

# DETERMINATION OF LEAD AND ARSENIC IN TOBACCO AND CIGARETTES: AN IMPORTANT ISSUE OF PUBLIC HEALTH

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## SUMMARY

Contents of lead and arsenic were determined in 617 tobacco samples and 80 samples of cigarettes. The mean content of lead in tobacco was 0.93 µg/g (range 0.02–8.56 µg/g) and arsenic was 0.15 µg/g (range <0.02–2.04 µg/g). The mean content of lead in cigarettes was 1.26 µg/g (range 0.02–6.72 µg/g) and arsenic was 0.11 µg/g (range <0.02–0.71 µg/g). There was a large variability in lead and arsenic content among samples of tobacco and samples of cigarettes. Positive correlation between lead and arsenic contents in tobacco was found ( $r=0.22$ ;  $p<0.0001$ ).

Based on our data and data from literature we compare the content of lead and arsenic in tobacco and cigarettes in other studies and discuss the influence of smoking to lead and arsenic exposure and health.

In conclusion, at the same time with the implementation of tobacco use prevention programmes it is advisable to implement continuous monitoring of lead and arsenic in tobacco and cigarettes in order to reduce the health risk due to exposure of these metals.

*Key words:* tobacco, cigarette, lead, arsenic

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## INTRODUCTION

According to the data of WHO – in the 20th century, 100 million people have died from tobacco use (1). At the same time, studies were focused on quantitative and qualitative analyses of tobacco constituents potentially responsible for the negative health effects.

More than 4,000 chemicals have been isolated from tobacco (hydrocarbons, aldehydes, ketones, aromatic hydrocarbons, heavy metals including lead and arsenic). Lead and arsenic have been identified and measured both in tobacco and tobacco smoke (2–6).

It is observed that contents of lead (7–9) and arsenic (10–12) in biological samples of human population are much higher in smokers than those in non-smokers.

Lead and arsenic are classified as carcinogenic to humans or possibly carcinogenic to humans (group 1 or 2) (13), but also various other negative effects of lead and arsenic on human health have been recognized (14).

Human population is exposed to lead and arsenic from many sources (air, water, soils, foodstuffs, and anthropogenic sources) (6). Smoking is not the main source of lead and arsenic exposure for humans, but cigarette smoking influences lead and arsenic toxicity.

Arsenic is methylated and eliminated from the body by urine. Cigarette smoking lowers methylation capacity of arsenic (15) and elimination of arsenic from the body. Cigarette smoking also can act synergistically with arsenic exposure to cause DNA damage in lungs (16). Mortality risk from heart disease, skin lesion, bladder cancer, and lung cancer associated with exposure to arsenic is higher among smokers (17–20).

Lead circulates in the bloodstream and accumulates in tissues and bones, or is eliminated from the body, primarily in urine. The blood brain barrier of children and infants is relatively impermeable to lead but they are at high risk of accumulating lead in the brain and central nervous system which may cause neurodegeneration (21). Tobacco is an important source of lead in secondhand tobacco smokers (children and adolescents) in the United States. Blood levels of lead were 14% and 24% higher in children who lived with 1 or with 2 or more smokers than in children living with non-smokers (22).

Lead plays a significant role in tobacco toxicity, especially radioactive <sup>210</sup>Pb. <sup>210</sup>Pb is a product of <sup>238</sup>U disintegration and its existence in tobacco depends on the tobacco origin and natural level of uranium in the soil where tobacco grows. In Italy, <sup>210</sup>Pb dose from inhalation of cigarette smoke is much higher than the dose from ambient air (23).

Exposure to metals through tobacco depends on the amount of metal present in tobacco, a percentage that is transferred to the tobacco smoke and the percentage that is absorbed.

The aim of this paper is to present content of lead and arsenic in tobacco and cigarettes analysed in the Institute for Public Health Niš (Serbia), and data from studies around the world available.

## MATERIALS AND METHODS

In the period from 2008–2010, the accredited laboratory (ISO 17025) in the Public Health Institute Niš, determined the content of lead and arsenic in 617 samples of imported tobacco and 60 samples

of imported cigarettes. The origins of analysed tobacco samples (Virginia, Burley, and Oriental tobacco types) was from different production areas around the world (Europe, Asia, America, and Africa). For the purposes of this investigation, 20 samples of different brands of cigarettes were purchased from the Niš (Serbia) market. Brand names are not discussed in this paper due to legal restrictions.

Usually, cigarette is made up of tobacco, paper and additives. For this reason, 19 samples of aroma and 5 samples of paper also were analysed by standard methods.

Samples were prepared before measurement by Electrothermal Atomic Absorption Spectrometry, instrument: Perkin Elmer AnAnalyst 600, with transversely-heated graphite atomizer (THGA) system, with Zeeman background correction.

10 g homogenized sample and 10 ml of magnesium nitrate solution (10% w/v in 95% ethanol) was mixed well in 100 ml beaker and evaporated on a steam bath and completely dried in oven for 1hr at 150°C. Samples are heated on a hot plate (200°C), gradually increasing temperature until organic matter was thoroughly charred (not exceeding 450°C). Beakers were placed in a muffle furnace and turned into ash overnight at 450°C. In removed and cooled beakers, a few drops of HNO<sub>3</sub> were added, dried on a hot plate, returned to muffle furnace for 1h until they turned into white ash, repeating the nitric acid treatment if necessary. Beaker was cooled and 10 ml of extraction acid were added carefully (200 ml conc. HCl + 650 ml H<sub>2</sub>O + 150 ml conc. HNO<sub>3</sub>) to dissolve ash (heated if necessary) (24).

Used beakers were soaked in 10% HNO<sub>3</sub> for 24 h; washed with de-ionized water and dried in order to avoid contamination.

All used chemicals were of high purity required for the trace metals analysis: extra pure nitric acid (69% HNO<sub>3</sub>), hydrochloric acid (36% HCl) (Merck, Darmstadt, Germany), and modifiers (NH<sub>2</sub>H<sub>2</sub>PO<sub>4</sub>, Pd(NO<sub>3</sub>)<sub>2</sub> and Mg(NO<sub>3</sub>)<sub>2</sub>, Merck, Darmstadt, Germany).

Samples were routinely analysed in laboratory, in duplicate and as spiked samples as QA/QC measures. The intralaboratory precision and accuracy of the results is controlled for every run starting with a control blank and quality control samples (duplicate and spiked).

This procedure was repeated in series after every fifth or tenth sample. Results vary for lead within 6% and 12% for arsenic (CV %). The results are interpreted according to current legal Book of regulation (24).

Descriptive statistics (mean, standard deviation, median), coefficient of variation (CV %), and Pearson's coefficient of correlation (r) were processed by Microsoft Excel software.

## RESULTS

Metal concentrations found in tobacco and cigarettes are shown in Table 1 (for lead) and Table 2 (for arsenic).

The Book of the Regulations stipulates maximum permitted levels (MPL) of lead (10 mg/kg) and arsenic (3 mg/kg) in tobacco and cigarettes. In all analysed samples of tobacco and cigarettes the concentration of lead and arsenic was within legal limits (Table 1 and 2).

For lead and arsenic in tobacco and cigarette, results showed the high coefficient of variation (CV %).

Table 3 showed significant positive correlation (p<0.0001) between lead and arsenic content in tobacco but not in cigarettes.

Cigarettes contain tobacco, paper, and additives. Many additives are used in cigarette manufacture as well as paper. They contain heavy metals including lead and arsenic (Table 4 and 5).

Content of lead and arsenic in paper and tobacco was lower than in tobacco and cigarettes.

## DISCUSSION

Metal content in tobacco depends on soil properties, atmospheric conditions, and requirements for tobacco farming (use of pesticide and fertilizer). Tobacco plants take up lead and arsenic from soil and concentrate these metals in leaves. For this reason, there are large variations in the content of metals in tobacco between countries.

In two studies content of lead and arsenic in tobacco and cigarettes from around the world was analysed. Watanabe et al. (25) analysed lead contents in 331 samples of cigarettes from various areas of the world. The mean content of lead was 1.76 µg/g (range 0.46–43.66 µg/g), and was higher than the lead content in our study (0.93 µg/g). Lugon-Moulin et al. (26) analysed 1,431 leaf samples of tobacco from Africa, Europe, South and North America. Content of arsenic (0.4 µg/g) was higher than arsenic content in tobacco found out in our study (0.15 µg/g).

**Table 1.** Content (µg/g) of lead in tobacco and cigarettes

	Number of samples (n)	Mean	Standard deviation	CV (%)	Median	Min-max	>MPL
Tobacco	617	0.93	1.1	118.3	0.58	0.02–8.56	–
Cigarette	80	1.26	1.7	134.9	0.53	0.02–6.72	–

**Table 2.** Content (µg/g) of arsenic in tobacco and cigarettes

	Number of samples (n)	Mean	Standard deviation	CV (%)	Median	Min-max	>MPL
Tobacco	617	0.15	0.19	126.7	0.11	<0.02–2.04	–
Cigarette	80	0.11	0.09	81.8	0.10	<0.02–0.71	–

**Table 3.** Correlations between lead and arsenic content in analysed samples of tobacco and cigarettes

	Coefficient of correlation (r)	p
Tobacco	0.220	p<0.0001
Cigarettes	-0.06	p>0.01

The results of present study showed lower content of lead and arsenic in tobacco and cigarettes than most other studies did and these were within legal limits. There was a large variability in lead and arsenic content among samples because the origin of tobacco and its variety influenced the metal content.

The mean levels of lead determined in cigarettes in our study were lower than those determined in cigarettes from markets in China (27, 28), Nigeria (29), Jordan (30), Pakistan (31), Russia (28), and similar to Korean domestic cigarettes (32), and ciga-

rettes commonly smoked in India, Germany, and Canada (28). Our results for lead content in cigarettes were higher than those observed in Poland (7), Brazil (33), and cigarettes imported to Korea from UK (32) (Table 6).

Compared to the values described in the literature for arsenic in cigarettes (Table 7), the mean levels of arsenic in the present study were lower than those reported in the studies in China (27), and from Korean domestic cigarettes (32), and similar to cigarettes sold in UK (32). Only in one study from Brazil (33) mean content of arsenic was lower compared to that reported in our study (Table 7).

A significant positive correlation was observed between lead and arsenic content in tobacco but not in cigarettes. These results suggest the possibility that these metals in tobacco originated from the same source.

Contents of lead and arsenic in paper and aroma of cigarettes are much lower than in tobacco and do not influence the contents of those metals in cigarettes.

**Table 4.** Content ( $\mu\text{g/g}$ ) of lead in additives (aroma) and paper for cigarette

	Number of samples (n)	Mean	Standard deviation	CV (%)	Median	Min-max
Aroma	19	0.06	0.04	66.7	0.07	0.01-0.13
Paper	5	0.38	0.14	36.8	0.36	0.2-0.55

**Table 5.** Content ( $\mu\text{g/g}$ ) of arsenic in additives (aroma) and paper for cigarette

	Number of samples (n)	Mean	Standard deviation	CV (%)	Median	Min-max
Aroma	19	0.023	0.01	4.3	0.02	<0.02-0.05
Paper	5	0.05	0.03	60	0.05	<0.02-0.08

**Table 6.** Comparison between the present result and available reported content of lead in cigarettes

Source of cigarettes	Mean ( $\mu\text{g/g}$ )	Range ( $\mu\text{g/g}$ )	Reference
Cigarettes sold in China	2.65	1.2-6.5	O'Connors et al. (27)
Cigarettes sold in Nigeria	10.8	-	Yebpella et al. (29)
Cigarette brands sold and/or produced in Jordan	2.67	2.10-3.23	Massadeh et al. (30)
Domestic cigarettes in Pakistan	14.39	10.16-27.33	Ajab et al. (31)
Imported cigarettes in Pakistan	8.749	2.5-14.0	
Cigarette brands sold in Korea	1.35	0.88-2.13	Jung et al. (32)
Cigarette brands sold in UK	0.74	0.45-1.35	
Cigarettes commonly smoked in:			Müller et al. (28)
China	4.48	-	
Canada	1.45	-	
India	1.57	-	
Germany	1.62	-	
Russia	1.86	-	
Cigarettes produced in Poland	0.7667	-	Galazyn-Sidorczuk et al. (7)
Cigarettes commercialized in Brazil	0.27	0.19-0.39	Viana et al. (33)
Our study	1.26	0.02-6.72	

**Table 7.** Comparison between the present result and available reported content of arsenic in cigarettes

Source of cigarettes	Mean (µg/g)	Range (µg/g)	Reference
Cigarettes sold in China	0.82	0.3–3.33	O'Connors et al. (27)
Cigarettes commercialized in Brazil	0.09	0.05–0.13	Viana et al. (33)
Cigarette brands sold in Korea	0.23	0.17–0.31	Jung et al. (32)
Cigarette brands sold in UK	0.12	0.07–0.18	
Our study	0.11	<0.02–0.71	

To reduce the risk of human population exposure to lead and arsenic, continuous monitoring of these two metals in tobacco is required as in other countries (34, 35) simultaneously with the implementation of tobacco control programmes in Serbia.

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