



## PM<sub>2.5</sub> Emissions from Hand-Held Sparklers: Chemical Characterization and Health Risk Assessment

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### ABSTRACT

The central goal of this work is to improve our understanding of the distinctive and unusual chemistry of aerosol particles released from firework displays and their potential health risks due to inhalation exposure. The chemical composition of fine particles (PM<sub>2.5</sub>) released from three commonly used sparklers (low smoke sparklers (LSS), whistling sparklers (WS) and colored sparklers (CS)) was investigated. In particular, total and water soluble elemental fractions (22 elements) and 13 polycyclic aromatic hydrocarbons (PAHs) in PM<sub>2.5</sub> were quantified. The particulate emissions from LSS had relatively lower particulate-bound metals and less water-soluble fractions in them compared to those from WS and CS. However, PAHs were found to be relatively higher in LSS compared to those from WS and CS. Health risk due to inhalation of PM<sub>2.5</sub> emitted from all the three types of sparklers was estimated for various dilution conditions including the case with little or no dispersion of particles. It was observed that WS had the highest carcinogenic risk ( $25 \times 10^{-6}$  for adults and  $75 \times 10^{-6}$  for children) followed by CS ( $2.6 \times 10^{-6}$  for adults and  $7.9 \times 10^{-6}$  for children) and LSS ( $7.6 \times 10^{-7}$  for adults and  $2.6 \times 10^{-6}$  for children) for the worst case scenario of no dilution of emitted particles during inhalation. This carcinogenic risk is pronounced only when there is no or very low dilution (~10) of emitted particles during inhalation. The health risk estimates for all sparkler types are below acceptable limits for dilution factors above 80 and thus exposure to PM<sub>2.5</sub> in sparkler emissions is unlikely to have serious health effects. The water soluble fraction of metals (bioavailable metals) made a major contribution to the carcinogenic health risk due to inhalation of PM<sub>2.5</sub> released from WS (~100%) and CS (~96%) while PAHs played a major role in the carcinogenic risk associated with PM<sub>2.5</sub> from LSS (~66%).

**Keywords:** Sparklers; Metals; Polycyclic aromatic hydrocarbons; Health risk estimates.

### INTRODUCTION

Fireworks release a range of fine (PM<sub>2.5</sub>; Aerodynamic Diameter (AED)  $\leq 2.5 \mu\text{m}$ ), ultrafine (UFPs; AED  $\leq 100 \text{ nm}$ ) and nanoparticles (NPs; AED  $\leq 50 \text{ nm}$ ) during their use in festive occasions and celebrations. Although firework displays last for a short duration of time, they increase the atmospheric particulate loading significantly (Moreno *et al.*, 2007; Wang *et al.*, 2007; Vecchi *et al.*, 2008; Joly *et al.*, 2010; Sarkar *et al.*, 2010). For example, Sarkar *et al.* (2010) reported an increase in airborne particulate matter (PM) concentration ( $317.2\text{--}616.8 \mu\text{g}/\text{m}^3$ ) by factors ranging from 6 to 12 compared to the WHO guidelines ( $50 \mu\text{g}/\text{m}^3$ ) during Diwali festival celebrations in India. Joly *et al.* (2010) found that the peak concentration of PM<sub>2.5</sub> during the firework displays was 1000 times higher than their corresponding

background levels ( $10\text{--}12 \mu\text{g}/\text{m}^3$ ). People present in the vicinity of firework displays may be exposed to extremely high levels of inhalable airborne particles (Joly *et al.*, 2010). The particulate emissions released from pyrotechnic (firework) displays are highly metallic in nature and thus pose a serious threat to human health, especially when the concentrations of toxic metals such as Cr, Cu, Ni, As and Ba are high (Becker *et al.*, 2000; Ravindra *et al.*, 2003; Kulshrestha *et al.*, 2004; Steinhäuser *et al.*, 2008; Vecchi *et al.*, 2008; Habib and Lohrer, 2012; Sterba *et al.*, 2013). The oxidative burden associated with particulate matter (PM) released from firework displays has been examined by Godri *et al.* (2010) who reported that the oxidative potential of PM per unit mass derived from fireworks is significantly greater than that from traffic generated PM. Most of the studies on pyrotechnic emissions focused on the chemical characterization of PM collected from the ambient air during festive seasons (Drewnick *et al.*, 2006; Wang *et al.*, 2007; Van der Kamp *et al.*, 2008; Vecchi *et al.*, 2008). However, little is known about the chemical composition of PM released from handheld pyrotechnics such as sparklers despite concerns over health risk associated with inhalation

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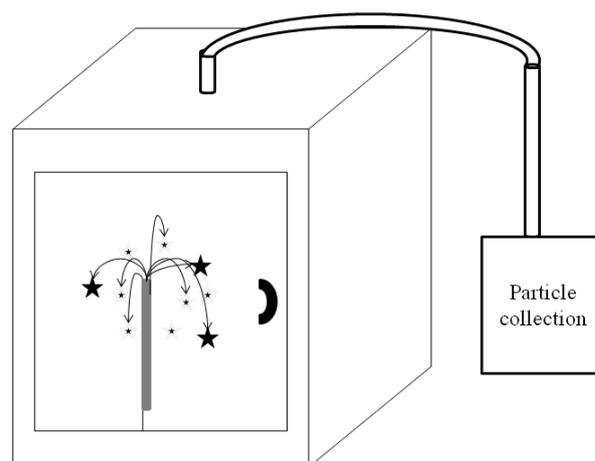
exposure (Betha and Balasubramanian, 2012). Sparklers mainly consist of a metal rod or a thin tube coated with an explosive mixture to burn producing colorful sparks (Russell, 2000). There are four major components in sparklers: a metallic component to generate sparks (aluminium, zinc, magnesium etc.); an oxidizing component (chlorates, nitrates, perchlorates, etc.); a combustible binder (dextrin, nitrocellulose, gum Arabic etc.); and a fuel component (e.g., charcoal) (McManus *et al.*, 1975; Helmenstine, 2012). The emission quality of sparklers depends on the relative proportions of various chemical constituents contain in them and the specific combustion dynamics.

Sparklers are used by people of different age groups ranging from children to adults and elderly during New Year celebrations, birthday parties and other festive occasions such as Diwali in India (Kulshrestha *et al.*, 2004). Our recent study has demonstrated that nearly 62–83% of airborne particles emitted from various sparklers are in the nanometer size range (Betha and Balasubramanian, 2012). Such small particles can easily penetrate deep into the respiratory system and be taken up by cells in the alveolar region (Oberdorster *et al.*, 2005; Buzea *et al.*, 2007). The metallic nature of these particles could potentially generate reactive oxygen species (ROS) such as hydroxyl radicals (OH) within the body through biochemical reactions with endogenously produced hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) (Halliwell, 1991; Li *et al.*, 2003; See *et al.*, 2007). These ROS are known to induce oxidative stress leading to several health problems (Garshick *et al.*, 2004; Patel *et al.*, 2011). Despite the wide use of sparklers and emissions of metallic aerosols from them, the chemical characteristics of these particulate emissions and the associated health risk remain poorly understood to date. In this study, the chemical characteristics of fine particles (PM<sub>2.5</sub>) released from three different types of commonly used sparklers (low smoke, whistling and colored sparklers) were investigated for the first time. The total particulate-bound metals in PM<sub>2.5</sub> and the corresponding water-soluble fractions together with particulate-bound polycyclic aromatic hydrocarbons (PAHs), emitted from the sparklers, were quantified. In addition, the human health risk due to inhalation of the particulate-bound elements and PAHs was estimated for both adults and children.

## MATERIALS AND METHODS

### Particulate Sampling

An experimental steel chamber (Fig. 1.) with 0.5 m × 0.5 m × 0.5 m dimensions was used to collect PM<sub>2.5</sub> (AED ≤ 2.5 μm) emitted from sparklers in a controlled environment (T = 25°C and RH = 60%). Sparklers were placed in the centre of the chamber and ignited to burn. The chamber was operated without any input of external clean air for dilution of emitted particles. Three most commonly available sparklers were chosen for this study: (1) whistling sparklers (WS), (2) low smoke sparklers (LSS) and (3) colored sparklers (CS). PM<sub>2.5</sub> samples from the sparkler emissions were collected on both polytetrafluoroethylene (PTFE) membrane (PALL corp, USA) and pre-combusted 47 mm quartz fiber filters (Whatman, USA) using portable MiniVol



**Fig. 1.** Experimental setup used for particulate collection.

air samplers (Air metrics, USA). PM emitted from sparklers was drawn at a flow rate of 5 L/min for 20 min onto the filters. Filter samples were collected in duplicates for each type of sparkler and for each filter type. These filters were conditioned in a dry box at a constant relative humidity and temperature (T = 22°C and RH = 32%) for 24 hours prior to weighing in a microbalance (Sartorius, MC 5). The filters were stored at 4°C until extraction and analysis. The experiment was repeated 3 times for each type of sparkler.

### Extraction of Total and Water Soluble Metals

Teflon (PTFE) filters were used for determination of total particulate-bound metals and water soluble metals. Each filter sample was cut into half using ceramic scissors to prevent any contamination. One half of the filter was used for total metal extraction using acid digestion (Karthikeyan *et al.*, 2006a). Briefly, 4 mL of 69.5% nitric acid (HNO<sub>3</sub>), 2 mL of 30% hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) and 0.5 mL of 48% hydrofluoric acid (HF) were added to the filters in a Teflon vessel. A MLS1200 mega closed-vessel microwave digestion system was used for digestion at 250 W for 10 min, followed by 400 W for 10 min and finally, 600 W for 4 min. The extracts were then filtered through a 0.45 μm PTFE membrane filter, diluted with ultrapure water and refrigerated at 4°C until analysis.

For water soluble metals and ions, the other half of the filter was used. The filter was extracted with 20 mL of ultrapure water in an ultrasonic bath and maintained at 60°C, for 1 hour. 10 mL of the extract was used for measurement of inorganic ions. The remaining 10 mL was used for measurement of water soluble metals. HNO<sub>3</sub> was added to the water soluble metal extract to lower the pH below 2 and was stored at 4°C until analysis. All the extracts were filtered through 0.45 μm PTFE membrane filter.

### Extraction of Polycyclic Aromatic Hydrocarbons (PAHs)

Quartz fiber filters were cut into half using ceramic scissors and extracted for PAHs using microwave digestion as described in Karthikeyan *et al.* (2006b). A 20 mL mixture of acetone: hexane (1:1) was used as extraction solvent. Filters were added into the solvent in a Teflon vessel and

were digested at 150 W for 20 minutes. The extracts were subsequently concentrated to approximately 3 mL using a rotary evaporator at room temperature and further to 0.5 mL by a nitrogen gas stream. The extracts were stored at  $-20^{\circ}\text{C}$  until analysis.

#### **Total and Water Soluble Metal Analysis**

The total metal and water soluble metal extracts were analyzed using ELAN 6100 Inductively Coupled Plasma Mass Spectrometer (ICP MS) equipped with a cross-flow nebuliser and a Quartz torch. The plasma gas, auxiliary gas and nebuliser gas flow rates were 16, 1 and 0.93 L/min, respectively. Filter extracts were introduced at a rate of 1 mL/min and waste was removed from the nebulizer through a Gilson Miniplus 2 peristaltic pump. Field blanks, obtained by placing filters in the filter holder with no samples being drawn through them, and laboratory blanks, unexposed filters, were also extracted and analyzed along with filter samples and were subtracted from the concentrations obtained from samples. Filter handling was done with stainless steel forceps. Spiked samples were tested after every 10 samples to confirm the validity of the system calibration as part of the quality control/quality assurance protocol. SRM 1649a (urban particulate matter) was used to assess the accuracy of analysis. The recovery ranged from 82–103% for the elements studied in this work.

A 5 point calibration curve using 20, 40, 60, 80 and 100 ppb (ng/mL) standards diluted from the inductive couples plasma (ICP) multi-element standard solution in diluted nitric acid was carried out before each run. The regression coefficients ranged from 0.9966 (Cu) to 0.9996 (Mg).

#### **PAHs Analysis**

The standard EPA 610 polycyclic aromatic hydrocarbon mix which consists of 16 native PAHs (naphthalene (Naph), acenaphthalene (Ace), acenaphthene (Acy), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzopyrene (BaP), indeno[1,2,3-cd]pyrene (Ind), dibenz[a,h]anthracene (DBA) and benzo[g,h,i]perylene (Bpe)) in methanol:methylene chloride (Supelco, USA) was used. A calibration curve was plotted using 1, 5, 10, 50, 100, 200, 500  $\mu\text{g/L}$  dilutions from the standard stock solution. The regression coefficients ranged from 0.990 (Flt) to 1.000 (Pyr).

Sample analysis was performed using a QP2010 GC-MS equipped with a Shimadzu AOC-5000 auto injector and a DB-5 fused silica capillary column (30 cm  $\times$  0.25 mm i.d.; film thickness 0.25 mm) with purified helium as carrier gas. The PAH standards as well as samples were analyzed in selective ion monitoring mode (SIM) with a detector voltage of 700 V and a scan range of 50 to 500 m/z. The most abundant ions were selected for quantification; and one to two ions were used for confirmation of each analyte. The GC temperature program used is as follows:  $70^{\circ}\text{C}$  for 2 minutes;  $15^{\circ}\text{C}/\text{min}$  to  $200^{\circ}\text{C}$ , hold for 4 minutes; and  $5^{\circ}\text{C}/\text{min}$  to  $300^{\circ}\text{C}$ , hold for 5 minutes.

#### **Human Health Risk Assessment**

Human health risk assessment was conducted based on the mean concentrations of water soluble metals and PAHs determined through the experimental study. Health risk assessment is especially useful in understanding the health hazard associated with inhalation exposure to PM emitted from the three different sparklers. The steps involved in health risk assessment are described in detail elsewhere (See and Balasubramanian, 2006). Briefly, it involves four recognized steps: (a) Hazard identification (b) Exposure assessment (c) Dose-response assessment and (d) Risk Characterization.

Elements such as Cd, Cr, Co and Ni and organic compounds such as Naph, BaA, BbF, BkF, BaP, Ind, DBA are classified as probable and possible carcinogenic compounds by International Agency for Research on Cancer (IARC) (IARC, 2013). Therefore these compounds were used in estimation of cancer risk. The risk depends on exposure concentration (EC) which is calculated by the following equation

$$\text{Exposure Concentration (EC) (mg/m}^3\text{)} = \frac{C_a \text{ (mg/m}^3\text{)} \times \text{ET (h/day)} \times \text{EF (day/year)} \times \text{ED (year)} \times \text{ADAF}}{\text{AT (h)}} \quad (1)$$

where  $C_a$  is the ambient air concentration ( $\mu\text{g/m}^3$ ) of a compound, ET is exposure time (h/day), EF is exposure frequency (day/year), ED is exposure duration (years), AT is averaging time (life time in years  $\times$  365 days/year  $\times$  24 h/day) and ADAF refers to age dependent adjustment factor.

Chronic daily intake (CDI) is then calculated from the following equation.

$$\text{CDI (mg/kg/day)} = \frac{\text{Total Dose (TD, mg/m}^3\text{)} \times \text{Inhalation Rate (IR, m}^3\text{/day)}}{\text{Body Weight (BW, kg)}} \quad (2)$$

$\text{TD} = \text{EC} \times \text{E}$  where EC is exposure concentration of particulate-bound pollutants and E is deposition fraction of particles by size (Volckens and Leith, 2003),  $\text{E} = -0.081 + 0.23 \ln(\text{Dp})^2 + 0.23 \sqrt{\text{Dp}}$  where Dp ( $\mu\text{m}$ ) is the diameter of particles. Two groups of people (adults and children) were considered for risk assessment. BW is typically assumed to be 70 kg for adults and 15 kg for children. IR is assumed to be  $20 \text{ m}^3/\text{day}$  for adults and  $10 \text{ m}^3/\text{day}$  for children. The excess life time cancer risk is given by  $(\text{ELCR}) = \text{CDI} \times \text{SF}$ , where SF is the inhalation slope factor ( $\text{SF, mg/kg/day}^{-1}$ ) calculated from inhalation unit risk values ( $\text{IUR, (mg/m}^3\text{)}^{-1}$ ) provided by USEPA.

## **RESULTS AND DISCUSSION**

#### **Total Metal Concentration in Sparkler Emissions**

$\text{PM}_{2.5}$  mass emissions for three different types of sparklers (a) LSS, (b) WS and (c) CS were found to be  $37.3 \pm 4.0 \text{ mg/m}^3$ ,  $140.1 \pm 2.9 \text{ mg/m}^3$ , and  $174.3 \pm 7.2 \text{ mg/m}^3$ ,

respectively. Total metal concentrations of the particles were analyzed and are shown in Fig. 2. A total of 22 elements which are commonly used constituents in manufacturing of pyrotechnics were quantified: Ag, Al, B, Ba, Bi, Ca, Cd, Co, Cr, Cu, Fe, In, K, Li, Mg, Mn, Na, Ni, Pb, Sr, Tl and Zn. The metals that had the highest concentrations for all the three sparkler types were K, Ca, Fe, Zn and Ba. These five metal components are essential elements for fireworks. K is a major component of fireworks (Vecchi *et al.*, 2008), with up to 74% of sparkler powder consisting of  $\text{KNO}_3$  as an oxidizer, while Fe is often used as a combustion agent (Croteau *et al.*, 2010).

The WS had the highest concentration of metals across the range of all 22 elements analyzed. The significant amount of metals in WS suggests a higher amount and more complex mix of sparkler ingredients to produce the whistling effect. It also had a much higher concentration of Pb, almost 100 times more than the other two types of sparklers. Pb is usually present in the form of lead (IV) oxide as an oxidant and igniter. However, it also produces crackling sound in pyrotechnics which might account for the high levels of Pb in WS to produce the whistling sound at the start of burning (Croteau *et al.*, 2010). The CS contained much higher concentrations of Sr compared to the low smoke and whistling sparklers as Sr is the metal used for displaying red color in the sparklers (Moreno *et al.*, 2007). CS and WS had higher concentrations of Ca, Mg and Al compared to LSS as these three metals are used to create more vividly colored sparks which were observed during burning compared to LSS (Morena *et al.*, 2008; Camilleri and Vella, 2010). LSS emissions were observed to have extremely low K concentrations compared to the other two types of sparklers, in the order of 100 times or less, reflects that small amounts of  $\text{KNO}_3$  or  $\text{KClO}_4$  are used as an oxidizer in those sparklers. Unlike other sparklers where charcoal forms the major fraction of the fuel, LSS have nitrocellulose-based fuels with a small percentage of charcoal added to it. Due to lower amounts of charcoal in

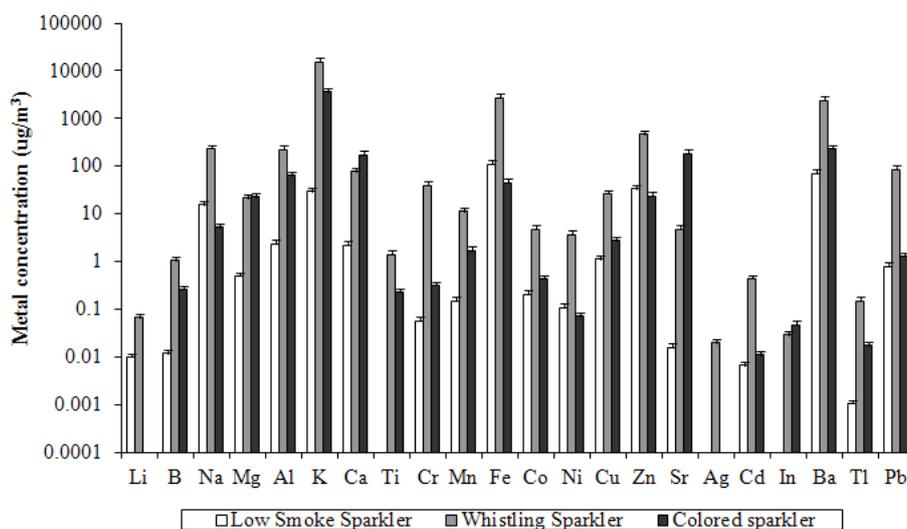
LSS, the amount of oxidizer required for combustion is understandably much lower compared to CS and WS.

#### **Water Soluble Metal Concentration in Sparkler Emissions**

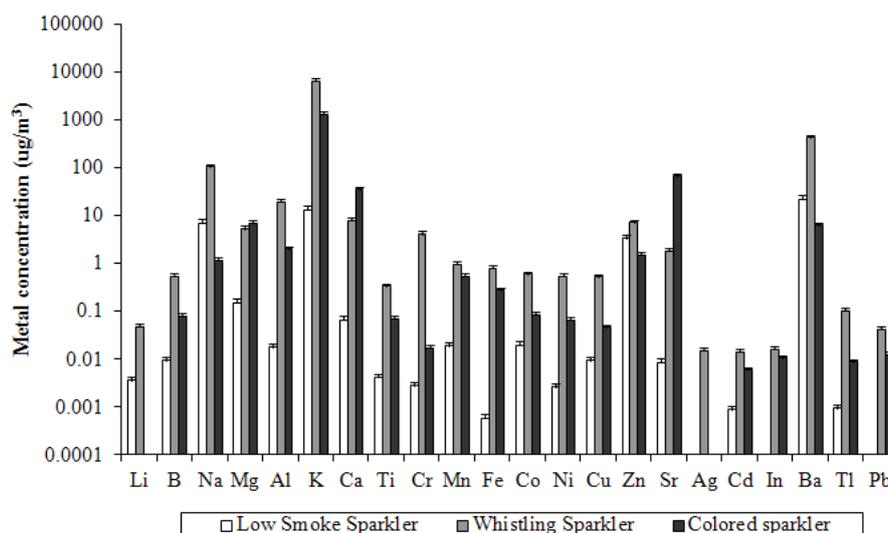
Ultimately, the impact of heavy metals on humans depends on their concentration levels, toxicity, and bioavailability, i.e., the extent to which they are absorbed by the blood or stored in internal organs. The toxicity and bioavailability of a metal depends in part on reactivity (size of particle) and solubility (chemical characterization) (Steinhauser *et al.*, 2006). Therefore a study of the solubility of the metals in sparkler emissions is crucial (Brown *et al.*, 1999).

From Fig. 3 it can be seen that K, Ba and Na have the highest water soluble concentrations for all three sparklers, which can be attributed to the highly soluble characteristics of these metals. The bioavailability of Ba is of concern as it is known to cause bronchoconstrictor effects and interfere with the heartbeat, and it is also suspected to be responsible for a significant increase in asthma cases during celebrations in India (Murti, 2000). Ba derived from pyrotechnics is more soluble than that from the traditional anthropogenic source, erosion of brake liner in automobiles, due to the presence of water soluble species such as  $\text{BaCl}_2$ , BaO and  $\text{Ba(OH)}_2$  in pyrotechnics compared to barite in brake liners.

In order to assess the bioavailability of the metals, it is important to assess the fraction of the total metals that is dissolved. WS have shown highest solubility for Li at 72.5%, Al at 8.7%, Cr at 10.2%, Cu at 2% and In at 55% of total metal concentrations. The CS has shown the highest percentages for Ca at 20.5%, Ti at 31.7%, Mn at 31.5%, Fe at 0.6%, Co at 20.3%, Ni at 94.2%, Cd at 54.1% and Pb at 1% while LSS shows the highest solubility for B at 81.8%, Na at 44.4%, Mg at 30.4%, K at 44.5%, Zn at 9.8%, Sr at 54.3%, Ba at 31.1% and Tl at 90.6% as compared to the other sparklers. The solubility of these metals in sparkler emissions is different from that of ambient particulate matter which could be related to the complex matrix contained in sparklers and the chemical forms in which they appear.



**Fig. 2.** Total metal concentrations of particles emitted from combustion of whistling sparklers (WS), Colored sparklers (CS), and Low smoke sparklers (LSS).



**Fig. 3.** Water soluble metal concentrations of particles emitted from combustion of whistling sparklers (WS), Colored sparklers (CS), and Low smoke sparklers (LSS).

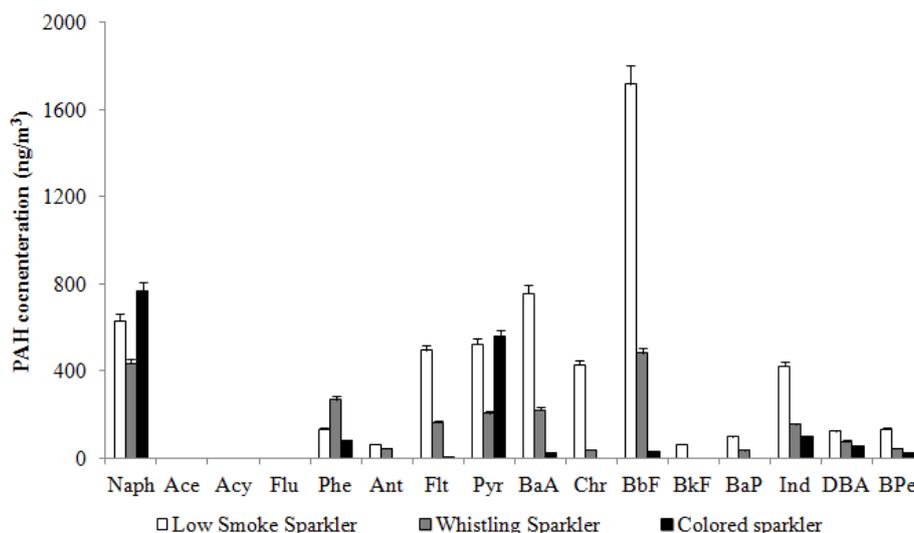
Similar to the trends of total metal concentrations, WS had the highest concentrations of water soluble metals among the three types of sparklers while LSS showed the lowest concentrations of water soluble metals. Elements such as Ti, Sr, K, Mg, Na and B had high percentage solubility and showed a strong correlation between total metal and water soluble metal concentration. However, for some metals such as Ba and Cd, the water solubility of the metal varied between sparklers. The solubility of Cd in CS was observed to be 54% while it was only 3% and 13% in WS and LSS, respectively. Similarly, the solubility of Ba in LSS was around 31%, but it was only 17% and 2% in WS and CS, respectively. This difference in water-solubility of the same elements in different sparklers could be related to difference in the chemical composition of the pyrotechnic mixture, causing the elements such as Ba and Cd to form chemical compounds with different solubility characteristics.

#### **PAHs Concentration in Sparkler Emissions**

According to the GC-MS used in this study, 13 out of the 16 priority PAHs listed by the Agency for Toxic Substances and Disease Registry (ATSDR) were detected in one or more types of sparklers. PAHs are usually the by-products of incomplete combustion processes and the PAHs analyzed are known or suspected carcinogens. As can be seen from Fig. 4, by and large LSS emit the highest concentrations of PAHs with a significant level of heavier PAHs containing four to six rings such as BbF, BaA, Pyr, Chr, Flt and Ind. The latter PAHs are known to be highly carcinogenic. The dominant fraction of low molecular weight PAHs, those containing two to three rings (Naph, Ace, Acy, Flu, Phe, Ant), was naphthalene for all three sparklers. The higher concentration of PAHs in LSS may be due to incomplete combustion as they contain relatively lower amounts of oxidizer(s). The CS and WS may undergo a more complete combustion which makes them emit lower concentrations of PAHs compared to LSS. The dominant PAHs emitted by CS were Naph and Pyr, while those

emitted from WS were Naph and BbF.

Diagnostic ratios (DRs) of Ant/(Ant + Phe), Flt/(Flt + Py), BaA/(BaA + Chr), and Ind/(Ind + Bpe) for the three different types of sparklers were calculated based on average concentrations and are shown in Table 1 along with the DRs of other source types reported in the literature. These DRs can serve as markers for sparkler emissions since they are unique for different sources. It can be noted that the DRs of Flt/(Flt + Py) for LSS and WS are similar to those of PAHs for cooking, incense and candle emissions reported in literature. However, BaA/(BaA + Chr) and Ind/(Ind + Bpe) are higher in LSS and WS sparkler emissions compared to other sources. DRs could not be calculated for CS since some of the PAHs are below detection limit. It was reported by Yunker *et al.* (2002) that Ant/(Ant + Phe) < 0.1 indicates a source of petroleum origin (low temperature processes) while a ratio > 0.1 is indicative of combustion origin (high temperature processes). Similarly, BaA/(BaA + Chr) > 0.35 indicates combustion origin and a ratio of < 0.2 indicates petroleum origin, while the ratios between 0.2–0.35 could be either of petroleum or combustion origin. Ind/(Ind + Bpe) > 0.5 indicates sources of solid fuel combustion (such as coal, wood, grass, etc.) and ratios < 0.2 indicate petroleum origin, while the ratios between 0.2–0.5 indicate liquid fossil fuel combustion (Vehicle and crude oil) (Yunker *et al.*, 2002). In our study, we observed that Ant/(Ant + Phe) > 0.1, BaA/(BaA + Chr) > 0.35, Ind/(Ind + Bpe) > 0.5 are suggestive of particle emissions derived from high temperature processes (combustion). However, the ratio of Flt/(Flt + Py) < 0.5 observed in this study indicates that it may not be originated from high temperature processes (combustion of coal, wood, biomass etc.) entirely (Yunker *et al.*, 2002). Therefore, total index which is the sum of single indices normalized for the limit value (low temperature – high temperature sources) reported in literature was calculated as described in Mannino and Orecchio (2008). The total index is calculated in cases of ambiguity where values of DRs are not in agreement with each other.



**Fig. 4.** Particulate-bound PAHs emitted from combustion of whistling sparklers (WS), Colored sparklers (CS), and Low smoke sparklers (LSS).

**Table 1.** Diagnostic ratios of PAHs.

Source type	Ant/(Ant + Phe)	Flt/(Flt + Py)	BaA/(BaA + Chr)	Ind/(Ind + Bpe)	References
LSS	0.32	0.49	0.64	0.76	This study
WS	0.14	0.44	0.86	0.78	This study
Gas Cooking	0.04–0.06	0.51–0.53	0.28–0.31	0.44–0.54	See and Balasubramanian, 2008
Incense	0.17–0.87	0.41–0.58	0.38–0.51	0.53–0.67	Yang <i>et al.</i> , 2012
Candle	0.28	0.49	0.35	0.71	Orecchio, 2011
Fireworks		0.83		0.23	Sarkar <i>et al.</i> , 2010

Total Index =  $\text{Flt}/(\text{Flt} + \text{Py})/0.4 + \text{Ant}/(\text{Ant} + \text{Phe})/0.2 + \text{BaA}/(\text{BaA} + \text{Chr})/0.1 + \text{Ind}/(\text{Ind} + \text{Bpe})/0.5$ . When the total index is  $> 4$ , the PAHs mainly originated from high temperature processes, while  $< 4$  indicate low temperature processes. In our study, we observed that the total index of 10.7 and 12 for LSS and WS sparklers implies that sparklers burn through high temperature combustion processes.

#### Human Health Risk Assessment

The human health risk assessment when exposed to  $\text{PM}_{2.5}$  emissions from LSS, WS, and CS was carried out as described in the material and methods section. In general, the  $\text{PM}_{2.5}$  released into the ambient air gets dispersed and thus the concentrations are lower compared to the concentrations measured in the vicinity of their emissions. Such a scenario is widely applicable for aerial pyrotechnics, or for individuals who are at some distance away from the firework display locations. In the case of hand-held sparklers as described earlier, the individuals who are directly involved with fireworks undergo inhalation exposure to high concentrations of particulate-bound chemical species. However, there are no data available on dilution factors for an appropriate exposure assessment. Consequently, various dilution factors were considered for estimation of health risk assessment due to inhalation exposure including a worst case scenario with no or little dilution.

Typically, festive celebrations (e.g., Diwali) with sparklers last for 1 or 2 hours, so an exposure time of 2 hour per day

was assumed. An exposure frequency of 10 days per year with an assumption of more than 1 festive celebration taking place with the use of sparklers within a year, and an exposure duration of 70 years for adults and 6 years for children were considered. The averaging time used in this study to assess the lifetime cancer risk of both adults and children was therefore 613,200 h and 52,560 h, respectively. The ADAF used in this study was 3 for people with age ranging from 0 to 15 and 1 for people with age ranging from 16 to 70 (USEPA, 2005).

The ELCR due to inhalation of particulate-bound metals and PAHs emitted from three types of sparklers was estimated for both adults and children, and is shown in Tables 2 and 3, respectively for no dilution conditions. Cr (VI) is considered as a possible carcinogenic compound. However, due to lack of available data on Cr (VI) we used total Cr determined in our study to estimate ELCR. Thus some uncertainty exists in our ELCR estimation. Mean concentrations of carcinogenic PAHs and water soluble metals were used for estimation of ELCR. ELCR exceeded acceptable limits (1 in a million) for both WS and CS, but not for LSS in the case of adults for no dilution conditions, while it exceeded acceptable limits for all three types of sparklers (WS, CS and LSS) in the case of children. It can be seen from Tables 2 and 3 that both ELCR and HQ are higher for children compared to adults for all three types of sparklers implying that children are more prone to health effects compared to adults. Because of lower immunity

**Table 2.** Health risk estimates for water soluble metals and PAHs in PM<sub>2.5</sub> emissions from (a) LSS (b) WS (c) CS for adults (no dilution condition).

<b>(a) Low smoke sparklers</b>				
	CDI* (mg/kg/day)	PEF*	SF*	ELCR*
Heavy metals				
Cr	$8.5 \times 10^{-10}$		$4.2 \times 10^0$	$3.6 \times 10^{-9}$
Co	$6.0 \times 10^{-9}$		$3.1 \times 10^1$	$1.9 \times 10^{-7}$
Ni	$8.2 \times 10^{-10}$		$8.4 \times 10^1$	$6.9 \times 10^{-8}$
Cd	$2.7 \times 10^{-10}$		$6.3 \times 10^0$	$1.7 \times 10^{-9}$
PAHs				
Naph	$1.9 \times 10^{-7}$	0.001	$1.2 \times 10^{-1}$	$2.3 \times 10^{-8}$
BaA	$2.3 \times 10^{-7}$	0.1	$3.1 \times 10^{-1}$	$7.1 \times 10^{-8}$
BbF	$5.3 \times 10^{-7}$	0.1	$3.1 \times 10^{-1}$	$1.6 \times 10^{-7}$
BkF	$1.9 \times 10^{-8}$	0.1	$3.1 \times 10^{-2}$	$6.0 \times 10^{-10}$
BaP	$3.0 \times 10^{-8}$	1	$3.1 \times 10^0$	$9.1 \times 10^{-8}$
Ind	$1.3 \times 10^{-7}$	0.1	$3.1 \times 10^{-1}$	$4.0 \times 10^{-8}$
DBA	$3.8 \times 10^{-8}$	1	$3.1 \times 10^0$	$1.2 \times 10^{-7}$
				$\Sigma = 7.6 \times 10^{-7}$
<b>(b) Whistling sparklers</b>				
Heavy metals				
Cr	$1.2 \times 10^{-6}$		$4.2 \times 10^0$	$5.2 \times 10^{-6}$
Co	$1.8 \times 10^{-7}$		$3.1 \times 10^1$	$5.6 \times 10^{-6}$
Ni	$1.7 \times 10^{-7}$		$8.4 \times 10^1$	$1.4 \times 10^{-5}$
Cd	$4.3 \times 10^{-9}$		$6.3 \times 10^0$	$2.7 \times 10^{-8}$
PAHs				
Naph	$1.3 \times 10^{-7}$	0.001	$1.2 \times 10^{-1}$	$1.6 \times 10^{-8}$
BaA	$6.7 \times 10^{-8}$	0.1	$3.1 \times 10^{-1}$	$2.1 \times 10^{-8}$
BbF	$1.5 \times 10^{-7}$	0.1	$3.1 \times 10^{-1}$	$4.5 \times 10^{-8}$
BkF		0.1	$3.1 \times 10^{-2}$	
BaP	$1.0 \times 10^{-8}$	1	$3.1 \times 10^0$	$3.2 \times 10^{-8}$
Ind	$4.7 \times 10^{-8}$	0.1	$3.1 \times 10^{-1}$	$1.4 \times 10^{-8}$
DBA	$2.3 \times 10^{-8}$	1	$3.1 \times 10^0$	$7.2 \times 10^{-8}$
				$\Sigma = 2.5 \times 10^{-5}$
<b>(c) Colored sparkler</b>				
Heavy metals				
Cr	$5.3 \times 10^{-9}$		$4.2 \times 10^0$	$2.2 \times 10^{-8}$
Co	$2.6 \times 10^{-8}$		$3.1 \times 10^1$	$8.1 \times 10^{-7}$
Ni	$2.0 \times 10^{-8}$		$8.4 \times 10^1$	$1.7 \times 10^{-6}$
Cd	$1.8 \times 10^{-9}$		$6.3 \times 10^0$	$1.1 \times 10^{-8}$
Heavy PAHs				
Naph	$2.4 \times 10^{-7}$	0.001	$1.2 \times 10^{-1}$	$2.8 \times 10^{-8}$
BaA	$7.7 \times 10^{-9}$	0.1	$3.1 \times 10^{-1}$	$2.4 \times 10^{-9}$
BbF	$8.1 \times 10^{-9}$	0.1	$3.1 \times 10^{-1}$	$2.5 \times 10^{-9}$
BkF		0.1	$3.1 \times 10^{-2}$	
BaP		1	$3.1 \times 10^0$	
Ind	$2.9 \times 10^{-8}$	0.1	$3.1 \times 10^{-1}$	$9.0 \times 10^{-9}$
DBA	$1.7 \times 10^{-8}$	1	$3.1 \times 10^0$	$5.2 \times 10^{-8}$
				$\Sigma = 2.6 \times 10^{-6}$

\* PEF – Potency Equivalency Factors; CDI – Chronic daily intake; SF – Slope factor; ELCR – Excessive lifetime cancer risk

levels, lower body mass and less effective detoxification, the health outcome of the inhalation exposure to particulate emissions from different types of sparklers is more severe for children compared to that of adults. In addition, it was observed that for both CS and WS the carcinogenicity is mainly contributed by particulate-bound metals. Total ELCR due to particulate-bound elements was found to be  $2.5 \times$

$10^{-5}$  and  $2.5 \times 10^{-6}$  in the case of adults and  $7.4 \times 10^{-5}$  and  $7.6 \times 10^{-6}$  in the case of children for WS and CS, respectively while ELCR due to PAHs was negligible ( $2 \times 10^{-7}$  and  $9.4 \times 10^{-8}$  (for adults);  $6 \times 10^{-7}$  and  $2.8 \times 10^{-7}$  (for children) for WS and CS respectively) for both WS and CS. In the case of LSS, the carcinogenicity of PM<sub>2.5</sub> is mainly due to particulate-bound PAHs ( $5 \times 10^{-7}$  for adults and  $1.5 \times 10^{-6}$

**Table 3.** Health risk estimates for water soluble metals and PAHs in PM<sub>2.5</sub> emissions from (a) LSS (b) WS (c) CS for children (no dilution condition).

<b>(a) Low smoke sparklers</b>				
	CDI* (mg/kg/day)	PEF*	SF*	ELCR*
Heavy metals				
Cr	$6.0 \times 10^{-9}$		$4.2 \times 10^0$	$1.1 \times 10^{-8}$
Co	$4.2 \times 10^{-8}$		$3.1 \times 10^1$	$5.6 \times 10^{-7}$
Ni	$5.7 \times 10^{-9}$		$8.4 \times 10^1$	$2.1 \times 10^{-7}$
Cd	$1.9 \times 10^{-9}$		$6.3 \times 10^0$	$5.0 \times 10^{-9}$
PAHs				
Naph	$1.4 \times 10^{-6}$	0.001	$1.2 \times 10^{-1}$	$6.9 \times 10^{-8}$
BaA	$1.6 \times 10^{-6}$	0.1	$3.1 \times 10^{-1}$	$2.1 \times 10^{-7}$
BbF	$3.7 \times 10^{-6}$	0.1	$3.1 \times 10^{-1}$	$4.9 \times 10^{-7}$
BkF	$1.4 \times 10^{-7}$	0.1	$3.1 \times 10^{-2}$	$1.8 \times 10^{-9}$
BaP	$2.1 \times 10^{-7}$	1	$3.1 \times 10^0$	$2.7 \times 10^{-7}$
Ind	$9.0 \times 10^{-7}$	0.1	$3.1 \times 10^{-1}$	$1.2 \times 10^{-7}$
DBA	$2.6 \times 10^{-7}$	1	$3.1 \times 10^0$	$3.5 \times 10^{-7}$
				$\Sigma = 2.3 \times 10^{-6}$
<b>(b) Whistling sparklers</b>				
Heavy metals				
Cr	$8.7 \times 10^{-6}$		$4.2 \times 10^0$	$1.6 \times 10^{-5}$
Co	$1.3 \times 10^{-6}$		$3.1 \times 10^1$	$1.7 \times 10^{-5}$
Ni	$1.2 \times 10^{-6}$		$8.4 \times 10^1$	$4.2 \times 10^{-5}$
Cd	$3.0 \times 10^{-8}$		$6.3 \times 10^0$	$8.2 \times 10^{-8}$
PAHs				
Naph	$9.4 \times 10^{-7}$	0.001	$1.2 \times 10^{-1}$	$4.8 \times 10^{-8}$
BaA	$4.7 \times 10^{-7}$	0.1	$3.1 \times 10^{-1}$	$6.2 \times 10^{-8}$
BbF	$1.0 \times 10^{-6}$	0.1	$3.1 \times 10^{-1}$	$1.4 \times 10^{-7}$
BkF		0.1	$3.1 \times 10^{-2}$	
BaP	$7.3 \times 10^{-8}$	1	$3.1 \times 10^0$	$9.6 \times 10^{-8}$
Ind	$3.3 \times 10^{-7}$	0.1	$3.1 \times 10^{-1}$	$4.4 \times 10^{-8}$
DBA	$1.6 \times 10^{-7}$	1	$3.1 \times 10^0$	$2.2 \times 10^{-7}$
				$\Sigma = 7.5 \times 10^{-5}$
<b>(c) Colored sparkler</b>				
Heavy metals				
Cr	$3.7 \times 10^{-8}$		$4.2 \times 10^0$	$6.7 \times 10^{-8}$
Co	$1.8 \times 10^{-7}$		$3.1 \times 10^1$	$2.4 \times 10^{-6}$
Ni	$1.4 \times 10^{-7}$		$8.4 \times 10^1$	$5.1 \times 10^{-6}$
Cd	$1.3 \times 10^{-8}$		$6.3 \times 10^0$	$3.4 \times 10^{-8}$
PAHs				
Naph	$1.6 \times 10^{-6}$	0.001	$1.2 \times 10^{-1}$	$8.4 \times 10^{-8}$
BaA	$5.4 \times 10^{-8}$	0.1	$3.1 \times 10^{-1}$	$7.2 \times 10^{-9}$
BbF	$5.7 \times 10^{-8}$	0.1	$3.1 \times 10^{-1}$	$7.5 \times 10^{-9}$
BkF		0.1	$3.1 \times 10^{-2}$	
BaP		1	$3.1 \times 10^0$	
Ind	$2.0 \times 10^{-7}$	0.1	$3.1 \times 10^{-1}$	$2.7 \times 10^{-8}$
DBA	$1.2 \times 10^{-7}$	1	$3.1 \times 10^0$	$1.6 \times 10^{-7}$
				$\Sigma = 7.9 \times 10^{-6}$

\* PEF – Potency Equivalency Factors; CDI – Chronic daily intake; SF – Slope factor; ELCR – Excessive lifetime cancer risk

for children). This health risk analysis suggests that although LSS generates less metallic particles compared to WS and CS, its health risk is mainly due to higher PAH emissions.

It should be noted the health risk estimates were made for the worst case scenario assuming very little dilution of particulate emissions from sparklers. Health risk estimates for various dilution conditions (0–1,000) are summarized

in Table 4. Considering a dilution factor of 10 would lower the ELCR for both adults and children considerably, with no exceedance of the limits set by USEPA, for both LSS and CS. However, exposure to PM<sub>2.5</sub> emissions from WS still poses cancer risk for both adults and children. Increasing the atmospheric dilution factor further to 80 or above would reduce the cancer risk below the acceptable limits.

**Table 4.** ELCR estimated at various dilution conditions.

Dilution factor	Low smoke sparkler		Whistling sparkler		Colored sparkler	
	Adults	Children	Adults	Children	Adults	Children
0	$7.6 \times 10^{-7}$	$2.3 \times 10^{-6}$	$2.5 \times 10^{-5}$	$7.5 \times 10^{-5}$	$2.6 \times 10^{-6}$	$7.9 \times 10^{-6}$
10	$7.6 \times 10^{-8}$	$2.3 \times 10^{-7}$	$2.5 \times 10^{-6}$	$7.5 \times 10^{-6}$	$2.6 \times 10^{-7}$	$7.9 \times 10^{-7}$
80	$9.5 \times 10^{-9}$	$2.9 \times 10^{-8}$	$3.1 \times 10^{-7}$	$9.4 \times 10^{-7}$	$3.2 \times 10^{-8}$	$9.9 \times 10^{-8}$
100	$7.6 \times 10^{-9}$	$2.3 \times 10^{-8}$	$2.5 \times 10^{-7}$	$7.5 \times 10^{-7}$	$2.6 \times 10^{-8}$	$7.9 \times 10^{-8}$
1000	$7.6 \times 10^{-10}$	$2.3 \times 10^{-10}$	$2.5 \times 10^{-8}$	$7.5 \times 10^{-8}$	$2.6 \times 10^{-9}$	$7.9 \times 10^{-9}$

Therefore, the usage of these sparklers in well ventilated places can enhance the dispersion of sparkler emissions by the prevailing winds favorably and thus reduce the health risk associated with inhalation exposure to PM<sub>2.5</sub> released from sparklers.

## CONCLUSIONS

Three types of sparklers (LSS, WS and CS) were investigated for particulate bound metals and PAHs. Emissions from the sparklers have elevated levels of K (the dominant metal component of the firework propellant), Ca, Fe, Zn and Ba as well as a complex mixture of different trace metals and some heavy metals such as Cd, Cr, Co, Cu, Mn, Ni, and Pb, with WS having the highest concentration of metals and LSS with the lowest. The metals in the emissions were also tested for water solubility as it provides the bioavailable fraction of metals. WS and CS showed a higher degree of water soluble metals as compared to LSS. However, LSS have higher levels of particulate bound PAHs in its emissions compared to WS and CS. Significantly high concentrations of large molecule PAHs such as BbF, BaP and Ind, which are highly carcinogenic, are found in LSS emissions. Health risk estimates indicate that the emissions from WS have higher carcinogenic potential compared to the other two types of sparklers. Carcinogenicity of emissions from WS and CS was mainly due to metals, whereas in the case of LSS, PAHs make a major contribution to its carcinogenic properties. Particle emissions from sparklers were collected onto filters without dilution which could result in condensation of the semi-volatile species. Therefore the estimated concentrations for PAHs and some metals might be slightly higher leading to overestimation of health risk associated with particle inhalation.

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