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Impact of tobacco smoke on the elemental composition of indoor different particles sizes

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Abstract

Environmental tobacco smoke is an important factor in indoor air quality and public health. In this study, the impacts of cigarette smoking on the characteristics of PM_{10} , $PM_{2.5}$, and PM_1 and their corresponding toxic metals in 50 indoor environments before and after smoking from 10 popular cigarette brands sold in Kuwait were investigated. Misleading advertisement branded 'medium strong' with less nicotine volume instigates the smokers use more cigarettes that may cause human health effects. Results showed high PM_{10} level (range 150–1100 µg m⁻³) followed by $PM_{2.5}$ (range 100–880 µg m⁻³) and PM_1 (range 52–530 µg m⁻³) after smoking. Establishment-wise, highest levels of particulate regardless the sizes were found in storehouse and lowest were observed in offices. The mean $PM_{2.5}/PM_{10}$ and $PM_1/PM_{2.5}$ ratios were in the range of 0.73–0.86 and 0.66–0.80, respectively, after smoking demonstrating that indoor PM_{10} was mostly composed of fine particles, which confirms that cigarette smoke had a dominate impact on the presence of smaller particles. Toxic metals level exhibited similar patterns as those observed in particulate in the sequence of Pb > Cd > Sb > Ba > Hg. This study recommends the preventive measures and suggests policy makers to take stringent measures against cigarette smoking in Kuwait to protect public health.

Keywords Cigarettes · Main- and sidestream smoke · Particulates · Trace metals

Introduction

Indoor air pollution is now fully recognized to be a substantial public health concern as people spend more than 90% of their time indoors (Jenkins and Counts 1999; Bennett and Koutrakis 2006). Among the indoor air pollutants, environmental tobacco smoke (ETS), particulate matter that is released either directly from a tobacco product or exhaled by a smoker, is entirely acknowledged as a key factor in indoor air quality (IAQ) and public health. Previous and recent statistics show obvious and credible evidence that there are momentous and alarming increases in cigarette smoking worldwide with around 1.1 billion people currently smoking cigarettes (World Health Organization, WHO 2018). Nearly, tobacco smoking kills more than 7 million annually

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M. AL-Harbi dr.meshari@ku.edu.kw (one in 10 deaths) and out of that 6 million deaths are as a results of direct tobacco use, whereas around 1 million deaths are the result of non-smokers being exposed to secondhand smoke (Ezzati and Loprz 2003; WHO 2018). Thus, many efforts worldwide have been made aiming to identify the smoke for different toxic metals and other components (Raju et al. 1999; Shaikh et al. 2002; Galažyn-Sidorczuk et al. 2008; Verma et al. 2010). In previous studies, it was observed that the exhaled mainstream smoke contributing 3-11% of CO, 15-43% of particles and 1-9% of nicotine to secondhand tobacco smoke (Baker and Proctor 1990; Flowers et al. 2006; Valente et al. 2007). During active smoking, particulate matter released by mainstream smoke enters the respiratory tract, whereas in the sidestream smoke, particulate matter is inhaled after dilution. Sidestream smoke particles are slightly smaller than mainstream smoke particles as they leave the cigarette (Guerin et al. 1992). Practically, sidestream smoke sustains over several minutes and results in the increase of particle size in secondhand tobacco smoke (David et al. 2014). The size of particulates (PM_{10} , PM_{25} , PM₁) is an essential feature as it defines the location of particle deposition in the respiratory system (Monn 2001), and smaller particles penetrate deeper in the respiratory system



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(Kaiser 2000). Sidestream smoke yields do not vary as they attribute to the weight of tobacco burned during smoldering. Earlier studies showed highly toxic compounds in low-tar than high tar cigarettes emitted from sidestream and secondhand smokes (Chortyk and Schlotzhauer 1989; Bodnar et al. 2012; Guo et al. 2012; Pourkhabbaz and Pourkhabbaz 2012).

Cigarette smoking was found to be the main cause for oral cancer and lung diseases than other causative factors (Hijazi et al. 2002; Abul et al. 2004; Al-Shammari et al. 2006). Satarug (2012) and Kweon et al. (2012) observed a definite link between ETS exposures and lung and heart diseases. ETS is found to cause several cases of lung cancer per year (USEPA (US Environmental Protection Agency) 1992). Tobacco smoke exposure leads to cancer, cardiovascular, respiratory diseases, and addiction (Doll et al. 2004). Al-Suraikh and Hamdy (2008) observed the incidence risk factors and possible cause for the hospital acquired pneumonia (HAP) in medical ward patients who were admitted over a 6-month period as a result of smoking habits (40.2%), besides other factors. The effect of cigarette smoking in blood and its functional activity levels in Arab population showed low white blood corpuscles (WBC) count compared to the WBC in non-smokers (Al-Awadhi et al. 2008).

Cigarette smoking is considered one of the substantial sources of toxic metals in both human body and environment (Schneider and Krivan 1993). Risk assessment tests in humans through inhalation and seafood consumption revealed the effect of Cd on glomerular and tubular damages among different genders and smokers when compared to Cd levels in non-smokers (Ju et al. 2012). Kiziler et al. (2007) correlated the elevated levels of blood Cd and Pb causing infertility in smokers in comparison to non-smokers. Cadmium (Cd) is absorbed in the blood stream easily (60%) when compared to Cd absorbed from food and water (5%). Wong and Lye (2008) recorded high mercury levels in the blood of smokers than that of non-smoking respondents. Joelle et al. (2004) reported the elevated Hg in smoking and non-smoking maternal blood of women who pass the Hg in cord blood of fetus. Besides the direct impact of ETS, the indirect effect of ETS is becoming prominent over the recent years. Previously, the elemental levels in urine of participants exposed and non-exposed to tobacco smoke was assessed (Willers et al. 2005; Scott et al. 2008; Richter et al. 2009). Their analysis was correlated among age, race, and poverty status. High cadmium, lead, antimony, and barium levels were observed in the urine of smokers compared to the elemental levels in urine of the non-smokers. Age-wise analysis showed high lead levels in the younger respondents than the older respondents. Exposure to antimony (Sb), molybdenum (Mo), barium (Ba), and cesium (Cs) primarily

occurs through dietary intake. However, exposure to barium and antimony is found to occur in contact with air, water, and soil. These elements were found high in the urine of tobacco smokers in relation to demographic characteristics (Cornelis et al. 1995). Bamgbose et al. (2007) and Jackson et al. (2011) revealed elevated trace metals level in urine and reproductive hormones among premenopausal women smokers than non-smokers. Barium becomes harmful when a person becomes susceptible to respiratory or cardiovascular illness (Scott et al. 2008). In view of the unknown health risks associated with inhaling high levels of toxic elements, Pappas et al. (2007) suggested to minimize the possible exposure of toxic substances.

Despite the fact that cigarette smoking was labeled as main precursor for numerous detrimental health impacts as mentioned above, seldom evidences are observed with reference to environmental tobacco smoke, main- and sidestream smoke, particulates from such smoke, and the factors responsible for indoor environmental pollution. Based on the above findings, this study focused in correlating the different particulate sizes (PM₁₀, PM₂₅, and PM₁), toxic metals from various cigarette types, in a range of indoor locations of Kuwait during different seasons of the years 2013 and 2015.

Materials and methods

Cigarette pretreatment

Ten different commercially branded cigarettes were selected from local market of Kuwait in this study. Prior to samples analysis, cigarettes in their original packaging were placed in pre-washed dried plastic bags separately and stored at 4 °C until tested. Subsequently, the weight of each cigarette and few characteristic features, namely taste, tar and nicotine weight after dried at 80 °C, were determined (Table 1). Ten replicates of each cigarette were selected at random from a carton containing the specific brands and from the same batch of production date. The cigarettes were weighed with and without tobacco content, with and without the filter as well, the weight of the paper that wrapped the tobacco and the filter (Table 1). Replicates of cigarettes were also individually burnt in muffle furnace at 500 °C for 2 h to produce ash. The filter and the wrapped paper were removed, and the tobacco was alone subjected to wet digestion.

Sampling sites and protocols

According to the survey conducted in this study and in the literature, the most five preferred indoor environments for



Cig. code	Cigarette brands	Weight analysis (g)				Tobacco net wt (g)	Tar (mg cig ^{-1})	Nicotine (mg cig ⁻¹)	Quality of flavor
		(I)	(I) (II) (III) (IV)		(V)				
A	Marlboro-Red	0.85	0.16	0.04	0.04	0.61	9	0.7	Strong
В	Wills F Kings	0.87	0.14	0.04	0.04	0.65	8	0.8	Moderate strong
С	Parliament Blue	0.86	0.29	0.04	0.12	0.40	9	0.8	Smooth light
D	Rothmans	0.87	0.18	0.04	0.04	0.62	8	0.7	medium blend
E	Bensen & Hedges	0.88	0.16	0.04	0.03	0.65	9	0.8	Medium strong
F	Dunhill	0.92	0.19	0.06	0.05	0.62	4	0.4	Mild blend
G	L&M	0.83	0.21	0.05	0.05	0.51	6	0.6	Thick smoke strong
Н	John Player Gold Leaf	0.86	0.16	0.04	0.04	0.63	8	0.8	Light taste
Ι	Scissors	0.83	0.14	0.04	0.04	0.61	8	0.7	Moderate light
J	Monte Carlo	0.82	0.16	0.04	0.04	0.59	10	0.7	Medium strong

Table 1 Specifications of the selected ten cigarette brands

I, cigarette with tobacco; II, filter bud; III, paper wrap without tobacco and filter; IV, filter paper wrap without filter bud, without cig. and without cig. wrap; V, net wt (I–II–III–IV); cig., ci

smoking were car parks, offices, storehouse, restaurants, and residential apartments. Thus, a total of 50 sites (10 each category) representing typical indoor venues scattered in the six governorates of Kuwait were selected. Public car parks were classified based on open car parks and closed car parks, and the aisle capacity, namely long-stay, standard and short-stay. Smoking unawares (though prohibited by law) is common at times in many parking areas. Public car parks for the ETS analysis were selected only as they were accessible for repeatability of the experiments and to avoid any confrontation in private parking areas. Most of the closed car parks in Kuwait housed approximately 1000 cars at a time in an area of 9000-192,00 sq m. The open car parks varied from 13,200 to 28,800 sq m, and they were mostly parked at ground level. Public and private offices selected ranged from 70 to 1000 sq m. The public offices are visited by approximately 50-180 visitors/day, whereas the private offices are commuted from 20 to 90 visitors/day. All offices chosen for the study had proper HVAC system. Based on the accessibility, private storehouses varying from 240 to 1080 sq m were chosen. These storehouses that packed materials of household accessories were subjected to high particulate level when compared to other selected establishments considered in this study. For restaurants, only those that allow cigarette smoking, but not water pipe smoking, in order to decouple the water pipe effects, were selected with different area ranging from 70 to 800 sq m and they are visited by approximately 40-120 visitors/day. Expatriates/ nationals apartments were also assessed for smoking habits. Sampling was conducted only in the living room (areas varied from 10 to 20 sq m) as all the other rooms were restricted to privacy of the respondents. The selection of apartment in each area also followed a pattern by conducting the analyses from a center positioned building and surrounded by other buildings in order to minimize the errors that may be attributed to outdoor environmental pollution.

The levels of particulates (PM_{10} , $PM_{2.5}$, and $PM_{1.0}$) emanating from local/branded cigarettes before and after burning (ashes) in the indoor environments described above were quantified using particulate analyzer (Hazdust EPAM 5000). Particulates were measured by gravimetric analysis, wherein particulates were collected by selective impactor and impinged on FRM filter for a specific exposure period. From each Governorate determine the seasonal variation of particulates (PM_{10} , $PM_{2.5}$, and $PM_{1.0}$) dispersed in the corresponding indoor environments. Particulate level and their inorganic composition were simultaneously assessed by both nephelometry and gravimetry methods by the EPAM-5000 analyzer.

Chemical analysis

Selected trace elements from PM_{10} , $PM_{2.5}$, and $PM_{1.0}$ samples such as barium (Ba), antimony (Sb), cadmium (Cd), lead (Pb), and mercury (Hg) were determined following the standard method (Flowers et al. 2006). The PM_{10} , $PM_{2.5}$, and $PM_{1.0}$ impacted on the FRM filter from each sampling area were pre-concentrated by acid digestion in 5% nitric acid (HNO₃) in 50-mL sterile polystyrene centrifuge tubes overnight. The samples were digested in a microwave digester (Questron Q-Wave 2000) according to the following program: power 1600 W, power setting 100%, ramp time



20 min, temperature 200 °C, hold time 15 min. In order to check for background interference, three sample preparation blanks were processed in the same manner as the samples for every digestion batch; cooled and digested samples were diluted to 50 mL with double distilled water. The resultant analyte was determined for elemental concentration in the inductively coupled plasma mass spectrometry (ICP-MS).

Contamination/pollution index (CPi)

Contamination/pollution index (CPi), which demonstrates the relative level of contamination or pollution contributed by each metal in the sample, was calculated. It is defined as the concentration of each trace metals against the reference value (Vincent et al. 2011). The reference values were obtained by using standards reported by Kabata-Pendias and Pendias (1992) for maximum permissible level of heavy metals in plants. Regulatory or reference values of heavy metals in plants and foodstuffs of plant origin are usually adopted as there are no global standards on heavy metals level in tobacco products. For the sake of brevity, contamination/ pollution index was calculated for only Pb and Cd, which pose higher threats and their levels were high compared with other trace elements. The reference values used for Pb and Cd were $(0.05-3.0 \text{ mg kg}^{-1})$ and $(0.1-0.5 \text{ mg kg}^{-1})$, respectively. The CPi values above 1.0 indicate pollution and classified into four categories, varying from moderate to excessive. CPi values below 1.0 demonstrate contamination and classified into three levels, ranging from very slight to high.

Quality assurance procedures

Quality assurance of the samples was ensured using internal standards of trace elements, blanks, and replicates. The precision of the instrument was assessed by sample recovery from Standard Reference Material (SRM-1649A, urban dust) from NIST, USA. Samples above 98% recovery were alone considered for assessment. Sample recoveries below this limit were repeated until consistent data were obtained and statistically validated.

Statistical methods

For the data treatment, both *t* tests and analysis of variance (ANOVA) procedures were used to determine the statistical significance (p < 0.05) of the differences between the determined means using Statistica[®] software.

Results and discussion

Toxic metals in different components of cigarettes

The concentrations of toxic metals (Cd, Pd, Hg, Ba, and Sb) obtained from tobacco and ash of 10 different commercially branded cigarettes during summer and winter are shown in Table 2. As multiple samples for each brand were examined, the only mean concentration along with the standard deviation (SD) for each cigarette was listed. As shown in Table 2, the mean concentration of Cd in all cigarette brands and during both seasons was $0.68 \pm 0.12 \ \mu g \ g^{-1}$, ranging from 0.18 to 1.4 μ g g⁻¹. Such observation is consistent with reported results for Cd in the UK (0.90 μ g g⁻¹), in Korean cigarettes (1.02 μ g g⁻¹) (Jung et al. 1998), and in various areas in the world (from 0.29 to 3.38 μ g g⁻¹, Watanabe et al. 1987; Ebisike et al. 2004). Lead levels ranged from 1.15 to 2.54 μ g g⁻¹, with a mean value of 1.83 ± 0.06 μ g g⁻¹. Again, Pb values in this study are comparable to those reported in UK (1.35 μ g g⁻¹), Korea (0.74 μ g g⁻¹), and Jordan (Pb at 2.67 μ g g⁻¹) (Massadeh et al. 2005; Watanabe et al. 1987). Mercury content varied in the 10 cigarette brands from 0.39 to 0.59 μ g g⁻¹, with a mean value of 0.51 \pm 0.001 μ g g⁻¹. The mean concentrations of Ba and Sb were $0.18 \pm 0.06 \ \mu g \ g^{-1}$ (range 0.11–0.31 $\mu g g^{-1}$) and 0.15 ± 0.06 $\mu g g^{-1}$ (range $0.09-0.22 \ \mu g \ g^{-1}$), respectively. High Pb and Cd concentrations followed by Hg, Ba, and Sb were observed in the ten brands of cigarettes during winter than in the summer season. Metals concentrations were found high before burning the cigarettes (tobacco) compared to after burning the cigarettes (ash) (Table 2). This indeed was expected as after smoking high percentage of toxic metals are absorbed and trapped by the filter of different branded cigarettes.

Cigarette-wise analysis revealed high concentrations of Cd in brand # D, Pb in brand # J, and Hg, Ba, and Sb in brand # C when compared to the metals level in the other brands. ANOVA analysis showed significant differences between the seasonal trace metals level in the ten cigarettes before (tobacco) and after smoking (ash) (p value < 0.05). The variation in metals level observed in ten cigarettes brands is plausibly due to the differences in the chemistry of individual tobacco leaves and later to its processing (Verma et al. 2010). Tobacco is grown over different soil types and exposed to different fertilizers and water types, and thus, the metals uptake by tobacco plants are likely to vary (Bell et al. 1992; Pappas et al. 2006). It is imperative to mention that if these metals present in higher concentrations than required



Table 2 Concentration of traceelements in tobacco and ashof different branded cigarettes $(\mu g g^{-1})$

Cig. code	ode Season		Cd	Pb	Hg	Ba	Sb
			Mean \pm SD	$Mean \pm SD$	$Mean \pm SD$	$Mean \pm SD$	Mean \pm SD
A	Summer	Tobacco	0.18 ± 0.0012	1.15 ± 0.022	0.39 ± 0.034	0.11 ± 0.009	0.09 ± 0.016
		Ash	0.05 ± 0.02	0.28 ± 0.071	0.10 ± 0.026	0.04 ± 0.032	0.03 ± 0.016
	Winter	Tobacco	0.31 ± 0.022	1.55 ± 0.042	0.45 ± 0.027	0.20 ± 0.036	0.14 ± 0.021
		Ash	0.08 ± 0.025	0.29 ± 0.046	0.11 ± 0.032	0.05 ± 0.032	0.04 ± 0.025
В	Summer	Tobacco	0.68 ± 0.032	1.57 ± 0.017	$0.46 \pm .024$	0.13 ± 0.013	0.12 ± 0.019
		Ash	0.19 ± 0.038	0.41 ± 0.038	0.13 ± 0.032	0.05 ± 0.024	0.04 ± 0.021
	Winter	Tobacco	1.19 ± 0.032	2.11 ± 0.034	0.53 ± 0.038	0.24 ± 0.029	0.19 ± 0.030
		Ash	0.31 ± 0.025	0.42 ± 0.049	0.14 ± 0.047	0.06 ± 0.032	0.05 ± 0.029
С	Summer	Tobacco	0.55 ± 0.041	1.71 ± 0.036	0.52 ± 0.034	0.17 ± 0.012	0.14 ± 0.021
		Ash	0.08 ± 0.028	0.40 ± 0.031	0.08 ± 0.024	0.03 ± 0.014	0.02 ± 0.014
	Winter	Tobacco	0.96 ± 0.042	2.30 ± 0.033	0.59 ± 0.027	0.31 ± 0.033	0.22 ± 0.042
		Ash	0.14 ± 0.036	0.42 ± 0.022	0.09 ± 0.028	0.03 ± 0.014	0.03 ± 0.012
D	Summer	Tobacco	0.80 ± 0.027	1.21 ± 0.043	0.50 ± 0.032	0.13 ± 0.021	0.12 ± 0.022
		Ash	0.23 ± 0.035	0.36 ± 0.047	0.15 ± 0.040	0.04 ± 0.016	0.04 ± 0.021
	Winter	Tobacco	1.40 ± 0.053	1.63 ± 0.039	0.57 ± 0.052	0.24 ± 0.037	0.19 ± 0.031
		Ash	0.36 ± 0.044	0.43 ± 0.044	0.16 ± 0.029	0.05 ± 0.032	0.04 ± 0.023
Е	Summer	Tobacco	0.52 ± 0.033	1.71 ± 0.029	0.51 ± 0.020	0.16 ± 0.017	0.13 ± 0.014
		Ash	0.15 ± 0.037	0.37 ± 0.056	0.15 ± 0.035	0.04 ± 0.022	0.05 ± 0.022
	Winter	Tobacco	0.91 ± 0.061	2.30 ± 0.055	0.58 ± 0.038	0.29 ± 0.051	0.20 ± 0.029
		Ash	0.24 ± 0.034	0.44 ± 0.036	0.16 ± 0.027	0.06 ± 0.027	0.06 ± 0.029
F	Summer	Tobacco	0.24 ± 0.021	1.41 ± 0.028	0.42 ± 0.022	0.13 ± 0.014	0.12 ± 0.015
		Ash	0.06 ± 0.029	0.35 ± 0.040	0.12 ± 0.020	0.04 ± 0.023	0.03 ± 0.019
	Winter	Tobacco	0.42 ± 0.062	1.90 ± 0.053	0.48 ± 0.046	0.24 ± 0.031	0.19 ± 0.037
		Ash	0.10 ± 0.026	0.36 ± 0.028	0.13 ± 0.028	0.05 ± 0.022	0.04 ± 0.015
G	Summer	Tobacco	0.57 ± 0.016	1.62 ± 0.025	0.51 ± 0.030	0.14 ± 0.013	0.13 ± 0.013
		Ash	0.12 ± 0.016	0.32 ± 0.025	0.11 ± 0.037	0.03 ± 0.013	0.03 ± 0.013
	Winter	Tobacco	1.00 ± 0.058	2.18 ± 0.041	0.58 ± 0.058	0.25 ± 0.048	0.20 ± 0.028
		Ash	0.20 ± 0.024	0.43 ± 0.038	0.12 ± 0.043	0.04 ± 0.015	0.03 ± 0.020
Н	Summer	Tobacco	0.23 ± 0.026	1.67 ± 0.027	0.47 ± 0.033	0.12 ± 0.016	0.11 ± 0.017
		Ash	0.06 ± 0.032	0.39 ± 0.041	0.13 ± 0.050	0.04 ± 0.024	0.03 ± 0.017
	Winter	Tobacco	0.40 ± 0.033	2.25 ± 0.027	0.54 ± 0.044	0.22 ± 0.030	0.17 ± 0.033
		Ash	0.10 ± 0.024	0.40 ± 0.063	0.14 ± 0.022	0.05 ± 0.025	0.04 ± 0.019
Ι	Summer	Tobacco	0.42 ± 0.029	1.67 ± 0.022	0.47 ± 0.014	0.12 ± 0.016	0.11 ± 0.026
		Ash	0.11 ± 0.039	0.42 ± 0.048	0.13 ± 0.036	0.04 ± 0.023	0.03 ± 0.015
	Winter	Tobacco	0.73 ± 0.053	2.25 ± 0.127	0.54 ± 0.038	0.22 ± 0.047	0.17 ± 0.037
		Ash	0.18 ± 0.043	0.42 ± 0.043	0.14 ± 0.020	0.05 ± 0.030	0.04 ± 0.020
J	Summer	Tobacco	0.76 ± 0.014	1.89 ± 0.021	0.48 ± 0.023	0.13 ± 0.016	0.12 ± 0.020
		Ash	0.19 ± 0.050	0.43 ± 0.049	0.12 ± 0.029	0.04 ± 0.02	0.03 ± 0.01
	Winter	Tobacco	1.33 ± 0.032	2.54 ± 0.089	0.55 ± 0.049	0.24 ± 0.035	0.19 ± 0.044
		Ash	0.20 ± 0.033	0.48 ± 0.043	0.13 ± 0.028	0.05 ± 0.022	0.04 ± 0.028

for the human body, they become very toxic. Several detrimental health outcomes were reported in the previous studies caused by tobacco smoke including cancer, cardiovascular, respiratory diseases, impaired fertility, arterial stiffness, and addiction (Doll et al. 2004; Pretorius2012; Raymond et al. 2012; Rom et al. 2012). Other health outcomes are also reported in the introduction section.



Indoor PM₁₀, PM_{2.5}, and PM₁ masses

Particulate (PM₁₀, PM₂₅, and PM₁) profiles of 50 indoor environments during the years of 2013 and 2015 in winter and summer seasons before and after cigarette smoking are illustrated in Figs. 1 and 2. Prior smoking and during winter (Fig. 1), minimum PM₁₀ concentration was detected in offices with a value of 95 μ g m⁻³ and maximum value of 750 μ g m⁻³ was observed in storehouses, with mean concentrations ranged from 250 (offices) to 480 μ g m⁻³ (storehouses). Similar observations were also observed with PM_{2.5} and PM_{1.0} with a range of 63–530 μ g m⁻³ and 48–350 μ g m⁻³, with means varied from 182 to 300 μ g m⁻³ and from 110 to 210 $\mu g\ m^{-3},$ respectively. As expected, the influence of smoking was evident where PM_{10} , PM_{25} , and PM₁ concentrations were significantly higher at the sites after cigarette smoking episode. The minimum and maximum span was increased to be from 150 to 1100, 100 to 880, and 52 to 536 μ g m⁻³ for PM₁₀, PM_{2.5}, and PM₁, respectively. Mean concentrations were also shifted to the range of 350–520 $\mu g~m^{-3}$ for PM_{10} and 220–410 $\mu g~m^{-3}$ and 220–410 μ g m⁻³ for PM_{2.5} and PM₁, respectively. Similar patterns were also observed during summer season (Fig. 2), though with less magnitudes. The differences observed between PM means seasonal-wise, establishment-wise, and (before/after) smoking were statistically significant (p value < 0.05). Additionally, PM_{10} concentrations were well correlated with PM2 5 and PM1 after cigarette smoking incident as Pearson correlation coefficients (R) were in the range of 0.88 and 0.93, for all indoor environments investigated. This indicates that PM fractions at these sites were influenced by similar sources.

To further study the association between PM fractions, PM_{2.5}/PM₁₀ and PM₁/PM_{2.5} mass concentration ratios were also evaluated for all tested indoor venues and results are shown in Table 3. The mean PM_{25}/PM_{10} and PM_{1}/PM_{25} ratios of before cigarette smoking event were in the range of 0.54-0.72 and 0.52-0.71, respectively, and the corresponding ranges were 0.73-0.86 and 0.66-0.80 when cigarette smoking was experienced. The ratios found in all indoor environments in this study were consistent with those reported in the previous studies (0.77, Slezakova et al. 2007) and (0.74–0.95, Slezakova et al. 2009). High values of PM2.5/PM10 and PM1/PM2.5 ratios demonstrates that indoor PM₁₀ were mostly composed of fine particles, which confirms that cigarette smoke had a dominate impact on the presence of smaller particles. The low values of PM_{2.5}/PM₁₀ and PM₁/PM_{2.5} ratios were possibly

caused by occasional ventilation of outdoor air with a higher proportion of coarse particles released from road constructions that were adjacent some venues during the monitoring campaign.

Particulate-wise analysis showed higher PM_{10} level than both $PM_{2.5}$ and $PM_{1.0}$ in the selected establishments. Among the selected establishments particulates, irrespective of particle size, were highly found in the sequence of storehouses > restaurants > car parks > residential apartment > offices during summer and winter (Figs. 1, 2). In general, observations showed high PM_{10} , $PM_{2.5}$ and $PM_{1.0}$ level in the selected establishments during winter than in the summer seasons (Figs. 1, 2). Comparatively, particulates levels were high 'after smoking' than 'before smoking' (Figs. 1, 2).

The high concentrations of particulates $(PM_{10}, PM_{25}, and$ $PM_{1,0}$ level in the selected public and private establishments could be related to the activities by the smoking and nonsmoking respondents during the winter season and the long hours spent by them in the indoor environment. This phenomenon was found in line with the earlier studies (Flowers et al. 2006; Repace and Lowrey 1985; Al-Mulla et al. 2008; Qibin et al. 2011). Particulates $(PM_{10}, PM_{2.5}, and PM_{1.0})$ were high after smoking in confined area because the particulates released from the ash and smoke of cigarettes added the existing particulates from the outdoor to the indoor environment. Such observation was observed in the earlier studies (Neergaard et al. 2007; Lydia et al. 2013). Particulates in storehouses attributed to the frequent transport of materials allowing the external particulates to gain entry into the storehouse. This phenomenon was found in line with the earlier studies (Jenkins and Counts 1999; Bennett and Koutrakis 2006; Chen and Zhao 2011) in addition to (a) the particulates released from the packed products, (b) circulation of deposited particulates in the confined area where cleaning was partially carried out and, (c) where HVAC system was temporarily shut down during the winter seasons or when it was partially functional. Succeeding to the high particulates level in storehouses was that of the particulates in most of the restaurants surveyed especially during winter seasons, when more customers and demand of consumption and food items were utilized and when air-conditioning system was temporarily reduced. Valente et al. (2007) reported such high particulate level and ETS levels were as high as 1.5-4.5 times in such public hospitality, viz. restaurants, when compared to people who lived with smokers. Offices showed low particulate level among the other selected establishments indicating some maintenance (vacuum cleaning), use of effective air-conditioning systems throughout the day, and restricted number of respondents inhabiting the offices



Fig. 1 Box plots of PM10, PM25, and PM1 concentration before smoking (BS) and after smoking (AS) during winter season

when compared to the number of people commuting the other establishments. Public offices showed more particulate level than private offices which reflects the extent of poor maintenance of public offices. Similar observations were on par with the earlier findings (Luoma and Batterman 2001; Qibin et al. 2011).





Fig. 2 Box plots of PM₁₀, PM_{2.5}, and PM₁ concentration before smoking (BS) and after smoking (AS) during summer season

Table 3	Descriptive statistics of	PM_{25}/PM_{1}	$_0$ and PM ₁ /PM ₂ 5	ratios at monitoring sites b	efore smoking (BS) and afte	r smoking (AS)
	1	64	V I/ 4	6		<u> </u>

Site	Season	Smoking status	Ave.	SD	Min	Max	25th percentile	Median	75th percentile
PM _{2.5} /PM ₁₀				1					
Offices	Summer	BS	0.62	0.00	0.6	0.64	0.61	0.62	0.63
		AS	0.74	0.01	0.73	0.75	0.73	0.75	0.75
	Winter	BS	0.72	0.05	0.68	1.11	0.68	0.68	0.68
		AS	0.74	0.00	0.7	0.75	0.71	0.74	0.75
Car parks	Summer	BS	0.68	0.10	0.29	1.13	0.62	0.62	0.62
		AS	0.76	0.25	0.34	1.71	0.73	0.78	0.79
	Winter	BS	0.69	0.11	0.43	1.43	0.68	0.68	0.68
		AS	0.74	0.06	0.51	1.21	0.74	0.74	0.74
Storehouses	Summer	BS	0.54	0.26	0.11	1.41	0.28	0.52	0.61
		AS	0.80	0.03	0.74	0.81	0.79	0.81	0.81
	Winter	BS	0.72	0.17	0.68	1.16	0.68	0.68	0.68
		AS	0.86	0.05	0.74	1.14	0.74	0.74	0.74
Restaurants	Summer	BS	0.56	0.15	0.12	1.31	0.29	0.62	0.62
		AS	0.78	0.03	0.67	0.79	0.79	0.79	0.79
	Winter	BS	0.74	0.19	0.68	0.90	0.68	0.68	0.68
		AS	0.79	0.15	0.74	1.01	0.74	0.74	0.74
Apartments	Summer	BS	0.70	0.01	0.1	0.90	0.28	0.64	0.85
		AS	0.76	0.00	0.75	0.76	0.76	0.76	0.76
	Winter	BS	0.70	0.10	0.68	1.13	0.68	0.68	0.68
		AS	0.80	0.18	0.74	1.27	0.74	0.74	0.74
PM _{1/} PM _{2.5}									
Offices	Summer	BS	0.58	0.12	0.56	1.11	0.56	0.56	0.56
		AS	0.67	0.16	0.17	1.21	0.41	0.63	0.85
	Winter	BS	0.61	0.05	0.16	0.61	0.67	0.67	0.67
		AS	0.66	0.00	0.6	0.78	0.61	0.62	0.63
Car parks	Summer	BS	0.52	0.00	0.52	0.57	0.53	0.55	0.56
		AS	0.72	0.10	0.13	1.09	0.46	0.63	0.82
	Winter	BS	0.60	0.00	0.55	0.63	0.62	0.63	0.67
		AS	0.68	0.00	0.6	0.86	0.61	0.64	0.65
Storehouses	Summer	BS	0.71	0.38	0.53	1.04	0.56	0.56	0.56
		AS	0.80	0.37	0.2	1.07	0.56	0.64	1.00
	Winter	BS	0.60	0.04	0.38	0.63	0.67	0.67	0.67
		AS	0.66	0.04	0.29	0.67	0.61	0.61	0.61
Restaurants	Summer	BS	0.71	0.15	0.18	0.75	0.52	0.62	1.37
		AS	0.77	0.14	0.21	0.79	0.56	0.65	0.77
	Winter	BS	0.61	0.12	0.42	0.64	0.67	0.67	0.67
		AS	0.66	0.03	0.33	0.69	0.61	0.61	0.61
Apartments	Summer	BS	0.64	0.14	0.56	1.08	0.56	0.56	0.56
		AS	0.80	0.17	0.29	1.09	0.70	0.75	0.92
	Winter	BS	0.62	0.05	0.31	0.67	0.67	0.67	0.67
		AS	0.69	0.00	0.62	0.77	0.63	0.65	0.66

Toxic elements constituents of indoor PM_{10} , $PM_{2.5}$, and $PM_{1.0}$

Levels of toxic elements cadmium (Cd), lead (Pb), mercury (Hg), barium (Ba), and antimony (Sb) in particulates in different venues before and after cigarette smoking were also investigated, and average summer and winter results are shown in Fig. 3. The concentration of toxic metals in PM_{10} after cigarette smoking ranged, Cd: from 7.20 to 11.40 µg g⁻¹ with mean of 9.80 ± 1.8 µg g⁻¹, Pb: from 19.90 to 35.90 µg g⁻¹ with mean of 29.60 ± 6.0 µg g⁻¹, Hg: from 3.90 to 7.22 µg g⁻¹ with mean of 6.30 ± 1.37 µg g⁻¹, Ba: from





Fig. 3 Comparison of average concentration of trace elements at monitoring sites before smoking (BS) and after smoking (AS)

4.26 to 7.54 μ g g⁻¹ with mean of 6.51 ± 1.31 μ g g⁻¹, and Sb: from 4.23 to 7.60 μ g g⁻¹ with mean of 6.84 ± 1.46 μ g g⁻¹. After cigarette smoking, the toxic elements concentrations increased by 1.0–1.30 times compared with prior smoking. Establishments-wise, toxic metals level exhibited similar patterns as those observed in PM₁₀. Similar profiles of toxic metals were also found with PM_{2.5} and PM_{1.0} (Fig. 3), but with lower magnitudes. High elemental concentrations in the particulates (PM₁₀ PM_{2.5} and PM_{1.0}) were found in the sequence of Pb>Cd>Sb>Ba>Hg irrespective of the seasons and during both 'before' and 'after' the smoking act. Results showed significant differences (*p* value < 0.05) between the elemental levels and establishments before and after smoking in PM₁₀, PM_{2.5}, and PM_{1.0}.

Trace metals were found high in establishments where smoking was either encouraged or where no stringent monitoring was not undertaken attributing the release of trace metals from the cigarette smoke and ash. High Pb and Cd in selected indoor environment 'after' a smoke indicated these predominant metals from tobacco besides the release of trace metals existing in the indoor ambient atmosphere. Similar observation was also recorded by Luoma and Batterman (2001), Wong and Lye (2008), Perez et al. (2011) and Satarug (2012).

Contamination/pollution index (CPi)

Elevated concentrations of heavy metals found in different cigarette brands in this study could be ascribed to several factors related to tobacco plants. These include composition of tobacco plants itself, constituents of soil and dust, merging of different tobacco types with different metal contents, and application of fertilizers (Vincent et al. 2011). Additionally, it was reported that land pollution is considered one of the main contributors for high level of toxic metal in cigarettes (Vincent et al. 2011). Contamination/pollution index (CPi) for Pb in tobacco and ash of the 10 different cigarette brands was calculated during summer and winter seasons (Fig. 4). All CPi values for Pb in tobacco were above 1.0, indicating pollution effects. The CPi values in tobacco (Fig. 4a) varied from moderate pollution (samples A and D) to very high (sample C) in summer, whereas in winter CPi values ranged from high pollution (samples A, B, D, and F) to excessive pollution (sample C). The level of samples pollution in winter was higher than that in summer, which is indeed expected as Pb concentrations in winter were higher than those in summer. The CPi values for Pb in ash were contradicted to those in tobacco samples, where all CPi values were below 1.0, indicating contamination effects (Fig. 4b). All cigarette types exhibited high contamination, and no changes were observed seasonal-wise. The CPi for Cd in tobacco and ash for all cigarette brands was also calculated during summer and winter seasons (Fig. 5) Again,

all CPi values for Cd in tobacco were above 1, demonstrating pollution effects. In summer, CPi values in tobacco (Fig. 5a) fluctuated from moderate pollution (samples A, F, and H) to excessive pollution (sample C, D, and J), while in winter CPi values altered from moderate pollution (samples A) to excessive pollution (samples B, C, D, E, G, I, and J). The CPi values for Cd in winter were higher than those in summer. The CPi values for Cd in ash were distributed between pollution and contamination effects (Fig. 5b). According to CPi values for Cd in ash, samples showed moderate to high contamination in summer whereas moderate to high pollution in winter.

Exposure to high level of heavy metals is associated with number of health concerns include lung disease, cancer (Fowles and Dybing 2003), and other systemic illnesses such as difficulties of pregnancy and peripheral artery disease (Milnerowicz et al. 2000; Navas-Acien et al. 2004). Elevated Pb and Cd found in smokers and passive smokers blood and urine have been classified as Group I and Group IIB carcinogens, respectively (Smith et al. 1997, 2003). It was also reported that Pb and Cd could replace Zn and Ca, leading to the harshness of hypertension problems attributable to their accumulation in kidneys, which hurt their aptitude to adjust the water balance in the body (Afridi et al. 2013). This would in turn cause water and salt retention, and high blood pressure (Staessen et al. 1996). The levels of Hg, Ba, and Sb were lower than Pb and Cd in this study. Health risks associated with Ba include muscle twitching, gastrointestinal dysfunction, paralysis, elevated blood pressure, and low blood potassium (Acobs et al. 2002). High Ba toxicity may cause kidney damage, respiratory failure, and death (Johnson and VanTassell 1991). In laboratory animals, several health outcomes were reported include pneumoconiosis, altered electrocardiograms, and stomach ulcers, as results of long-term exposure of antimony compounds (Cooper and Harrison 2009). Mercury toxicity includes incidence of cognitive impairment, especially in children (Cobbina et al. 2011). Reports also stated their chronic ailments observed elsewhere the Globe (Doll et al. 2004; Joelle et al. 2004; Al-Shammari et al. 2006). Therefore, a combined effect of smoking cigarettes and their constituents enhanced by particulates concentration besides the dietary influence of trace metals was found to cause detrimental effects in the humans (Eatough et al. 1989; Al-Suraikh and Hamdy 2008; Raymond et al. 2012).

Lead is a crucial environmental hazard for children, predominantly known for its neurocognitive effects (Lanphear et al. 2000). Cigarette smoking can not only hurts the smokers but also induces severe health outcomes to passive smokers, mostly in children and old-age person (Maninno et al. 2003). Cd is volatile in nature, and thus, it can easily find its way in the blood stream of the smoker and accumulate simply in the organs (Wu et al. 1997; Stephens et al. 2005).





Fig. 4 Contamination/pollution index for Pb in a tobacco and b ash

The present study recommends the environmentalists and policy makers to undertake suitable measures to curb particulates and the various constituents that emanate from the cigarette smoke to protect public health. Such measures may include establishing de-addiction/healthcare centers in various parts of the country that could decrease the number of smokers in the near future, restrict marketing of cigarettes and tobacco products through vendors, distant sales (internet), to children and adolescent, and advertise the detrimental impacts of cigarettes. Additionally, encourage research studies with priorities in order to waylay mechanisms that may prevent not only smoking in the public but also augment a total ban to cigarette and related smoking products.

Conclusion

The finding of this study confirms that environmental tobacco smoke (ETS) is a notable source of different particulate matter sizes (PM_{10} , $PM_{2.5}$, and PM_1) and several toxic metals, particularly Pb, Cd, Hg, Ba, and Sb in the 50 indoor environments tested. Establishment-wise analysis showed





Fig. 5 Contamination/pollution index for Cd in a tobacco and b ash

high trace metals concentrations in particulates found in storehouse and lowest observed in offices, demonstrating the influence of population density and compact spaces in addition to the cigarette smoke that added the pollutant levels in the selected indoor environment and affecting the respondents. Furthermore, it is speculated that dietary intake of toxic elements (Pb, Cd, Hg, Ba, and Sb), most probably through smoking cigarettes, may increase the risk of several health outcomes such as cancer, cardiovascular, and respiratory diseases. Thus, this study suggests the availability of proper enlightenment programs to create awareness on the dangers of particulates matter, especially with small sizes (i.e., $PM_{2.5}$ and PM_1) and their corresponding toxic metal poisoning among the main and passive smokers to enable a smoke free environment and protect human health.

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