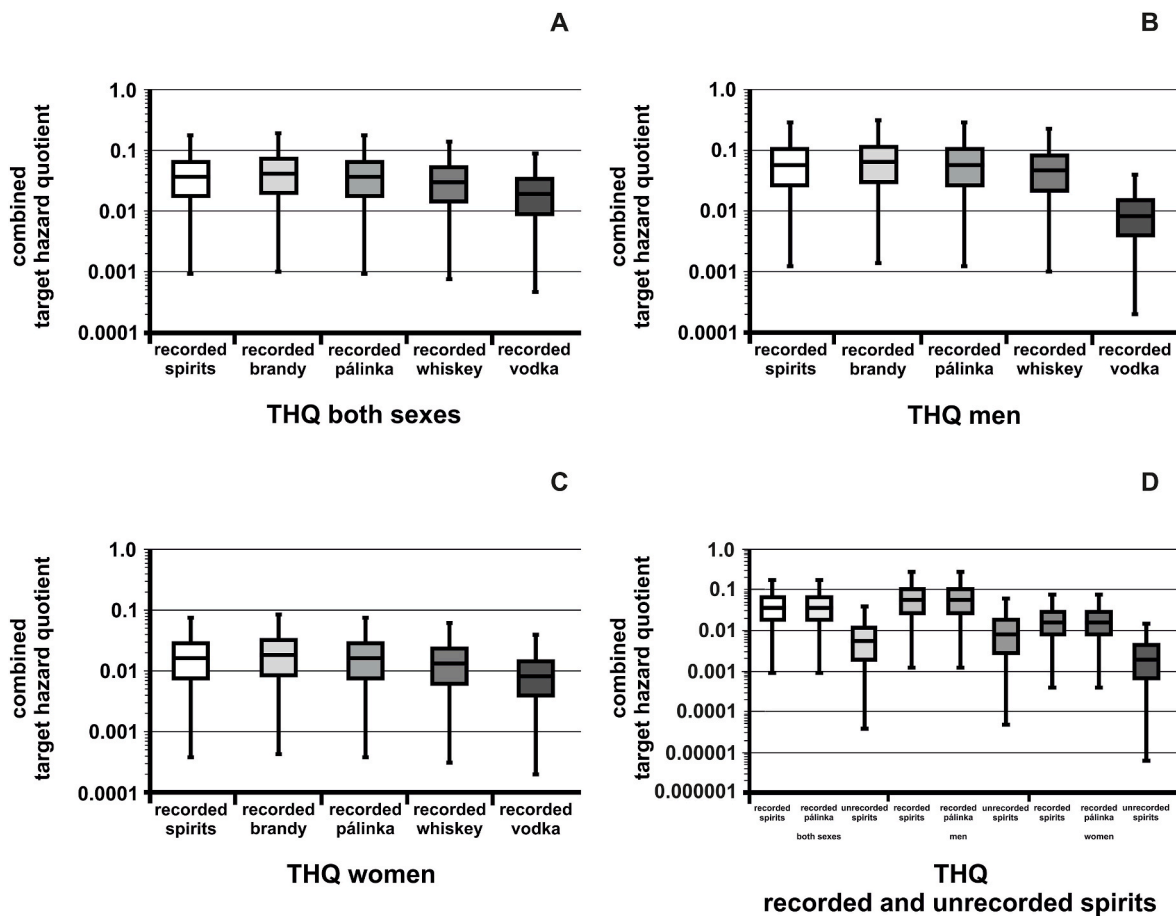
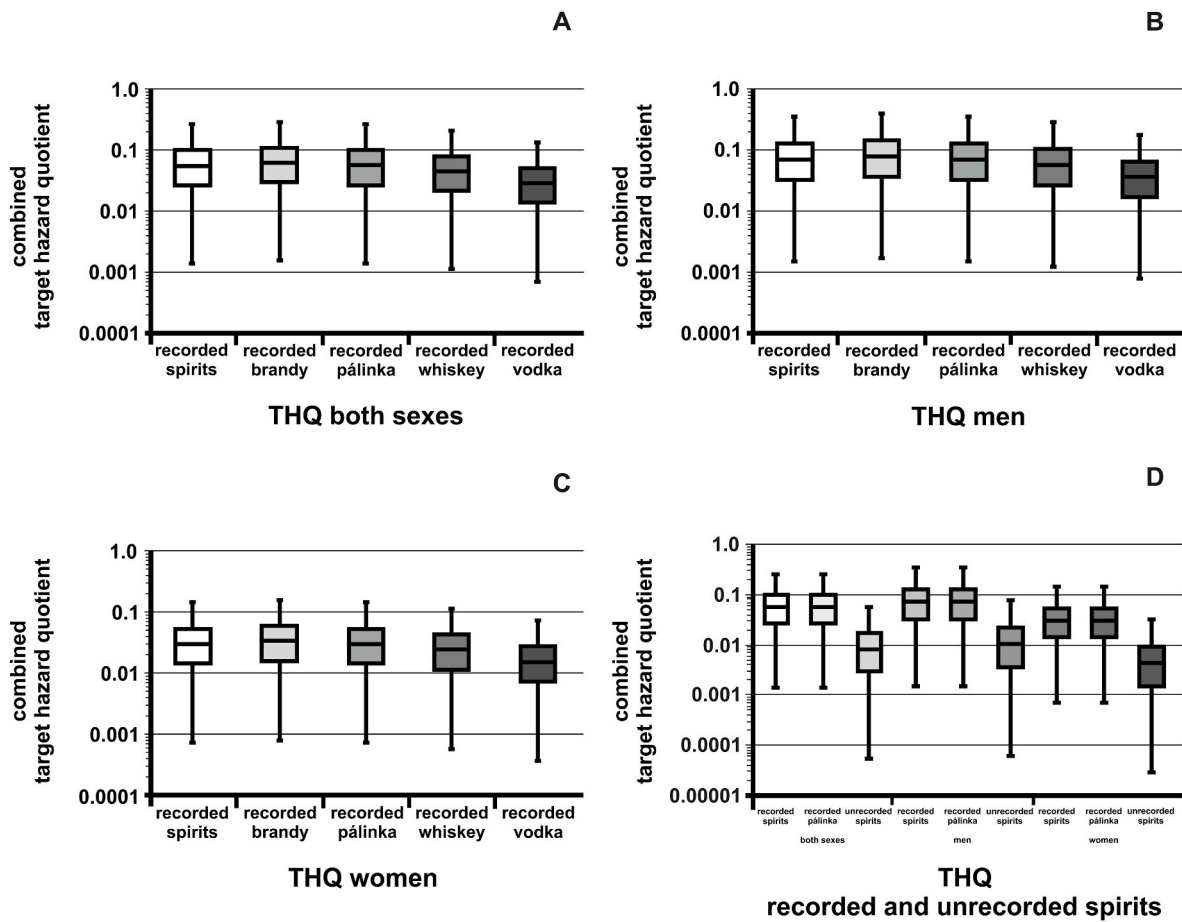


Fig. 3. Health risk from combined exposure to heavy metals in recorded and unrecorded spirits by average drinkers. Panels A, B, and C show the Combined Target Hazard Quotient (THQ<sub>c</sub>) values for both sexes, men, and women consuming recorded spirits, respectively. Panel D illustrates THQ<sub>c</sub> values for both sexes, men, and women drinking recorded and unrecorded spirits. Median values of THQ<sub>c</sub>, their interquartile ranges, 1st and 99th percentiles are depicted.



**Fig. 4.** Health risk from consumption of recorded and unrecorded spirits by regular drinkers. Panels A, B, and C show the Combined Target Hazard Quotient (THQ<sub>c</sub>) values for both sexes, men, and women consuming recorded spirits, respectively. Panel D depicts THQ<sub>c</sub> values for both sexes, men, and women drinking recorded and unrecorded spirits. Median values of THQ<sub>c</sub>, their interquartile ranges, 1st and 99th percentiles are demonstrated.

extraction and rotation method, respectively. Groups of heavy metals with eigenvalues higher than 1.0 were considered to be related. Results of the FA are shown in [Supplementary Tables 5–7](#) and in [Fig. 2](#). Statistical analyses were carried out using IBM SPSS statistics 25.0 software (IBM Inc, Armonk, New York, USA). Values of  $p < 0.05$  were considered statistically significant. Median concentrations of heavy metals, their interquartile ranges, and 1.5 times the interquartile ranges (as whiskers) are shown in [Fig. 1](#), panels A–G. Outlier values are indicated by open circles. Median values of THQ<sub>c</sub>, their interquartile ranges, 1st and 99th percentiles are depicted in [Figs. 3–6](#).

**3. Results**

As shown in [Fig. 1](#), panels A–F, Cu, Fe, Mn, Ni, Zn, and Sn were detected both in recorded and unrecorded spirits, Co and Cr were found

only in recorded spirit samples ([Fig. 1. G](#)). The concentrations of Cu ([Fig. 1. A](#)), Zn ([Fig. 1. F](#)) and Sn ([Fig. 1. E](#)) were significantly higher ( $p < 0.001$ ) in unrecorded spirits than those in their recorded counterparts. Compared to unrecorded spirits, significantly higher levels of Fe ([Fig. 1. B](#),  $p < 0.001$ ), Mn ([Fig. 1. C](#),  $p < 0.05$ ), and Ni ([Fig. 1. D](#),  $p < 0.001$ ) were measured in recorded spirits. The level of Pb was below the limit of quantitation. No Cd was detected in our samples.

[Tables 4 and 5](#) show that Cu, Zn, and Sn were found more frequently in unrecorded than recorded spirits, with Cu present in 99% and 62%, Zn in 95% and 76%, and Sn in 52% and 33% of the samples, respectively. In contrast, Fe, Mn, and Ni were detected more often in recorded spirits when compared with their unrecorded counterparts, with Fe present in 23% and 4%, Mn in 18% and 13%, and Ni in 26% and 11% of the samples, respectively ([Tables 4 and 5](#)). Comparison with AMPHORA threshold values showed that 33% and 26% of the samples of recorded

**Table 5**  
Average concentrations of heavy metals in unrecorded spirit samples and compliance with AMPHORA<sup>a</sup> threshold values

		heavy metals detected in unrecorded spirit samples					
		copper	iron	manganese	nickel	zinc	tin
spirit category	AMPHORA threshold values of heavy metals [mg/litre]	2.0	2.0	0.5	0.2	5.0	1.0
unrecorded spirits (n = 100)	average concentration of heavy metals ± SD [mg/litre]	5.51 ± 8.06	0.13 ± 1.19	0.01 ± 0.08	0.21 ± 0.93	1.08 ± 2.62	1.59 ± 1.56
	minimum – maximum concentration of heavy metals [mg/litre]	0.0–51.60	0.0–11.91	0.0–0.79	0.0–6.86	0.0–16.96	0.0–4.10
	proportion of samples in which heavy metals were detected [%]	99	4	13	11	95	52
	proportion of samples in which level of heavy metals were above the AMPHORA threshold values [%]	51	1	1	10	7	52

<sup>a</sup> AMPHORA: Alcohol Measures for Public Health Research Alliance.

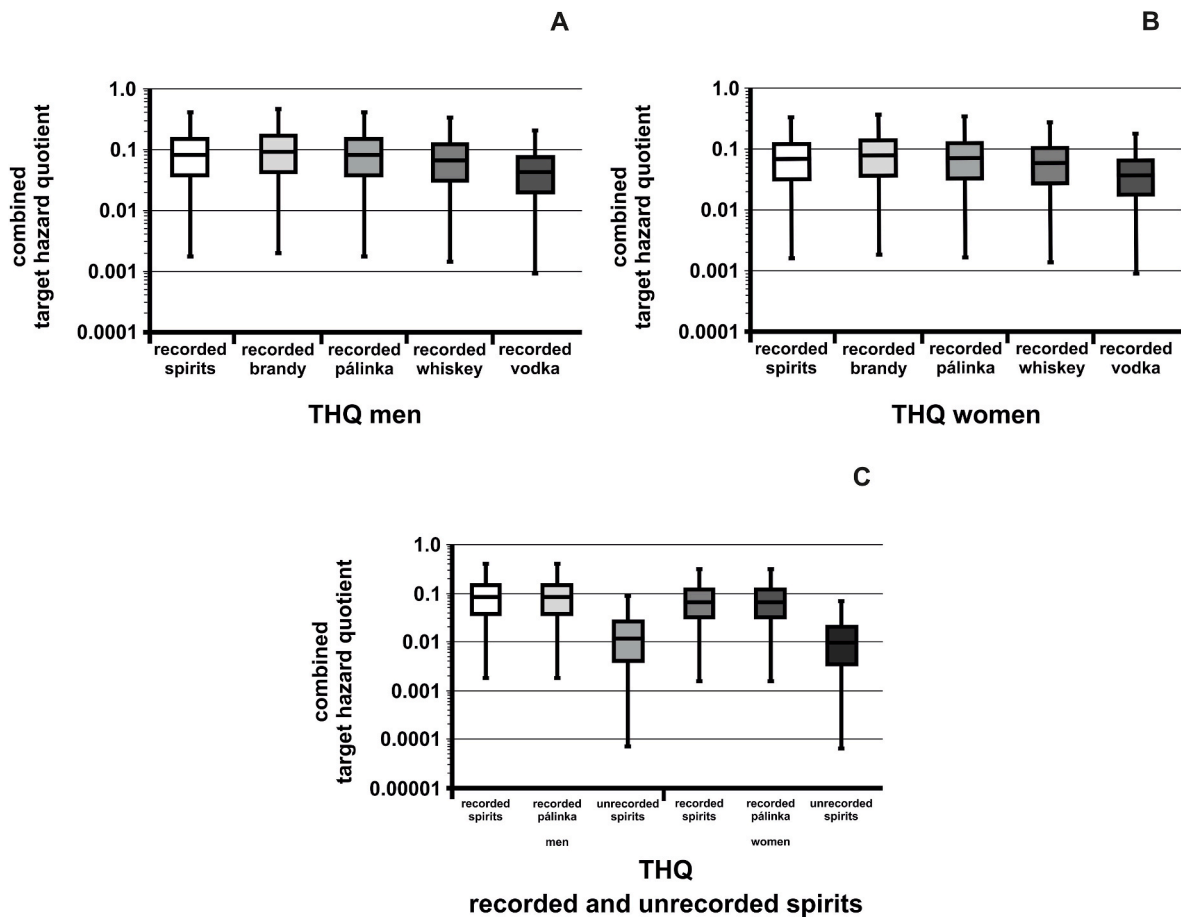


Fig. 5. Health risk from consumption of recorded and unrecorded spirits by heavy drinkers (version A). Panels A and B show the Combined Target Hazard Quotient (THQ<sub>c</sub>) values for men and women consuming recorded spirits, respectively. Panel C depicts THQ<sub>c</sub> values for men and women drinking recorded and unrecorded spirits. Median values of THQ<sub>c</sub>, their interquartile ranges, 1st and 99th percentiles are illustrated.

spirits contained Sn and Ni above recommended limits respectively (Table 4). Table 5 shows that Sn and Cu were detected in 52% and 51% of unrecorded spirit samples at concentrations higher than AMPHORA threshold values, respectively. The occurrence of heavy metals in recorded brandy, whiskey, and vodka samples and their relationship with AMPHORA limits are presented in Supplementary Tables 1–4.

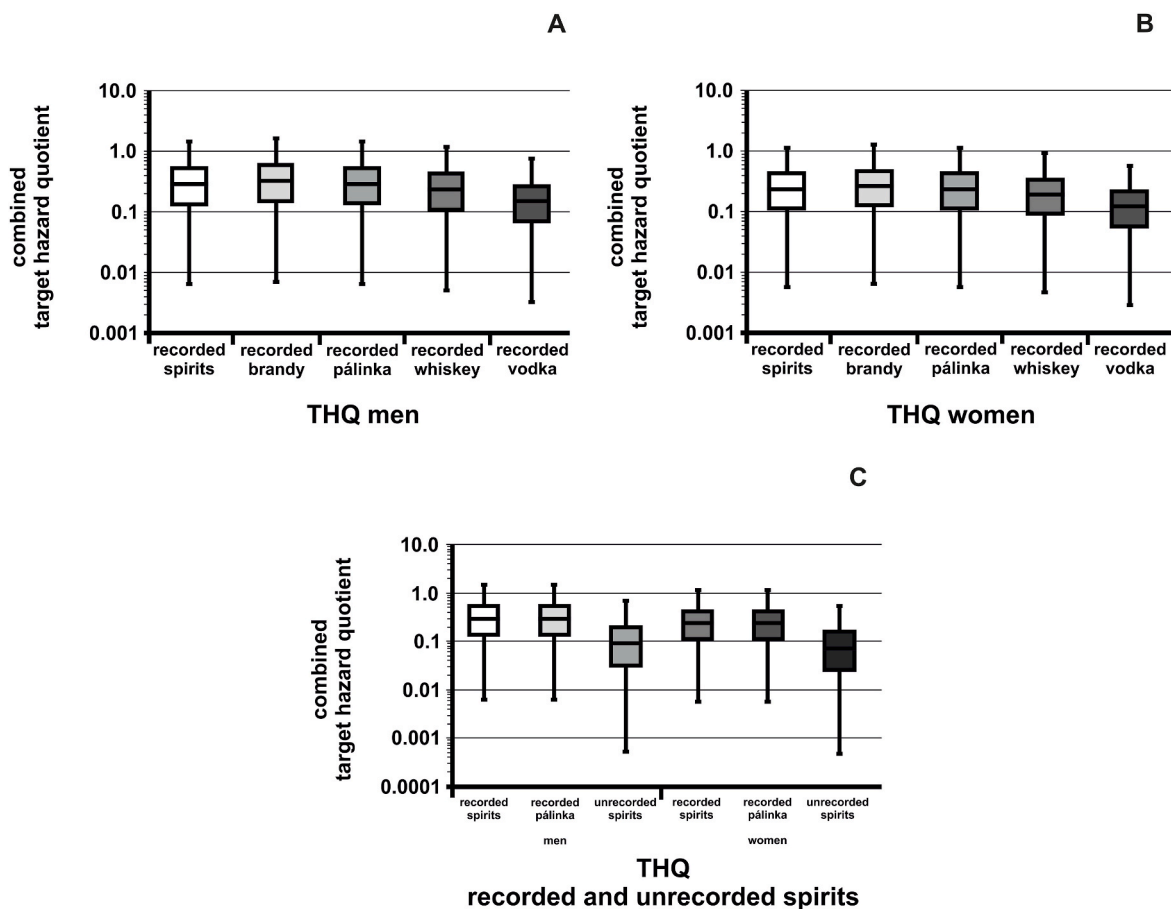
Following factor analysis, three groups (factors) of heavy metals detected in recorded spirits were identified (75.6% of total variance, Fig. 2.). The first, second, and third group (factor) included Mn, Zn, and Cu (36.6% of total variance), Co, Cr, and Ni (24.7% of total variance), and Fe and Sn (14.3% of total variance), respectively (Supplementary Tables 5–7, Fig. 2.). Data on the concentration of heavy metals determined in unrecorded spirits did not fulfil the assumptions necessary for factor analysis.

The results of the population-based comparative risk assessment are presented in Figs. 3–6 using the population average, regular and chronic heavy drinkers (version A and B), respectively. Figs. 3–5 show that distributions of THQ<sub>c</sub> values of heavy metals detected in recorded spirits, brandy, pálinka, whiskey, and vodka were below 1.0 for both sexes, and separately for men and women. Compared to those who drink recorded brandy, the THQ<sub>c</sub> values were significantly lower for average (Fig. 3, panels A–C), regular (Fig. 4A–C), and chronic heavy drinkers (version A, Fig. 5A and B) consuming recorded pálinka, whiskey, and vodka. When drinking unrecorded spirits, the THQ<sub>c</sub> values were also below 1.0 at each consumption level (Fig. 3, D, 4, D, 5C, and 6.C). Fig. 6,

panel A shows that distributions of THQ<sub>c</sub> values of heavy metals determined for recorded spirits, brandy, pálinka and whiskey were above 1.0 for men and significantly lower for chronic heavy drinker men (version B) consuming recorded vodka. Fig. 6, panel B depicts that distributions of THQ<sub>c</sub> values of heavy metals calculated for recorded spirits, brandy and pálinka were above 1.0 for women and significantly lower for chronic heavy drinker women (version B) consuming recorded whiskey and vodka. Compared to those chronic heavy drinkers (version B) who consume unrecorded spirits THQ<sub>c</sub> values of heavy metals determined in recorded spirits and recorded pálinka were significantly higher for men and women (Fig. 6C).

#### 4. Discussion

Heavy metals are frequently detected both in recorded and unrecorded spirits (Bonic et al., 2013; Ibanez et al., 2008; Iwegbue et al., 2014a; Lachenmeier et al., 2008; Newman et al., 2017, 2018; Tatarková et al., 2019), but little attention so far has been paid to their adverse effects on health (Ibanez et al., 2008; Lachenmeier et al., 2011a). Although there are toxicological threshold values for several heavy metals (Lachenmeier et al., 2011a), measurement of their concentrations in spirit samples only indicates whether their levels are below or above threshold limits. However, this approach does not capture the overall exposure and is not suitable for quantitative health risk assessment because levels above toxicological threshold values do not



**Fig. 6.** Health risk from consumption of recorded and unrecorded spirits by heavy drinkers (version B). Panels A and B show the Combined Target Hazard Quotient (THQ<sub>c</sub>) values for men and women consuming recorded spirits, respectively. Panel C depicts THQ<sub>c</sub> values for men and women drinking recorded and unrecorded spirits. Median values of THQ<sub>c</sub>, their interquartile ranges, 1st and 99th percentiles are demonstrated.

necessarily indicate increased health risk. Consequently, more complex methodologies are required, including THQ analysis (Iwegbue et al., 2014a, Iwegbue et al., 2014b; Otim et al., 2019). However, only a few studies using THQ analysis to estimate risk from exposure to heavy metals in spirits have been published (Iwegbue et al., 2014a, Iwegbue et al., 2014b; Otim et al., 2019). In addition, the previous studies provided only a point estimate of the health risk and did not take into account the diversity of risk arising from differences in sex and patterns of alcohol consumption (Iwegbue et al., 2014a, Iwegbue et al., 2014b; Otim et al., 2019). Although a probabilistic risk assessment was carried out previously, it examined only the health risk from consumption of heavy metals from unrecorded spirits using a margin of exposure approach (Lachenmeier and Rehm, 2013). Nor has the health risk from consumption of Cr, Co, Cu, Fe, Mn, Ni, Sn, and Zn in recorded and unrecorded spirits been compared (Lachenmeier et al., 2012). To overcome these limitations and provide a comprehensive health risk estimation, we used a probabilistic risk assessment approach, including THQ analysis with different exposure scenarios.

To carry out the health risk assessment, first the concentrations of heavy metals in recorded and unrecorded spirits were determined. The concentrations of heavy metals reported in previous studies were also recorded (Bonic et al., 2013; Ibanez et al., 2008; Iwegbue et al., 2014a; Lachenmeier et al., 2011b). Bonic et al. (2013) found that the average concentrations of Cu, Fe, Mn and Zn in unrecorded plum brandies were 3.9, 1.4, 0.4, and 0.3 mg/L, respectively. They also reported mean levels of these metals in recorded plum brandies as 3.3 (Cu), 0.4 (Fe), 0.4 (Mn), and 1.1 (Zn) mg/litre (Bonic et al., 2013). By reviewing a large number of studies on the concentration of metals in recorded alcoholic

beverages, Ibanez et al. (2008) found that vodka, whiskey, and brandy contained Cu, Fe, and Zn at concentrations of 0.1–14.6, 0.0–2.3, and 0.0–20 mg/L, respectively. Our measurements of levels of Cu, Fe, Mn, and Zn (see Tables 4 and 5) are comparable to those reported by these studies (Bonic et al., 2013; Ibanez et al., 2008). Concentrations of Sn in recorded and unrecorded spirits have not been reported so we could not make any comparison. In addition, only a few investigations have reported data on the concentration of Ni in spirits (Iwegbue et al., 2014a; Lachenmeier et al., 2011b). Lachenmeier et al., 2011b and Iwegbue et al., 2014a reported that average levels of Ni in unrecorded and recorded spirits were 0.23 mg/L and 0.13 mg/L, respectively. In our results, this heavy metal was present in 11% of unrecorded and 26% of recorded spirit samples, with mean concentrations of 0.21 and 4.29 mg/L in unrecorded and recorded spirits, respectively (Tables 4 and 5). Comparing our data to those reported by Lachenmeier et al., 2011b and (Iwegbue et al., 2014a), the average concentration of Ni in our samples were similar in unrecorded, and higher in recorded spirits. Several factors could be responsible for the discrepancy with recorded spirits. First, the type of alcoholic beverages was different from those analysed by Iwegbue et al., 2014a. Unlike their investigation, which included alcoholic beverages with ethanol content ranging from 5% to 60% (% v/v), our recorded spirit samples contained 31%–80% (% v/v) ethyl-alcohol. To ensure comparability of the measured concentrations in spirits, the levels of heavy metals have to be expressed in mg/litre of pure ethanol (Lachenmeier and Rehm, 2013). We did convert concentrations to mg/litre of pure alcohol but it was not clear whether this was done in their study (Iwegbue et al., 2014a), making direct comparison of levels of Ni difficult. Second, we had several outliers in terms of Ni



concentrations, which could explain the higher average levels in our recorded samples (Kirkwood and Sterne, 2003). Although the level of Pb was below the limit of quantitation in our samples, we agree with the conclusions of previous studies suggesting that consumption of unrecorded alcohols containing Pb at higher concentrations can pose a health risk for the consumers in the CEE countries (Lachenmeier, 2020; Tatarková et al., 2019).

The factor analysis identified three clusters of heavy metals in recorded spirits. The first (factor 1) and second (factor 2) included Mn, Zn, Cu and Co, Cr, Ni, respectively. Previous studies have shown that heavy metals can be released into spirits from equipment used for distillation and storage (Fuller et al., 2010; Ibanez et al., 2008). Stills are often made of metal alloys containing Mn, Zn, and Cu (Fuller et al., 2010; Ibanez et al., 2008) while stainless steel containers used for storage of recorded spirits contain Co, Cr, and Ni (Fuller et al., 2010; Ibanez et al., 2008). Our findings suggest that heavy metals in the first and second group could originate from these common sources. The third group (factor 3) comprised Sn, Fe, with concentrations negatively correlated. Thus, we assume that these heavy metals were from different sources.

Since several recorded and unrecorded spirit samples contained one or more heavy metals at concentrations above the AMPHORA threshold values, it was reasonable to investigate whether there is any health risk from combined exposure to them. Our population-based comparative risk assessment showed that while the concentration of heavy metals exceeded the AMPHORA limits in a large proportion of recorded and unrecorded spirit samples, this was not so much that would pose any health risk to average, regular, and chronic heavy drinkers (version A). In contrast, chronic heavy drinkers (version B) may be at increased health risk when consuming recorded spirits. However, this increased risk is likely to be negligible and may affect only a few percentages of chronic heavy drinkers. Consequently, there are no grounds to worry about the adverse effects of heavy metals from spirits.

#### 4.1. Strengths and limitations

Our study has certain strengths but also limitations. Our investigation is the first to compare the concentration of heavy metals in recorded and unrecorded spirits. Another strength is that there have been no previous studies that assessed health risks from combined exposure to heavy metals in recorded and unrecorded spirits and considered differences by sex and patterns of alcohol consumption. In addition, our health risk assessments were based on internationally accepted reference doses. The limitations include that our spirit samples were limited to those available in Hungary, which limits the generalizability of our results. Second, drinking patterns can change over time but the data required to consider how they change by sex, age, and location currently are not available (Griswold et al., 2018). Third, our risk assessments were based on the calculation of combined THQs. However, an important limitation of this method is that  $THQ_c$  does not consider the target organ toxicity and the health effects that could arise from the interaction among heavy metals and between them and ethanol when consumed simultaneously (Lachenmeier et al., 2012; Sarigiannis and Hansen, 2012). Furthermore,  $THQ_c$  values do not completely consider the differences in toxicity of various heavy metals and their antagonistic or synergistic effects. This could result in an under- or overestimation of health risk in our study. Therefore, further toxicological studies are needed to determine the interactions between heavy metals to provide more precise health risk assessments.

#### 4.2. Conclusion

Our results showed that the concentrations of Cu, Zn, and Sn in unrecorded spirits were significantly higher than those of in their recorded counterparts. We also found that, when compared to their unrecorded counterparts, recorded spirits contained significantly higher

levels of Fe, Mn, and Ni. The findings of our comparative risk assessment demonstrated that combined exposure to heavy metals in recorded and unrecorded spirits posed no health risk for average, regular and chronic heavy drinkers (version A) and the health risk associated with consumption of recorded spirits was increased in chronic heavy drinkers (version B). However, when compared to the health risk arising from drinking large volumes of ethanol, the risk is negligible. Therefore, we should not worry about the adverse health outcomes of heavy metal intake from recorded and unrecorded spirits.

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#### Declaration of competing interest

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

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#### Appendix A. Supplementary data

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