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Heavy metals in cigarettes for sale in Spain

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ABSTRACT

The aim of the present study was to determine the concentrations of eight metals (Al, Cd, Co, Cr, Mn, Ni, Pb, Sr) in 33 cigarette brands for sale in Spain. Samples were analysed by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES). Mean values obtained were 428 µg Al/g, 0.810 µg Cd/g, 0.558 µg Co/g, 1.442 µg Cr/g, 112.026 µg Mn/g, 2.238 µg Ni/g, 0.602 µg Pb/g and 82.206 µg Sr/g. Statistically significant differences were observed with respect to concentrations of Co (0.004), Cr (0.045), Mn (0.005) and Sr (0.005) between black and blond tobacco and between levels of Mn (0.027) among manufacturers. Considering a Cd inhalation rate of 10% and a Cd absorption rate of 50%, absorption of Cd for smokers of 30 cigarettes/day was estimated at 0.75 µg Cd/day. An inhalation rate of 2–6% and an absorption rate of 86% were considered for Pb, Pb absorption in smokers of 30 cigarettes/day was therefore 0.18–0.54 µg/day. In view of the significant toxic effects of these metals, quantification and control of their concentrations in this drug are of the utmost importance.

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1. Introduction

Tobacco (*Nicotiana tabacum*) is highly addictive due to its nicotine alkaloid content (Tuesta et al., 2011; Nnorom et al., 2005; Pappas et al., 2007). It is estimated that 22% of the global population over the age of 15, around 250 million women and 1 billion men (Ajab et al., 2008), are smokers (WHO, 2012). The tobacco habit is associated with a death every 6 s (Verma et al., 2010; WHO, 2012) and is responsible for approximately 5 million deaths each year, i.e., 1 out of every 10 deaths in the world (Verma et al., 2010), and is expected to cause 10 million deaths in 2025 (Ajab et al., 2008) due to an increase in the number of smokers in developing countries (de Sousa Viana et al., 2011). China, where 57% of the male population and 3% of females are smokers, is the leading country (O'Connor et al., 2009). In Spain, 29.6% of the population smoke regularly and 68.5% have consumed tobacco at some time (EDADES, 2007). In the Canary Archipelago 26.4% of its inhabitants are smokers and 62.0% have consumed tobacco occasionally (Gobierno de Canarias, 2008). The number of smokers continues to decrease in developed countries due to effective campaigns focused on reducing tobacco consumption (Ashraf, 2012), such as Law 42/2010 (BOE, 2010) of the Spanish Government.

All tobacco products pose risks to health. The harmful effects of tobacco have been known for over 50 years (US Surgeon General's report, 2010), due to its content in both organic and inorganic toxic, genotoxic, mutagenic and carcinogenic components (Ashraf,

2012; Massadeh et al., 2005; Pappas et al., 2007; Verma et al., 2010). Tobacco processing uses between 600 and 1400 additives (Kazi et al., 2009; Nnorom et al., 2005; Pappas et al., 2007). Around 4700 compounds, many of which are toxic, such as aromatic hydrocarbons, aldehydes, ketones, and heavy metals are found in tobacco (Ajab et al., 2008; de Sousa Viana and Menezes-Filho, 2011). Inhalation of the product, either directly (smoking) or indirectly (passive smoking), is related to multiple pathologies including most notably diverse types of cancer (larynx, oesophagus, trachea, lungs, stomach, pancreas, kidneys, ureter, cervix, bladder) in addition to chronic diseases (cerebral infarction, blindness, cataracts, periodontitis, aortic aneurysm, coronary disease, pneumonia, asthma, COPD, atherosclerosis) (US Surgeon General's report, 2010; de Sousa Viana et al., 2011; Galazyn-Sidorczuk et al., 2008; Kazi et al., 2009; Nnorom et al., 2005). The high exposure of toxic metals as a result of cigarette smoking may be associated with diabetic mellitus and rheumatoid arthritis (Afridi et al., 2014; Afridi, 2015b, 2015c).

The most noteworthy of these diverse toxic components are heavy metals (Galazyn-Sidorczuk et al., 2008; Massadeh et al., 2005; Nnorom et al., 2005). Some of these metals are essential at very low concentrations (Verma et al., 2010), but others are toxic at very low concentrations (Nada et al., 1999; Rubio et al., 2012). The presence of metals in the *Nicotiana tabacum* plant is a result of its ability to absorb them from the soil where it grows (de Sousa Viana et al., 2011; Galazyn-Sidorczuk et al., 2008; O'Connor et al., 2010; Rodríguez-Ortiz et al., 2006), in addition to various other

factors, such as climatic conditions, plant variety, use of pesticides and fertilisers, soil pH, and manufacturing process, among others (de Sousa Viana et al., 2011; Järup, 2003; Kazi et al., 2009; Nada et al., 1999; Verma et al., 2010). Tobacco farming is an economic activity in more than 30 countries, including Argentina, Brazil, China, Greece, Italy, Malawi, Mozambique, Tanzania, Spain, Turkey and the United States. The *Nicotiana tabacum* plant has a preference for absorbing metals such as Pb, Cd and Zn, although it absorbs much more Cd than Pb due to the greater mobility of the latter, and preferably accumulates them in its leaves (Kazi et al., 2009; Sand y Becker, 2012; Satarug and Moore, 2012).

The IARC has classified Cd as a group 1 carcinogen and Pb has recently been raised from category 2B to 2A (Ashraf, 2012; de Sousa Viana et al., 2011; IARC, 2012; Järup, 2003; Pappas et al., 2007). Cd is in seventh place on the list of the 275 most hazardous materials, and is considered to be one of the ten most problematic chemicals for public health (Satarug, 2012). Several studies suggest that the carcinogenic ability of the afore-mentioned tobacco metals is comparable to that of N-nitrosamines and polycyclic aromatic hydrocarbons (Fowles and Dybing., 2003).

Concentrations of Cd in biological fluids of smokers are greater than in those of non-smokers (de Sousa Viana et al., 2011; Järup, 2003) as well as in tissues (Elinder et al., 1983), although the same pattern is not observed in the case of Pb, the latter providing controversial data its concentrations are compared in smokers and non-smokers (Galazyn-Sidorczuk et al., 2008). The levels of Cd and Pb are much greater in the pulmonary tissue of a smoker than in that of a non-smoker (O'Connor et al., 2010). Likewise, an association has been observed between blood Pb levels and tobacco and alcohol consumption (Massadeh et al., 2005; Ashraf, 2012).

It has been estimated that tobacco smokers are exposed to 1.7 µg Cd per cigarette, and about 10% is inhaled when one cigarette is smoked (Morrow, 2001; NTP, 2005). While the fatty tissue of smokers presents a four-fold concentration of Cd with respect to a non-smoker, (Galazyn-Sidorczuk et al., 2008), the blood Cd levels are up to 29% higher, and an analysis of female oral cancer patients reveals a significantly higher concentration of Cd in blood and hair (de Sousa Viana et al., 2011). The mean values of Cd in venous blood of smokers (118 ng/mL) are greater than those of non-smokers (0.46 ng/mL) (Rubio et al., 2006). In regard to the Cd concentration in liver and kidney tissues, the values in smokers are twice those of non-smokers (Scherer and Barkemeyer, 1983). The levels of Cd in human breast milk are strongly related to whether or not the mother is a smoker and if she is exposed to cigarette smoke (Nnorom et al., 2005). Increased toxic elements and decreased essential elements as a result of cigarette smoking are associated with the development and pathogenesis diabetes mellitus. The high exposure to toxic metals as a result of cigarette smoking have been considered to be synergistic with risk factors associated with diabetic mellitus (Afridi et al., 2015a; Afridi et al., 2015b).

A harmful plasma concentration of Pb is characterised in the case of adults by haemotoxic effects, reproductive failure, nephropathies, lower intellectual coefficient, behavioural problems (Ashraf, 2012; Massadeh et al., 2005; Kazi et al., 2009) and Alzheimer's disease (Rubio et al., 2005). Low to moderate levels of Pb induce an increase in IgE, which favours the development of asthma (Lutz et al., 2012).

With regard to Cd, foodstuffs constitute the main source of exposure for non-smokers (Massadeh et al., 2005; Sand and Becker, 2012). However, the main source of exposure to this metal in both active and passive smokers is tobacco (Ashraf, 2012; Ajab et al., 2008; Galazyn-Sidorczuk et al., 2008; Järup, 2003; Nnorom et al., 2005; Satarug, 2012). Cd has the ability to bioaccumulate, since its mean half-life is from 10 to 30 years (Sand and Becker, 2012; Satarug and Moore, 2012) causing particular damage to the

kidneys, bones and liver, and causing pathologies such as irritation of the stomach lining (Ashraf, 2012; Massadeh et al., 2005; Satarug, 2012). Damage caused to renal tubules is irreversible (Järup, 2003). A positive relationship exists in regard to exposure to Cd and the incidence and severity of diabetes, since Cd reduces insulin levels and has direct cytotoxic effects on the pancreas; it also enhances and exacerbates diabetic nephropathy (Edwards and Prozialeck, 2009).

Al, a metal with no function in the human body, is known for its neurotoxic nature and its effects on bone tissue (Domingo, 1995; González-Weller et al., 2010). Mn, Ni and Co are essential elements, since they form part of the metalloenzymes and metalloproteins of the human body, as do other metals (Rubio et al., 2012; Verma et al., 2010). However, an increase in the concentrations of Mn and Co implies toxic effects, predominantly on the central nervous system (Ajab et al., 2008), while high doses of Ni are associated with carcinogenicity (Kazi et al., 2009).

It is noteworthy that legislation exists to regulate the metal content of foodstuffs (Rubio et al., 2012), whereas it is absent in regard to tobacco (O'Connor et al., 2010).

Scientific literature focusing on heavy metals in tobacco does exist (Nada et al., 1999), but changes in cultivation techniques and product manufacturing (Verma et al., 2010) justify this research line.

In view of the above, the aim of the present study was to determine the mean concentrations of eight significant metals (Al, Cd, Co, Cr, Mn, Ni, Pb and Sr) in light, normal, Virginia and dark tobacco cigarettes.

2. Materials and methods

2.1. Samples

A total of 33 samples of cigarettes marketed in Spain were analysed. Each sample of 1.5 g of cigarette tobacco was taken from 3 cigarettes and this was repeated three times per brand using three different packets (3 cigarettes per packet 3 packets =9 cigarettes per brand were used). The average measurement of the three measurements was assigned to each brand. Samples were classified as light (10 samples) and normal (23 samples). A cigarette was considered to be light when the nicotine concentration was ≤ 0.6 mg. Samples were classified as Virginia (30 brands) and Dark (3 brands), also in agreement with the manufacturer: Philip Morris (8 brands), Imperial Tobacco (4 brands), British American Tobacco (3 brands) and Japan International Tobacco (18 brands) (Table 1).

2.2. Preparation of the samples and analytical procedure

Samples of 1.5 g of cigarette tobacco were weighed after removing the filter and paper. The mean weight of each cigarette was obtained by weighing its content (without filter or paper) three times using a PB153-S/FACT balance (Mettler Toledo, Switzerland).

Samples were placed in a porcelain crucible previously sterilized in a muffle oven (Nabertherm[®], Germany) at 800 °C for 24 h. Approximately 10 drops of 65% HNO₃ (Emsure[®], Merck, Germany) were added, the samples were ground with a glass rod and placed on a hotplate (P Selecta, Spain) to evaporate the acid, using a laminar flow hood (Romero[®] Flowtronic[®] F-3M, Spain).

The samples were then placed in a muffle oven (Nabertherm[®], Germany) and the temperature was raised to 450 °C for 48 h, to achieve incineration of the organic matter of the samples. Once incinerated, 4 drops of 65% HNO₃ were added and evaporated by placing the samples on the hotplate, followed by further

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