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Concentrations of heavy metals (Mn, Co, Ni, Cr, Ag, Pb) in coffee

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Aim: Technologies involved in roasting coffee beans, as well as the methods used to prepare infusions, vary according to culture, and contribute to differences in the concentration of elements in the drink. Materials and Methods: Concentrations of six elements: manganese (Mn), cobalt (Co), nickel (Ni), chrome (Cr), silver (Ag) and lead (Pb) were investigated in coffee infusions from eleven samples of coffee, roasted and purchased in four countries: Bosnia and Herzegovina, Brazil, Lebanon and Poland. Metal concentrations were determined using an induction coupled plasma technique in combination with mass spectrometry (ICP-MS, Perkin Elmer) which measures total metal (ionic and non-ionic) content. Results: Metal intake estimated for individual countries (in the respective order; mean consumption per person per year) was as follows: Mn: 26.8-33.1, 28.3-29.5, 29.7, 12.6-18.9 mg; Co: 0.33-0.48, 0.42-0.35, 0.32, 0.12-0.17 mg; Ni: 3.83-5.68, 4.85-5.51, 4.04, 2.06-2.24 mg; Cr: 0.17-0.41, 0.21-0.47, 0.17, 0.09-0.28 mg; Ag: 0.16-1.13, 0.26-0.70, 0.61, 0.33-1.54 mg, Pb: 4.76-7.56, 3.59-5.13, 3.33, 1.48-2.43 mg. Conclusions: This finding gives new data for Mn, Co, Ni, Cr, and Ag intake from coffee, and suggests that the amounts are negligible. However, the data for Pb consumption in heavy drinkers, for example in Bosnia and Herzegovina, indicate that Pb intake from coffee may contribute to the disease burden. The high lead level in some coffees suggests the need for a more precise control of coffee contamination.

Key words: coffee infusion, global disease burden, lead poisoning, toxicity

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INTRODUCTION

Coffee is the second most popular drink after water in the world. Coffee consumption varies widely according to geographical location. The highest consumption has been observed in Northern Europe (Finland; 12.0 kg per capita/year) whereas in Southern Europe the highest consumption is in Bosnia and Herzegovina (6.1 kg per capita/year). According to the International Coffee Organization, in 2008 coffee was consumed at a rate of 2.5 billion cups per day (1 cup = 30 mL). This consumption is a model for addictive behavior. Genetic investigations in twins suggest that the heritability of coffee intake can be estimated to be in the range of 39 to 56% (Laitala et al., 2008; Vink et al., 2009). Coffee seems to have distinct acute and long-term effects on health.

Interestingly, its consumption has been suggested to be beneficial in dementia, Alzheimer's disease, Parkinson's disease and diabetes mellitus type 2 (Gongora-Alfaro, 2010; Hjellvik et al., 2011). However, other researchers have associated coffee drinking with an increased risk of developing coronary heart disease (Montagnana et al., 2012). Higdon et al. estimated that consumption of more than 5 cups per day increased the risk of coronary heart disease from 40 to 60% (Higdon & Frei, 2006). Opinions on the influence of coffee drinking on the development of cancer is unclear. Some researchers suggest that coffee intake increases certain types of cancer (Silvera et al., 2007; Sugiyama et al., 2010), but others have provided contradictory results (Li et al., 2013). The consumption of large quantities of this beverage is not recommended for pregnant women because it increases the risk of miscarriage (Higdon & Frei, 2006). It is also not known to what extent caffeine is the cause of these negative effects, or whether other constituents are also of importance, for example heavy metals.

Heavy metals are widely dispersed in the environment. They enter the food chain and occur in varying concentrations in human food (Roychowdhury et al., 2003). The contamination of food is a serious problem as heavy metals are taken up from the digestive tract and exhibit harmful influence on many tissues. Some metals exhibit toxic properties in relatively low doses and, moreover, they gradually accumulate in tissues (Beckett et al., 2007). Metals disturb the ionic balance and mineral regulation, induce oxidative damage to cell structures, produce injury to DNA and induce cancer transformations (Wallkers, 2003).

The average annual coffee consumption per capita is: 6.1 kg in Bosnia and Herzegovina; 5.8 kg in Brazil; 4.8 kg in Lebanon and 2.4 kg in Poland. The chemical composition of coffee is very complex and depends on the place of origin and species/cultivar of the coffee plant. The technology used in the preparation and industrial processing of green beans, as well as the methods consumers use to prepare their coffee modify the concentrations of the substances in the final product. Additionally, potential contamination may derive from package and storage. The chemical composition of coffee is so complex that it is not possible at present to provide a complete list of all constituents. Therefore we discuss here only a few chemical species (namely some

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metals), our choice being based on their importance and also on the availability of data and their effect on public health. We investigated the concentration of six elements: Mn, Co, Ni, Cr, Ag and Pb in coffee infusions from eleven samples of coffee roasted and purchased in four countries: Bosnia and Herzegovina, Brazil, Lebanon and Poland. (Our previous study focused on Fe, Zn and Cu and showed no health hazard associated with exposure to these metals *via* coffee consumption (Iwanowska *et al.*,2013)).

MATERIALS AND METHODS

The study comprised eleven samples of coffee: three from Bosnia and Herzegovina, three from Brazil, one from Lebanon and four from Poland. The coffees were roasted in the country of purchase. Coffee plants for the Bosnia and Herzegovina, Poland and Brazilian samples were grown in Brazil (the origin of coffee plants in other samples is not known). Mineralization was performed in three replicates for each sample, as follows.

Mineralization of coffee infusions: an infusion (1g coffee in water, volume 27 ml) was filtered through a Whatman paper. 10 mL of the filtrate was mineralized in tubes with 1 mL HNO₃ and and 0.1 mL HClO₄ at 140°C for 5 h. Concentrations of the heavy metals: Mn, Co, Ni, Cr, Ag and Pb were determined by an induction-coupled plasma technique in combination with mass spectrometry (ICP-MS, Perkin Elmer, Warsaw, Poland). Each metal concentration was estimated 3 times from each sample of coffee.

RESULTS

Table 1 shows the countries of purchase of the coffees, and heavy metal contents in the eleven samples of coffee used in the investigation.

The metal content in the coffee infusions for individual countries was as follows: (Bosnia and Herzego-

vina, Brazil, Lebanon, Poland, respectively) Mn: 4.40–5.43; 4.87–5.08; 6.19 and 5.24–7.86 µg/g of coffee; Co: 0.0543–0.0780; 0.0610–0.0718; 0.0660 and 0.0510–0.0714 µg/g of coffee, the Ni: 0.6274–0.9307; 0.8366–0.9496; 0.8423 and 0.8588–0.9326 µg/g of coffee, the Cr: 0.0277–0.0672; 0.0359–0.0816; 0.351 and 0.0376–0.1183 µg/g of coffee, Ag: 0.0255–0.1851; 0.0451–0.1202; 0.1267 and 0.0643–0.1367 µg/g of coffee and Pb: 0.781–1.2393; 0.6185–8840; 0.6933 and 0.6154–1.0143 µg/g of coffee.

The lowest Mn, Ni, Cr and Ag content was found in coffee from Bosnia and Herzegovina: 4.40, 0.6274, 0.0277 and 0.0255 µg/g of coffee, respectively, and the lowest Co and Pb in that from Poland: 0.0510 and 0.6154 µg/g of coffee (respectively).

The highest Co, Ag and Pb content was found in coffee from Bosnia and Herzegovina: 0.0780; 0.1851; 1.2393 µg/g of coffee, respectively. The highest Mn and Cr content was found in coffee from Poland: 7.86 and 0.1183 µg/g of coffee, respectively, and the highest Ni was in coffee from Brazil, 0.9496 µg/g of coffee.

DISCUSSION

The estimated intake of Mn, Co, Ni, Cr, Ag and Pb per capita per year, based on coffee consumption values obtained from (http://chartsbin.com, 03.2013) are shown in Table 2. The highest coffee consumption is in Bosnia and Herzegovina, where people tend to brew the coffee the Turkish way i.e., the coffee is boiled together with the water. Note that the consumption values are averaged over the entire population- which means that coffee drinkers will tend to ingest more coffee than the values shown. In Bosnia on average 64% of the adult population declare themselves to be coffee drinkers, including 69% women and 59% men.

Some recent epidemiological and experimental studies have shown positive effects of coffee drinking on various aspects of health. On the other hand other studies have shown harmful effects, as listed in the Introduc-

Table 1. Concentrations of Mn, Co, Ni, Cr, Ag and Pb in coffee infusions (µg/g coffee)

	Concentration of Metal (µg/g coffee, s.d.)											
Country of purchase of coffee	Mn		Со		Ni		Cr		Ag		Pb	
	Mean	s.d.	Mean	s.d.	Mean	s.d.	Mean	s.d.	Mean	s.d.	Mean	s.d.
Bosnia and Herzegovina	5.43	0.16	0.055	0.020	0.6274	0.087	0.028	0.001	0.185	0.011	1.239	0.103
	4.40	0.19	0.078	0.027	0.9307	0.050	0.031	0.001	0.026	0.004	1.114	0.020
	5.22	0.35	0.054	0.021	0.7770	0.096	0.067	0.002	0.169	0.013	0.781	0.078
Grand Mean	5.02	0.52	0.06	0.02	0.78	0.15	0.04	0.02	0.13	0.08	1.05	0.22
Brazil	5.08	0.02	0.061	0.023	0.8366	0.034	0.082	0.001	0.120	0.016	0.884	0.042
	4.87	0.04	0.071	0.019	0.9496	0.065	0.036	0.001	0.065	0.006	0.619	0.023
	4.96	0.07	0.072	0.017	0.8679	0.091	0.036	0.001	0.045	0.008	0.628	0.021
Grand Mean	4.97	0.10	0.07	0.02	0.88	0.08	0.05	0.02	0.08	0.04	0.71	0.13
Lebanon	6.19	0.02	0.066	0.007	0.8423	0.023	0.035	0.001	0.127	0.008	0.693	0.019
Poland	6.03	0.09	0.055	0.025	0.8588	0.026	0.118	0.002	0.137	0.006	0.615	0.019
	5.99	0.02	0.071	0.015	0.8708	0.087	0.043	0.001	0.130	0.039	1.008	0.022
	5.24	0.02	0.051	0.021	0.9326	0.062	0.038	0.001	0.099	0.007	1.014	0.014
	7.86	0.03	0.067	0.013	0.8805	0.083	0.039	0.001	0.064	0.007	0.803	0.008
Grand Mean	6.28	1.01	0.06	0.02	0.89	0.07	0.06	0.04	0.11	0.04	0.86	0.17

	Heavy metal intake, range mg/per capita/year									
Country (coffee consumption kg/per capita/year)	Mn	Со	Ni	Cr	Ag	Pb				
Bosnia and Herzegovina (6.1)	26.8–33.1	0.33-0.48	3.83-5.68	0.17-0.41	0.16–1.13	4.76–7.56				
Brazil (5.8)	28.3–29.5	0.35-0.42	4.85–5.51	0.21–0.47	0.26-0.70	3.59–5.13				
Lebanon (4.8)	29.7	0.32	4.04	0.17	0.61	3.33				
Poland (2.4)	12.6–18.9	0.12-0.17	2.06–2.24	0.09–0.28	0.33–1.54	1.48–2.43				

tion. Therefore a study of metal concentrations in coffee is useful because the possible impacts of coffee drinking on health have not yet been clearly defined.

Manganese (Mn)

Food is the most important source of manganese exposure in the general population. The intake can be higher for vegetarians because higher levels of manganese occur in food of plant origin. The highest tissue concentrations of manganese have been observed in the liver, kidney, pancreas, and adrenals. Available data clearly show that manganese can cause adverse effects in humans, the most important target being the central nervous system. The syndrome known as manganism is characterised by apathy, anorexia, muscle pain and slow clumsy movement of the limbs.

The highest intake of Mn from the coffee drink was estimated to be in Lebanon: 29.7 mg per capita/year, and the lowest in Poland, ranging from 12.58–18.86 mg per capita/year. The estimated safe dietary intake per year for adults is 730–1825 mg, established by the U.S. National Research Council (Freeland-Graves, 1994). Thus, the conclusion from our studies is that Mn intake from coffee is negligible compared to the demand. The amount of manganese intake from coffee during a year does not cover even a small percentage of the annual demand for this element.

Cobalt (Co)

In the human body cobalt typically occurs as a component of vitamin B12. This element appears in most tissues, with the highest amount found in the kidneys, liver, and bones. In the human body it is involved in the production of red blood cells and proper functioning of the nervous system. A high cobalt ion concentration in serum has toxic effects on the heart and liver, but no significant effect on the kidney in mice (Liu *et al.*, 2010). The annual demand for cobalt is estimated at 2 mg. In our studies the highest level of Co intake with coffee drinking was estimated to be in Bosnia and Herzegovina and ranged from 0.33–0.48 mg per capita/year, the lowest intake was in Poland and ranged from 0.12–0.17 mg per capita/year. These quantities are much smaller than the annual demand for the element.

Nickel (Ni)

Nickel influences iron absorption and metabolism and the haematopoietic process. Humans may be exposed to nickel by breathing polluted air, drinking water, eating food or smoking cigarettes. Skin contact with nickel-contaminated soil or water may also result in nickel exposure. In small quantities nickel is essential, but when the uptake is too high it can be a danger to human health. The carcinogenicity of nickel and certain nickel compounds have been recognized by the International Agency for Research on Cancer (IARC). An uptake of too large quantities of nickel has the following consequences: lung embolism, respiratory failure, birth defects, asthma and chronic bronchitis, allergic reactions, development of lung, larynx and prostate cancers and heart disorders. The tolerable upper intake level for Ni is 1 mg/day. Our results therefore suggest that Ni intake from coffee, even in Brazil where it was the highest and ranged from 4.85–5.51 mg per capita/year, will have negligible effects on health.

Chromium (Cr)

Chromium is ubiquitous, occurring in water, soil and biological systems. Evaluation is usually limited to the trivalent chromium (Cr III) because this is an essential element in human nutrition. Chromium influences carbohydrate, lipid, and protein metabolism via an effect on insulin action. However, the mechanism is still not clear and neither is the exact structure of the biologically active form of chromium. Chromium-deficient diabetics have seen beneficial effects from chromium intake, since supplementing the diet with chromium decreased fasting blood glucose levels, improved glucose tolerance, lowered insulin levels, and decreased total cholesterol and triglyceride levels (Mooradian et al., 1994). Currently, there is no formal Recommended Dietary Allowance (RDA) for chromium. The highest coffee consumption patterns for Cr were in Brazil and ranged from 0.21-0.47 mg per capita/year.

Silver (Ag)

Silver is one of the transition metals we encounter in our everyday life. The silver ions contained in packages, for example sausage casings, extend the freshness, inhibit bacterial growth and division, easily spread in the environment and penetrate into the food chain. Silver ions are a means of inhibiting the growth of bacteria, mold and mildew. The human organism contains approximately 1 mg of silver, but if this metal accumulates it could cause a skin disease called argyria, manifested by discoloration or blue-grey darkening of the eyes, nose, and throat, or even lead to death. However, research carried out in Spain indicates that silver nanoparticles (AgNPs) are cytotoxic to human tumors, and that it might be of clinical importance that tumor cells are more sensitive to the cytotoxic effect of these nanoparticles (Avalos Funez et al., 2013). The smallest amount of silver reported to produce generalized argyria in humans ranges from 4-5 g to 20-40 g. The lethal toxic dose in humans is

50–500 mg/kg body weight. There are no data on the acceptable daily intake. An important fact is that there are isolated reports of more serious neurologic, renal, or hepatic complications caused by ingesting colloidal silver (Payne *et al.*, 1992). Our results suggest that Ag intake from coffee in Poland, where it was the highest and ranged from 0.33–1.54 mg per capita/year, would have negligible effect on health.

Lead (Pb)

Lead is a heavy metal commonly occurring in nature. The general population is exposed to lead from air and food in roughly equal proportions. Chronic exposure to lead is associated with multiorgan toxicity. No safe threshold for lead exposure has been determined so far, which means that the amount of lead that can be consumed without it being harmful to the body remains unknown (Rossi, 2008). Lead poisoning is known as "plumbism" and is defined by a lead blood concentration above 25 μg/dl for adults (in whole blood) and 5 μg/ dl for children (data from: Agency for Toxic Substances and Disease Registry). Long-term exposure to lead or its salts can cause nephropathy, and colic-like abdominal pains, and might also cause weakness in fingers, wrists, or ankles and increased blood pressure (D'souza et al., 2011; Baranowska-Bosiacka et al., 2013). Unlike adults, who absorb about 6% of ingested lead, children absorb about 50%, which means that they are at higher risk of lead poisoning than adults. Maternal exposure to Pb can lead to impaired intellectual and motor development in breast fed infants. New research has also shown an impact of lead on gene expression; for example, Pb causes increased secretion of chemokine IL-8, proinflammatory IL-6 and TNF-alpha (Gillis et al., 2012). Experimental evidence suggests that lead induces immunotoxicity (Lall & Dal, 2000). The highest coffee consumption patterns for Pb were in Bosnia and Herzegovina and ranged from 4.76-7.56 mg per capita/year. Assuming that only 64% of residents drink coffee, this gives a highest value of 7.56/0.64 = 12 mg per coffee drinker per year = 33 μg per day.

In general the lead intake varies widely according to geographical location. For example, data sourced from WHO (1995), INSERM (1999) and FAO (1989) give very high exposure rates of 83 µg per day = 30 mg/year from water, air, dust and food for adults in urban areas of the Russian Federation, Poland, Cuba, Thailand and India in the 1980s. Low urban exposure was found in Austria, Scandinavia, Netherlands, Germany, Luxemburg after 1990: 9 µg per day = 3.3 mg/year.

It can be said, therefore, that an additional 12 mg Pb per year from coffee would increase the intake considerably above these levels. Fewtrell *et al.* (2004) state that a range of adverse health outcomes can be attributed to lead, some inflicted at low concentrations (Fewtrell *et al.*, 2004). Lead was estimated to account for 0.9% of the total global disease burden, due to lead-induced mental retardation and on consequences of increased blood pressure. Coffee would therefore be expected to add to this disease burden.

However, coffee drinking would not contribute to levels of Pb which are considered to cause lead poisoning. The provisional tolerable weekly intake suggested by FAO/WHO is 214 µg per day for a 60 kg person, or 91 mg per year for a 70 kg person (http://www.hc-sc.gc.ca/fn-an/securit/chem-chim/environ/lead_plombeng.php). In the area of the Ribiera valley in Brazil the concentration of lead in water has been measured to be

as high as 230 μ g Pb/L i.e., roughly 168 mg per year (Da Cunha *et al.*, 2005). In Glasgow, Scotland, where the water was plumbosolvent lead levels were known to be often above 100 μ g/L, giving 73 mg per year assuming an adult consumption of 2 L of water per day, WHO (1995).

In conclusion, it can be said that lead in coffee can increase the intake considerably and add to the disease burden, but not to the extent as to cause lead poisoning.

CONCLUSIONS

In our study the levels of Mn, Co, Ni, Cr, and Ag were shown to be too low to influence health. However Pb, if accumulated in the body, for example by Bosnians (who are among the heaviest coffee drinkers in the world and brew coffee the Turkish way) might be harmful, but not to the extent as to cause lead poisoning.

The high level of lead in some coffees suggests that such products need more precise control of metal contamination.

Conflict of Interest

The authors report that there is no conflict of interest in the authorship and publication of this article.

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