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Preliminary communication

SAUCING OF TOBACCO-CUT SO AS TO INCREASE MICROELEMENT CONTENT IN THE CIGARETTE SMOKE

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Tobacco plant is known to easily absorb heavy metals from soil and accumulate them in leaves. Part of these metals is transferred by the smoke into the human body, where they accumulate, damage the organs (mainly kidney and liver) and act as promoters in conjunction with carcinogens. Application of the essential elements can be effective in the prevention of toxicity and curative manner against the negative effects of toxic heavy metals.

Reducing the harmful health effects of tobacco smoke is one of the major tasks of our research. Essential elements, first of all Zn and Fe, were added in an artificial way on tobacco-cut so that these elements should pass by the smoke to the smokers' bodies.

The toxic metal content of our tobacco-sample was consistent with other data published in international literature (Cd: 1.55 μg g^{-1}; Pb: 1.51 μg g^{-1}). The transfer rates of these metals from tobacco-cut to smoke were significant (Cd: 15–34%; Pb: 8–20%).

Twenty-thirty percent of the artificially added Zn and 10–23% of Fe transfer into the smoke was mainly in total particulate matter (TPM). The transmission of Zn to the smoke seems not to be dependent on temperature of smoke formation. However, higher Fe concentration was detected in smoke forming at 600 °C than at 900 °C. The more considerable part of Zn and Fe can be found in the ash.

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

Tobacco plants have a special ability to absorb Cd from soil and to accumulate it in unusually high concentrations in the leaves (ranging form 0.77 to 7.02 μg g^{-1}). 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 μg g^{-1}), 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).

Part of the heavy metals in tobacco leaf is transferred to the smoke. Tobacco smoking is considered to be one of the causes of heavy metals in human body. These

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metals tend to accumulate in the body and could possibly act as promoters in connection with carcinogens in cigarette smoke.

Smoking is an important source of cadmium. In cigarettes Cd concentrations range from 0.5 to 3.5 μg g^{-1} (CHIBA & MASIRONI, 1992). A large proportion of Cd contained in cigarettes passes into the smoke (MUSSALO-RAUHAMAA et al., 1986). In many studies Cd concentrations in the human body were found to rise with increased smoking. Lung Cd levels were significantly and positively correlated with total smoking time and pack-years (PAAKKO et al., 1988). Cd level in lung was found to be higher (3.0 μg g^{-1} dry weight) in smokers than in ex-smokers (1.1 μg g^{-1}) and non-smokers (0.4 μg g^{-1}). According to POCOCK and co-workers (1998) Cd concentrations increased with increased smoking. ANGERER and co-workers (1988) measured urinary Cd concentrations in non-smokers, ex-smokers and smokers. The mean Cd concentrations in kidney cortex were 10.4, 23.5 and 42.5 μg g^{-1} wet weight, respectively. According to MUSSALO-RAUHAMAA and co-workers (1986) the number of years of smoking and the number of cigarettes smoked per day were predictive of the effect of smoking on Cd content in fat tissues of smokers. Mean Cd content was four times (10.1 ng g^{-1}) higher in smokers than in non-smokers (2.5 ng g^{-1}).

Inhalation of Cd causes irritation and possibly an acute inflammatory reaction of the lungs. Long-term exposure produces chronic bronchitis and increases susceptibility to infections and emphysema. Cd causes excessive urinary loss of calcium. This is likely to decrease calcium absorption and bone mineralization and thus lead to osteoporosis and osteomalacia. Kidney damage has been observed in people who are exposed to excess Cd. The effects of Cd on the kidney take the form of renal tubular dysfunction and subsequent pathological changes (proteinuria, aminoaciduria, glucosuria).

Cigarette smoke is a source of lead, people who smoke tobacco 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 (range 0.96 to 2.00) (WATANABE et al., 1985).

Smokers and former smokers have higher blood Pb levels than non-smokers (QUINN & DELVES, 1987). Pb exposure has been shown to decrease intelligence scores (IQ), and it can cause hearing problems for infants or young children. Exposure to high level of Pb can cause bad damage in the brain and kidneys of adults and children. Pb exposure may increase blood pressure in middle-aged men. Also, a couple may have trouble with having children, if the man is exposed to Pb because high levels of Pb may affect his sperm or damage other parts of the male reproductive system. The effect of Pb to cause cancer in humans has not been shown.

In the last two decades more and more details were found in international literature in connection with the element interactions: antagonism and synergism. Figure 1 is focusing our attention on the central role of some trace elements, first of all of zinc and iron. The three "toxic heavy metals", namely Pb, Hg and Cd are in the centre, while around them are macro- and microelements, which are essential for life processes.

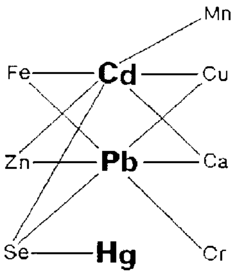

Fig. 1. Element-interactions according to CHOWDHURY & CHANDRA (1987)

Extensive literature is available regarding the protective (or antagonistic) effects of zinc on cadmium toxicity. Zn is provided to be the potent inducers of metallothionein-synthesis (HARFORD & SARKAR, 1991). Metallothionein (MT) is a small cysteine-rich protein that binds to heavy metals and its synthesis is induced by some of the metals to which it binds. BREMNER and BEATTIE (1990) were able to establish in their study a direct relation between Cd-induced MT and Zn uptake and the retention of Zn in the MT form.

The deficiency of essential elements in humans, which occurs not only in the developing countries, cannot be left out of consideration either. Evidence from a variety of experiments on animals suggests that iron deficiency also promotes lead uptake and retention. The evidence for humans is conflicting but in some studies substantial increases in lead uptake are said to have occurred when iron status were low (WATSON et al., 1980; FLANAGAN et al., 1982).

Low dietary iron also enhances the absorption and retention of cadmium by experimental animals (KOSTIAL et al., 1980). Although much less is known about such interactions in humans, it is important to bear in mind the possibility that a low iron status may well increase sensitivity to potentially toxic concentrations of such metals.

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

Laboratory tests on animals have indicated that sidestream smoke is more toxic than mainstream smoke. Cigarette smoke itself may be broken down into two categories of smoke: mainstream smoke (MS) and sidestream smoke (SS). Mainstream smoke, which is inhaled by the smokers from the cigarette during puff, forms at 900 °C. Sidestream smoke, which is emitted by the burning cigarette between puffs, forms at 600 °C.

Reducing the harmful health effects of tobacco smoke is one of the major tasks of our research. In this period of our work the purpose was to add essential elements, first of all Zn and Fe, in an artificial way on tobacco-cut so that these elements should pass by the smoke to the smokers' bodies.

1. Materials and methods

Tobacco-cut was put at our disposal by a Hungarian tobacco factory. The sample contained the main tobacco-types (Virginia, Burley, Oriental), however, the exact composition was not brought to our knowledge by the factory. This untreated European blend was considered as the control sample. Before analytical measurements the sample was conditioned for 48 h at 22 °C and 60 % humidity to achieve constant weigh and moisture (Hungarian Standard, 1994).

The tobacco-cut was sauced with Zn-salt and Fe-salt dissolved in ion-free water. The salts were required to be odourless, soluble in water, non-toxic and not to influence the flavour of the smoke. It was zinc sulfate ($ZnSO_4$) and ferrous ascorbate ($C_{12}H_{14}O_{12}Fe$), which met all requirements. Five concentrations of Zn and Fe solutions were sprinkled on the surface of the tobacco-samples, increasing the element content of tobacco-cut. The aim was to achieve 15, 30, 60, 120 and 240 mg Zn per 1 g tobacco-cut and 7.5, 15, 30, 60, and 120 mg Fe per 1 g tobacco-cut. Increasing the efficiency of casing, tobacco-samples were treated by heat placing at 60 °C for 2 h in a conditioning oven.

Twelve-twelve g of tobacco-cut was burnt in a quartz-tube (2) placed in a heater (1). The apparatus is illustrated in Fig. 2.

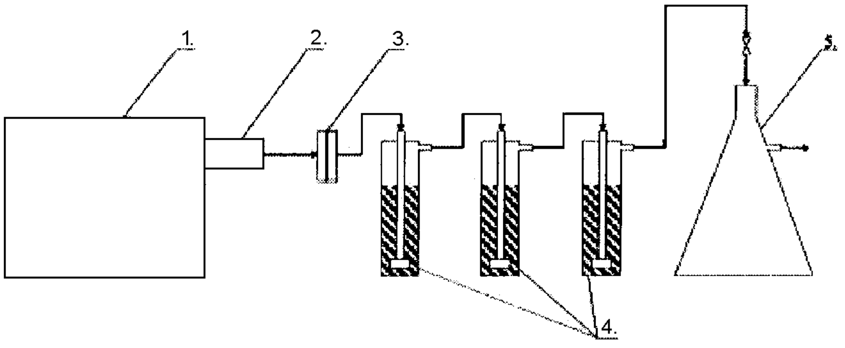

Fig. 2. Model on burning of tobacco

This apparatus was directly connected with a control unit using a NiCr-Ni thermo-element. Burning was carried out both at 600 °C (forming sidestream smoke) and at 900 °C (forming mainstream smoke). A water-jet pump drew the smoke through ash-free paper-filter pads (pore size: 2 µm) (3) and liquid traps (4) containing mixture of HCl and HNO_3 (in scrubbing bottles). This liquid, containing gas phase, could be analysed directly.

Before analysis tobacco-cut was digested with concentrated nitric acid and hydrogen peroxide. Destruction was carried out at 100 °C for 20 min under pressure. Filter pads, collecting total particulate matter, were digested in the same way as tobacco-cut. 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 the Budapest University of Technology and Economics. Chemicals were purchased from SIGMA Chemical Company (Budapest, Hungary).

2. Results

Results obtained from the analysis of control tobacco-cut and smokes forming at 600 °C and at 900 °C are presented in Table 1. The mean levels of these elements were consistent with other published data in the international literature (TSO, 1990).

Table 1. Toxic and essential elements concentrations in tobacco-cut and smoke (µg g^{-1})

	Tobacco-cut		Smoke forming at 600 °C		Smoke forming at 900 °C	
	Mean[a]	SD	Mean[a]	SD	Mean[a]	SD
Cd	1.55	0.31	0.54	0.15	0.23	0.06
Pb	1.51	0.31	0.31	0.06	0.12	0.02
Zn	37.1	2.81	4.60	0.47	3.77	0.53
Fe	413	42.3	46.0	2.04	28.7	1.52

[a] Average of 3 determinations

The transfer rate of Cd from tobacco-cut to smoke was significant (15–34%). However, in case of Pb these values were lower (8–20%) but considering the average cigarette consumption (20 cigarettes per day) it means that human body is exposed to Pb significantly "thanks to" smoking. The transfer rate of Cd and Pb was lower in smoke forming at 900 °C than at 600 °C.

Table 2 shows the measured Zn and Fe content in tobacco-samples sauced by different concentrations of Zn- and Fe-solutions.

We managed to get the 86–91% of the aimed amounts of Zn and 80–89% of Fe diffused into tobacco-tissues. These values are slightly lower than the calculated values.

Tables 3 and 4 show the transmission of Zn and Fe into the products of combustion, ash and smoke, which is divided into two parts: gas phase and total particulate matter (TPM).

Table 2. The aimed and the achieved Zn and Fe concentration in tobacco-cut (mg g^{-1})

Zn			Fe		
The aimed concentration	The achieved concentration		The aimed concentration	The achieved concentration	
	Mean[a]	SD		Mean[a]	SD
15	13.2	1.90	7.5	6.53	0.48
30	26.8	2.52	15	13.3	1.79
60	55.0	4.14	30	26.7	1.87
120	106	12.2	60	52.5	4.72
240	208	15.7	120	96.9	7.51

[a] Average of 3 determinations

Table 3. The Zn content of TPM, gas phase and ash (μg g^{-1})

		15 mg g^{-1}		30 mg g^{-1}		60 mg g^{-1}		120 mg g^{-1}		240 mg g^{-1}	
		Mean[a]	SD	Mean[a]	SD	Mean[a]	SD	Mean[a]	SD	Mean[a]	SD
600 °C	TPM	2830	62.3	5700	71.9	12310	370	26080	630	61210	550
	Gas phase	114	1.12	224	4.33	505	4.90	876	4.29	1890	11.5
	Ash	10260	144	20890	157	42220	666	78620	903	145130	1406
900 °C	TPM	2840	53.1	6140	78.5	12750	281	25860	410	60400	915
	Gas phase	111	2.23	220	4.13	486	3.47	795	8.20	1770	11.2
	Ash	10260	143	20450	118	41800	841	78920	867	146050	1922

[a] Average of 3 determinations

Table 4. The Fe content of TPM, gas phase and ash (μg g^{-1})

		7.5 mg g^{-1}		15 mg g^{-1}		30 mg g^{-1}		60 mg g^{-1}		120 mg g^{-1}	
		Mean[a]	SD	Mean[a]	SD	Mean[a]	SD	Mean[a]	SD	Mean[a]	SD
600 °C	TPM	1490	14.8	2740	25.2	5920	66.4	10680	165	20790	307
	Gas phase	15.7	0.32	27.9	0.85	48.7	0.65	152	2.41	301	7.12
	Ash	5030	52.0	10510	157	20760	246	41620	540	75790	817
900 °C	TPM	700	7.18	1940	15.5	2660	35.8	6910	86.5	11030	192
	Gas phase	16.1	0.14	35.2	0.30	82.0	0.65	191	1.78	352	7.47
	Ash	5830	38.7	11290	110	23990	349	45350	473	85500	801

[a] Average of 3 determinations

A more considerable part of these metals can be found in the ash both at 600 °C and 900 °C. Comparing the control sample with cased samples our goal was reached, namely that the Zn and Fe amounts in the smoke were increased significantly.

For example, in the 5th sample the Zn concentration was modified from 3–4 μg g^{-1} to 62–63 mg g^{-1}, in case of Fe these values were changed from 28–45 μg g^{-1} to 11–21 mg g^{-1}. In gas phase the metal content could hardly be detected, the metal content is concentrated in the TPM.

The distributions of Zn and Fe in ash, gas phase and TPM of smoke were expressed in percentage as illustrated in Figs 3, 4 and 5, 6.

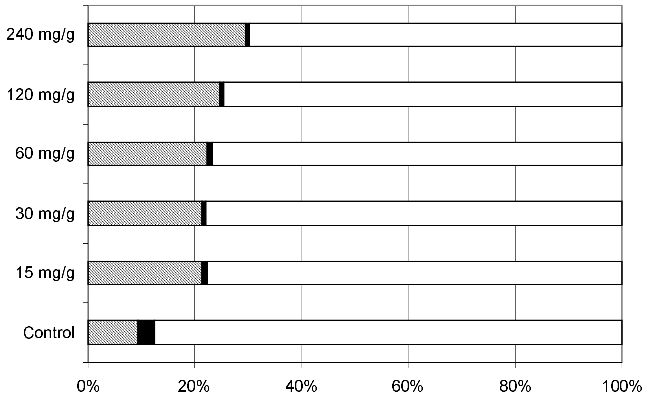

Fig. 3. Distribution of Zn content at 600 °C. ▨: TPM; ■: Gas-phase; ☐: Ash

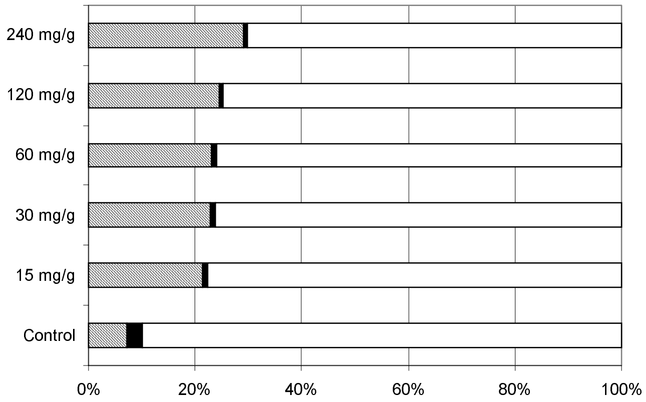

Fig. 4. Distribution of Zn content at 900 °C. ▨: TPM; ■: Gas-phase; ☐: Ash

Considering samples sauced by Zn-solution slight differences were shown between the metal content of smoke forming at 600 °C and at 900 °C. So the transmission of Zn to the smoke seems not to be dependent on temperature. In case of samples sauced by Fe-solution, 10–14% of the Fe content of tobacco-cut transferred to the smoke forming at 900 °C and 20–23% of this concentration passed to the smoke forming at 600 °C.

Ten-twelve % of natural Zn content, which was built in by the tobacco plant, passed to the smoke, the considerable part of which could be detected in TPM.

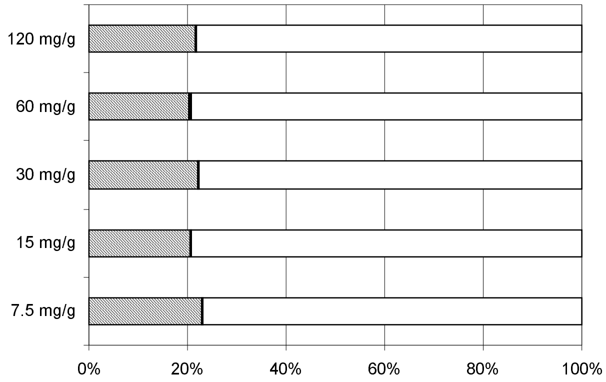

Fig. 5. Distribution of Fe content at 600 °C. ▨: TPM; ■: Gas-phase; □: Ash

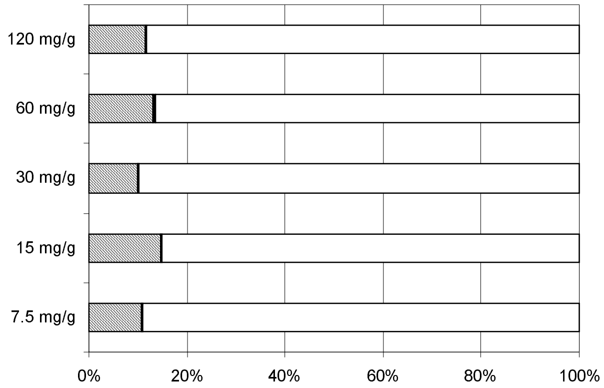

Fig. 6. Distribution of Fe content at 900 °C. ▨: TPM; ■: Gas-phase; □: Ash

However, 22–30% of the artificially added Zn was present in the smoke. An explanation could be that the Zn was not present in the same form: tobacco plant builds in Zn mainly as phosphate-salt, whereas we cased tobacco-cut by $ZnSO_4$. The volatilities of these salts are different. Similar tendency was observed in case of Fe. Transmission of Fe from control sample to smoke was 6–11%, whereas these values of cased samples were significantly higher (10–23%).

Comparing the control samples with the sauced ones the ratio of the Zn contents in TPM and that of gas phase were shifted powerfully. Considering the control sample the Zn concentration in TPM was 7–9%, in gas phase 2.8–3.1%; on the other hand in the cased samples it was 21–29% and 0.8–0.9%, respectively. Transmission of Fe to the TPM and gas phase was analogous to Zn, namely the 4–9% of the Fe content of tobacco-cut was detected into the TPM of the control sample and 1–2% of it in the gas phase; in case of sauced samples these values were 9–22% and 0.1–0.3%, respectively.

Increasing the metal concentration of casing-solution, changes could not be experienced in percentage of Zn and Fe getting over to smoke.

3. Conclusions

Non-negligible part of Cd and Pb contents pass from the tobacco-cut to the smoke during burning.

We managed to increase the Zn and Fe contents of cigarette smoke by saucing. However, significant differences between the Zn and Fe concentrations of smoke from control sample and cased samples are unambiguous, using high significance levels (0.001) these differences were confirmed by statistical method (T-test).

The metal content of smoke is concentrated in TPM. The transmission of Zn to the smoke seems not to depend on temperature, whereas the Fe content of smoke forming at 600 °C is greater, than smoke forming at 900 °C. More significant ratio of metals added in an artificial way to the tobacco pass to the smoke than from the natural metal content of tobacco.

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