Acta Alimentaria, Vol. 31 (3), pp. 279–288 (2002)

TRANSFER RATE OF CADMIUM, LEAD, ZINC AND IRON FROM THE TOBACCO-CUT OF THE MOST POPULAR HUNGARIAN CIGARETTE BRANDS TO THE COMBUSTION PRODUCTS

J. CSALÁRI and K. SZÁNTAI*

Department of Grain and Industrial-Plant, Faculty of Food Science, Szent István University, H-1118 Budapest, Somlói út 14-16. Hungary

(Received: 25 May 2001; accepted: 30 August 2001)

Interest is increasing in the content of toxic metals in cigarette smoke for both their harmful health effects and the possible antagonistic influence with the essential microelements. Numerous factors influence the metal concentration found in tobacco, including soil type and pH, genotype, stalk position, application of metal-containing pesticides to leaves. The rate of transfer to the smoke is dependent on the volatility, the temperature and the filter-type. In cigarette smoke element concentrations vary among brands and even within the same brand.

No comprehensive information is yet available on the toxic heavy metal content of the major Hungarian cigarette-brands. The purpose of the study was to obtain current information on the metal contents of selected brands of cigarettes being sold in Hungary. The work described in this paper had two objectives: firstly to determine the cadmium, lead, zinc and iron content of raw materials (tobacco-cut, cigarette paper, filter-rod) and secondly to measure the amount of these metals in the combustion products (cigarette smoke, ash, filter-rod).

Non-negligible part of the toxic metal content of the tobacco cut gets into the mainstream smoke, but the measured values are not higher than the similar data published in the international literature. Filters are not really efficient to decrease the toxic metal content of the mainstream smoke. The toxic metal concentration in the sidestream smoke is higher than in the mainstream smoke.

Keywords: cigarette, smoke, heavy metals, microelements, cadmium, lead, zinc, iron

In 1999 approximately 200 brands of cigarettes were present on the Hungarian market, 180-185 types of which are domestic. Cigarettes, which are manufactured in Hungary, are composed of blends representing Virginia, Burley and Oriental tobacco. In Hungary considering the yearly cigarette consumption (22–24 billion) and the number of active smokers (3.5 million people), an average smoker smokes 18 cigarettes per day.

Tobacco-plant is known to easily absorb heavy metals, especially Cd, from the soil and accumulate them in the leaves in unusually high concentration. In Hungary a comprehensive investigation has been carried out on the heavy metal content in tobacco leaf. The most commonly studied toxic metal contents such as Cd (0.50–1.89 $\mu g\ g^{-1}$),

* To whom correspondence should be addressed.
Fax: +36 1 372-6355; e-mail: kszantai@omega.kee.hu

Ni (0.33–5.40 µg g^{-1}) and Pb (0.38–1.17 µg g^{-1}) were found lower than shown in other international studies (GONDOLA & KÁDÁR, 1993). The mean level of the essential elements, such as Zn and Fe, was consistent with other foreign published data.

Part of these metals is transferred by the smoke into the human body, where they are accumulated, damage the organs (mainly kidney and liver) and act as promoters in conjunction with carcinogens. In cigarettes Cd concentrations range from 0.5 to 3.5 µg g^{-1} (CHIBA & MASIRONI, 1992). A large proportion of the Cd contained in the cigarette is passed into the smoke (MUSSALO-RAUHAMAA et al., 1986). Smoking is a source of Pb, so people who smoke or who breathe in tobacco smoke may be exposed to more Pb than people who are not exposed to cigarette smoke. Japanese cigarettes contain 1.29 µg Pb per cigarette (WATANABE et al., 1985). The Pb concentration in the tobacco-cut of Venezuelan cigarettes is much higher, it ranges from 6.48 to 12.4 µg g^{-1} (ALVARADO & CRISTIANO, 1993). MÜLLER and co-workers (2000) carried out a comprehensive investigation in order to survey the heavy metal content in tobacco-cut of the commonly smoked cigarettes purchased in Germany, China, Russia, India and Canada. The highest Cd contents were detected in Chinese brands with about five times higher concentrations than in Indian cigarettes. In case of Pb China is the "winner" again with concentrations at least three times higher than in all other countries. BELL and MULCHI (1990) obtained information on the concentrations of heavy metals in ten selected brands sold in the Unites States. The range of metal concentration found among the 10 brands (in µg g^{-1}) was 33.8 to 36.7 in case of Zn, 328 to 420 in case of Fe, 0.89 to 1.08 considering Cd and 2.11 to 2.60 considering Pb.

In the last two decades more and more details were found in international literature in connection with the element interactions. The three "toxic heavy metals", namely lead, mercury and cadmium, are in the centre of Fig. 1, while around them there are macro- and microelements, which are essential for life processes.

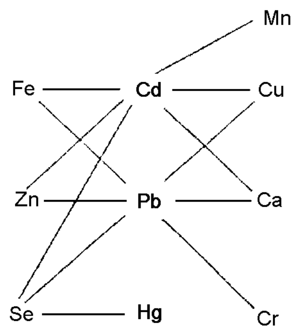

Fig. 1. Element-interactions according to CHOWDHURY & CHANDRA (1987)

The application of the essential elements can be effective in the prevention of toxicity or can help against the well-known negative physiological effects of some toxic heavy metals.

No comprehensive information is yet available about the heavy metal content of cigarettes being sold in Hungary. The object of this study was to survey the metal content (Cd, Pb, Zn, Fe) of the most popular Hungarian brands and to compare the metal content of ash, mainstream smoke (formed during puffs, inhaled by active smokers), sidestream smoke (formed between puffs, smoked by active and passive smokers, too) and filter.

1. Materials and methods

Four of the most popular brands of cigarettes (35% of Hungarian consumption) produced in Hungary were sampled. They were full flavour cigarettes with cellulose acetate filters. Before analytical measurements cigarettes were conditioned for 48 h at 22 °C and 60% humidity to achieve constant weight and moisture (HUNGARIAN STANDARD, 1994).

Thirty cigarettes from each brand were smoked by routine analytical cigarette-smoking machine (HUNGARIAN STANDARD, 1993). (Puff duration: 2 s Interpuff: 1 min Puff volume: 35 ml) The total particulate matter (TPM) of mainstream smoke was collected with ash-free paper filter (pore size: 2 µm), the gas phase was occluded by scrubbing bottles filled with 3.25 M nitric acid (50 ml). The smoke-trapping apparatus is illustrated in Fig. 2.

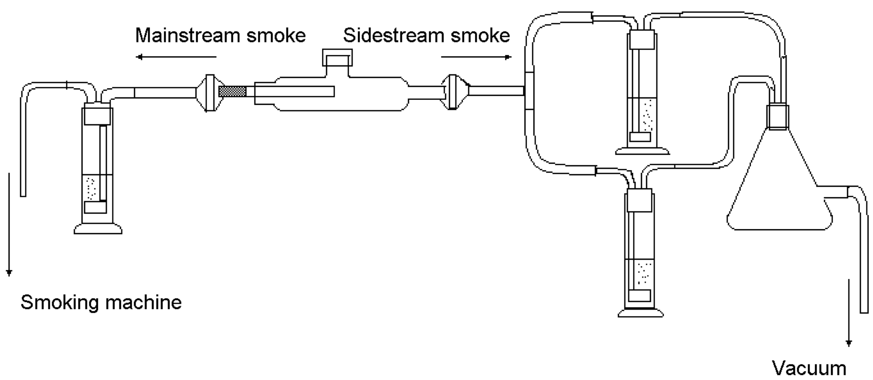

Fig. 2. Smoke trapping apparatus

Cigarettes were inserted horizontally into the sidestream smoke-chamber through the hole. The holder of Cambridge filters (a glass fiber filter pad), which collect the TPM of sidestream smoke, was positioned to the chamber. The occlusion of gas phase was similar to mainstream smoke. According to the results of our pre-experiments two scrubbing bottles were necessary to occlude the sidestream smoke, because it contains much more particulate matter and is richer in heavy metals than mainstream smoke

(SAKUMA et al., 1984). The necessary air flow – to maintain the requisite burn rate and to obtain the same number of puffs as in standard smoking – was supported by a vacuum-pump.

Tobacco-cut, cigarette-paper, filter-rod, ash and butt were digested with concentrated nitric acid and hydrogen peroxide. Destruction was carried out at 100 °C for 20 min under pressure. The occluding liquid could be analysed directly. The metal content was measured by ICP AES (ICAP-61, Thermo-Jarrel-Ash, USA).

Glassware (produced according to our own plans) was obtained from the glass-shaper of Technical University in Budapest. Chemicals were purchased from SIGMA Chemical Company (Budapest, Hungary).

2. Results

Mean metal contents in the tobacco-columns and in the cigarette-paper of each brand are given in Table 1. Iron is found in the highest concentration in tobacco followed in decreasing order by zinc, lead and cadmium. Among the different cigarette brands selected for this study, there is not a specific one that could be said to contain all the analysed elements at the highest concentration.

The levels of cadmium in the tobacco-cut vary within the range of 0.52 to 0.71 µg/cigarette, the concentrations of lead lie within the range of 1.42 to 2.21 µg/cigarette. These results are consistent with other foreign published data, however, cadmium contents are a bit lower than the average values reported in international literature (CHIBA & MASIRONI, 1992). The results of Table 1 indicate that cadmium and lead could not be detected in the cigarette-paper by the analytical method used.

Table 1. Cadmium, lead, zinc and iron content of tobacco-cut and cigarette-paper (µg/cigarette)

Cigarette-brands		Cadmium Tobacco-cut	Cadmium Cigarette-paper	Lead Tobacco-cut	Lead Cigarette-paper	Zinc Tobacco-cut	Zinc Cigarette-paper	Iron Tobacco-cut	Iron Cigarette-paper
1	Mean[a]	0.65	<DL	1.75	<DL	19.4	0.58	228	2.35
	SD	0.01		0.14		0.69	0.02	7.4	0.11
2	Mean[a]	0.65	<DL	1.42	<DL	21.5	0.43	173	5.01
	SD	0.03		0.14		1.34	0.01	9.9	0.21
3	Mean[a]	0.52	<DL	1.62	<DL	20.3	0.47	138	4.46
	SD	0.03		0.06		0.94	0.01	7.8	0.13
4	Mean[a]	0.71	<DL	2.21	<DL	19.2	0.36	180	3.74
	SD	0.03		0.16		0.78	0.02	9.8	0.24

[a] Average of 3 determinations
<DL Under detection limit

Zinc and iron could be detected both in tobacco-cut and in cigarette-paper. The zinc levels in the tobacco-column of each brand are generally similar (19.2–21.5 μg/cigarette). Some differences are noted from brand to brand in iron concentration of tobacco-cut (the range is from 138 up to 228 μg/cigarette). The zinc and iron contents of cigarette-paper are very low, they are only 1–3% of the metal concentrations of tobacco-cut. It is possible that the variability in iron content of cigarette-paper could be due to the different origins and processing ways.

Table 2 shows the element concentrations in mainstream smoke of each brand. It is evident from the values that the metals determined in the mainstream smoke of the different cigarette brands cover a wide range of concentrations. This shows that vaporization is almost completely responsible for the appearance of metals in smoke. The temperature in the burning zone of a cigarette has been determined to be in the range of 850–920 °C. Compounds have different volatility-characteristics: substances with extremely low vapour pressures at these temperatures may not be expected in higher quantity in the smoke.

Table 2. Cadmium, lead, zinc and iron content of mainstream smoke (μg/cigarette)

Cigarette-brands		Cadmium	Lead	Zinc	Iron
1	Mean[a]	0.031	0.29	0.85	8.18
	SD	0.001	0.01	0.02	0.31
2	Mean[a]	0.036	0.31	1.45	4.97
	SD	0.001	0.01	0.09	0.29
3	Mean[a]	0.034	0.26	1.31	4.87
	SD	0.001	0.01	0.02	0.13
4	Mean[a]	0.031	0.41	1.16	4.57
	SD	0.001	0.01	0.02	0.13

[a] Average of 3 determinations

Table 3. Characteristics of filter rods

Cigarette-brands	Length of filter rod	Material of filter rod	Additional material
1	20 mm	cellulose acetate	granulated activated carbon (12 mm)
2	20 mm	cellulose acetate	–
3	20 mm	cellulose acetate	granulated activated carbon (10 mm)
4	20 mm	cellulose acetate	–

Toxic metals are detected in mainstream smoke, by which these harmful elements pass into smokers' bodies and damage the organs. An ordinary smoker's body is burdened by 0.68 μg cadmium and 6.2 μg lead per day (supposing a 20-cigarette consumption per day). It supports the fact that these metals are detected in smokers' organs in higher concentration than non-smoker's and ex-smoker's. A close relation can not be experienced between the toxic metal contents of tobacco-cut and the mainstream smoke. It means that not only the harmful metal content built in tobacco leaves determines the element concentration of mainstream smoke; it can depend on the paper properties (such as porosity), the filter type and the circumstances of burning.

Table 4. Cadmium, lead, zinc and iron content captured by filter (mg/cigarette)

Cigarette-brands		Cadmium	Lead	Zinc	Iron
1	Mean[a]	0.061	0.32	1.08	8.03
	SD	0.003	0.01	0.11	0.16
2	Mean[a]	0.097	0.12	1.45	11.4
	SD	0.008	0.01	0.15	0.58
3	Mean[a]	0.072	0.26	1.56	6.3
	SD	0.009	0.01	0.02	0.58
4	Mean[a]	0.099	0.11	0.91	11.9
	SD	0.009	0.01	0.09	0.85

[a] Average of 3 determinations

The zinc content of mainstream smoke ranges from 0.85 to 1.45 μg/cigarette and the iron concentrations is from 4.57 to 8.18 μg/cigarette. Significant differences are experienced from brand to brand in zinc and iron contents of mainstream smoke. Correlation is not shown between the essential element contents of tobacco-cut and the mainstream smoke either.

Characteristics of filter rods are summarized in Table 3. The levels of metals captured by filters are found in Table 4.

Efficiency of filters to capture metals is not too good, namely these are developed to reduce the nicotine and tar content of smoke. Effectiveness is not improved significantly by additional material in case of cadmium (proved by T-test). With regard to lead content filters with activated carbon trap twice or three times more amount than filters without activated carbon. This phenomenon can be experienced inversely in case of iron; pure cellulose acetate filters trap more iron.

During smoking some part of toxic metals contaminates the environment not only by the smoke but also by the ash. The metal content of ash and sidestream smoke are given in Table 5.

After burning the considerable part of metals can be found in ash. The cadmium contents of ash range from 0.23 to 0.28 μg/cigarette; the lead levels are from 0.66 to 1.02 μg/cigarette. Non-negligible part of toxic metals pollutes the air and pass into the passive smokers' bodies. It means that only one cigarette contaminates the air with 0.17–0.33 μg cadmium and 0.31–0.66 μg lead.

Table 5. Cadmium, lead, zinc and iron content of sidestream smoke and ash (µg/cigarette)

Cigarette-brands		Cadmium		Lead		Zinc		Iron	
		Sidestream-smoke	Ash	Sidestream-smoke	Ash	Sidestream-smoke	Ash	Sidestream-smoke	Ash
1	Mean[a]	0.27	0.28	0.33	0.81	3.26	14.7	38.2	176
	SD	0.009	0.02	0.006	0.01	0.16	1.03	2.49	9.81
2	Mean[a]	0.27	0.23	0.32	0.66	2.89	16.1	33.1	129
	SD	0.008	0.01	0.01	0.01	0.02	1.24	2.35	7.31
3	Mean[a]	0.17	0.25	0.31	0.79	2.81	15.0	28.7	103
	SD	0.01	0.007	0.01	0.01	0.13	1.21	1.91	3.95
4	Mean[a]	0.33	0.24	0.66	1.02	2.23	15.3	27.6	140
	SD	0.01	0.01	0.01	0.02	0.16	1.52	1.33	3.61

[a] Average of 3 determinations

Figures 3, 4, 5 and 6 show distribution of measured metals in combustion products: mainstream smoke, trapped by filter, sidestream smoke and ash. The transfer efficiencies of the various metallic constituents present in tobacco vary over quite a wide range.

Cadmium mainly contaminates environment by sidestream smoke and ash, more than 80% of cadmium content of tobacco cut gets into the air and soil. However, 9–14% of original amount is filtrated, 4–6% of it passes into human body by mainstream smoke.

Nearly half the amount of lead gets into ash, significant part is transferred to smoke: 16–22% into mainstream smoke and 18–30% into sidestream smoke. This Figure shows, too, that filters with activated carbon are more effective to reduce lead content of smoke. Pure cellulose acetate filters adsorb 5–8% of lead content, whereas filters treated activated carbon trap 16–18% of it.

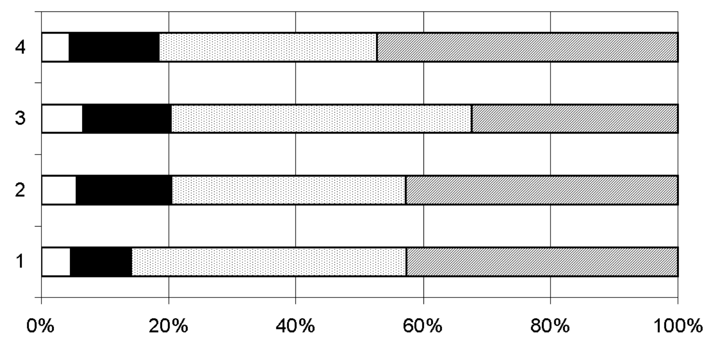

Fig. 3. Distribution of Cd in combustion-products. □: Mainstream smoke; ■: captured by filter; ▒: ash; ▨: sidestream smoke

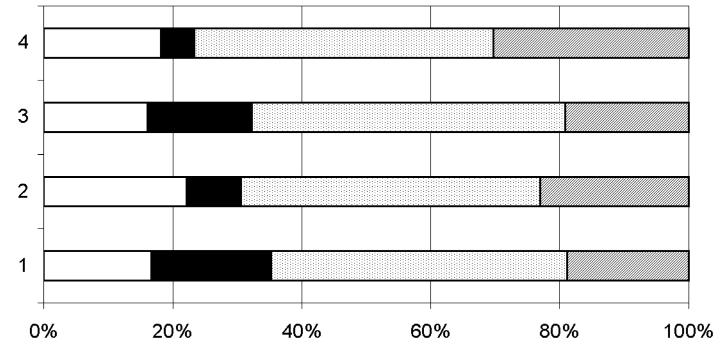

Fig. 4. Distribution of Pb in combustion-products. □: Mainstream smoke; ■: captured by filter; ▒: ash; ▨: sidestream smoke

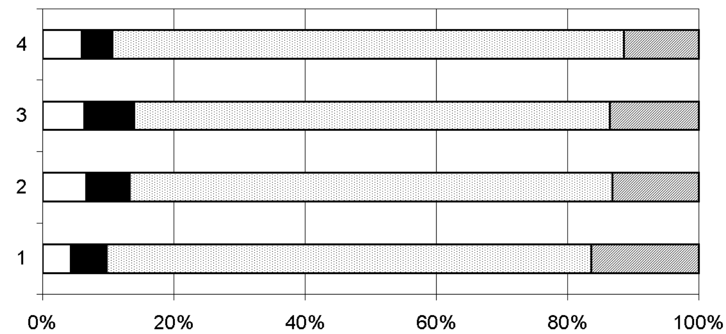

Fig. 5. Distribution of Zn in combustion-products. □: Mainstream smoke; ■: captured by filter; ▒: ash; ▨: sidestream smoke

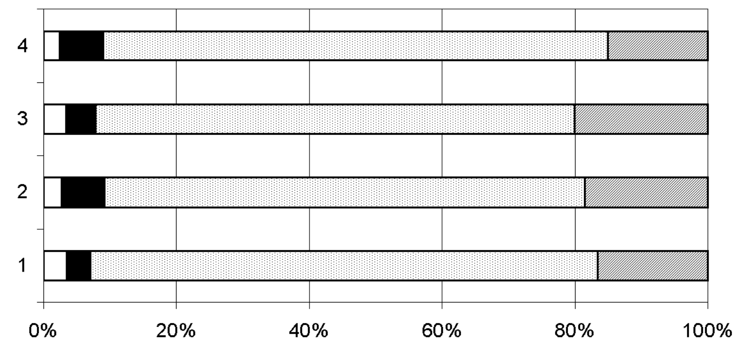

Fig. 6. Distribution of Fe in combustion-products. □: Mainstream smoke; ■: captured by filter; ▒: ash; ▨: sidestream smoke

More than 70% of essential zinc remains in ash and 10–14% gets into air by sidestream smoke. Only 4–6% passes into human body by mainstream smoke. Additional material in filters does not cause changing in efficiency of filtration.

Nearly 80% of iron content of tobacco cut gets into ash and 14–19% escapes into environment by sidestream smoke. Merely 2–3% of the original amount is transferred by mainstream smoke into smoker's body. Filters with additional material capture 3–4% of iron content, pure filters trap 6% of it.

3. Conclusion

The developed trapping apparatus is suitable for capturing simultaneously mainstream and sidestream smoke – formed under standard smoking conditions – without loss in order to analyse their metal content. It would be worth testing further filters with smaller pore size to capture the TPM of mainstream smoke. The trapping apparatus was used to analyse cadmium, lead, zinc and iron content of the smoke. We have to test and make it perfect to collect other toxic elements such as nickel.

Non-negligible quantity of toxic metals gets by mainstream smoke into smokers' bodies, where they can damage the organs and cause harmful effects (Cd: 0.03 μg/cigarette, Pb: 0.26–0.41 μg/cigarette).

Toxic metal content of sidestream smoke is high, which is worthy to note considering the passive smokers' health.

Filters are not able to remove greater part of harmful metals from smoke.

Most part of essential elements remains in ash and only a slight proportion is transferred to smoke. If more zinc and iron got into body by smoke, they would reduce the harmful health effects of cadmium and lead in active and passive smokers' organs.

References

ALVARADO, J. & CRISTIANO, A. R. (1993): Determination of cadmium, cobalt, iron, nickel and lead in Venezuelan cigarettes by electrothermal atomic absorption spectrometry. *J. Anal. At. Spectrom.*, *8*, 253–259.

BELL, P. & MULCHI, C. L. (1990): Heavy metal concentrations in cigarette blends. *Tob. Sci.*, *34*, 32–34.

CHIBA, M. & MASIRONI, R. (1992): Toxic and trace elements in tobacco and tobacco smoke. *Bull. Wld Hlth Org.*, *70*, 269–275.

CHOWDHURY, B. A. & CHANDRA, R. K. (1987): Biological and health implications of toxic heavy metals and essential trace element interactions. *Prog. Fd Nutr. Sci.*, *11*, 55–113.

GONDOLA, I. & KÁDÁR, I. (1993): Heavy metal content of flue-cured tobacco leaf in different growing regions of Hungary. *Acta agron. Hung.*, *43*, 243–251.

HUNGARIAN STANDARD (1993): *Cigaretták. Az összes és a nikotinmentes száraz részecskefázis meghatározása rutinanalitikai cigarettaelszívató gép alkalmazásával.* (Cigarettes. Determination of total and nicotin-free dry particulate matter using a routine analytical smoking machine.) MSZ 4387.

HUNGARIAN STANDARD (1994): *Dohány és dohánygyártmányok. Kondicionáló és vizsgálati légterek.* (Tobacco and tobacco products. Atmosphere for conditioning and testing.) MSZ 3402.

MUSSALO-RAUHAMAA, H., SALMELA, S. S., LEPPANEN, A. & PYYSALO, H. (1986): Cigarettes as a source of some trace and heavy metals and pesticides in man. *Archs envir. Hlth*, *41*, 49–55.

MÜLLER, G., EGGERSGLUESS, D. & NAJARDHANU RAJU, N. (2000): *Heavy metal (Cd, Cu, Pb, Cr, Zn, Hg) concentrations in tobacco of commonly smoked cigarette brands purchased in Germany, China, Russia, India and Canada*. Paper presented at the International Conference on Heavy Metals in the Environment, 6–10 August 2000, Ann Arbor, Michigan, USA.

SAKUMA, H., KUSAMA, M., YAMAGUCHI, K., MATSUKI., T. & SUGAWARA, S. (1984): The distribution of cigarette smoke components between mainstream and sidestream smoke II. *Beitr. Tabakforsch. Int.*, *12*, 199–209.

WATANABE, T., FUJITA, H., KOIZUMI, A., CHIBA, K., MIYASAKA, M. & IKEDA, M. (1985): Baseline level of blood lead concentrations among Japanese farmers. *Archs envir. Hlth*, *40*, 170–176.