EXPOSURE TO ATMOSPHERIC PMS, PAHS, PCDD/FS AND METALS NEAR AN OPEN AIR WASTE BURNING SITE IN BEIRUT

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ABSTRACT

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Since July 2015, Lebanon has experienced the worst solid waste management crisis in its history. Consequently, open-air waste burning in the vicinity of highly populated areas in Beirut has become a common practice. This study evaluates the effects of open-air dumping and burning on local air quality and public health. The levels of particulate matter (PM_{10}, $PM_{2.5}$), gaseous and particle-bound polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-dioxins and furans (PCDD/Fs), and particle-bound metals at a residence close to waste burning are reported. Concentrations, determined between October 2 and December 2, 2015, were compared either to previous measurements or to measurements taken away from a nearby burning incident, and after it had rained. Subsequently, the cancer risk due to exposure to these chemicals was assessed. Results showed alarming increases in pollutant concentrations which was translated into an increase in short-term cancer risk from about 1 to 20 people per million on the days when waste was being burned. Findings were shared with the public to warn the community against the dangers of waste mismanagement, and underline the obvious need for solid waste management at the governmental and municipal levels.

Keywords: dioxins, cancer and non-cancer risk, heavy metals, open waste burning

INTRODUCTION

Open dump waste burning is believed to largely contribute to air pollution worldwide. A recent study estimated that around 40% of the world's generated waste is treated by uncontrolled burning (Wiedinmyer *et al.*, 2014). In most developed countries, open waste

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burning is prohibited by law but still occurs at residential levels especially in rural areas (IPCC, 2006; Lemieux *et al.*, 2000; Quaß *et al*., 2004). Active environmental protection agencies (or similar bodies) have warned against these practices and have shared a wealth of information with the community in fact sheets and documents to emphasize the danger associated with waste burning activities (CIB, 2014; DEP, 2016; DNR, 2016; EPD, 2015; IDEM, 2016). In some developing countries, proper waste management practices are absent which results in increased open waste burning activities both at the residential areas and at dump sites.

Waste burning is known to emit a wide variety of atmospheric pollutants including, but not restricted to, greenhouse gases (CO_2, CH_4), sulfur dioxide (SO_2), nitrogen oxides (NO_x), volatile organic compounds (VOCs), particulate matter (PM), polycyclic aromatic hydrocarbons (PAHs), hydrochloric acid (HCl), hydrogen cyanide (HCN), hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), polychlorinated dibenzodioxins and polychlorinated dibenzofurans (PCDD/Fs) in addition to metals such as Hg, As and Pb (Akagi *et al.*, 2011; Estrellan & Iino, 2010; Lemieux *et al*., 2000; Lemieux *et al*., 2004; Park *et al.*, 2013; Solorzano-Ochoa *et al*., 2012; Wiedinmyer & Friedli, 2007). Thus, open waste burning largely influences air quality. A recent study in China reported that about 20% of China's anthropogenic PM_{10} emissions originate from open domestic burning (Wiedinmyer *et al*., 2014).

Waste burning in Lebanon has always been a common practice in rural areas. Farmers and villagers tend to burn tree pruning, leaves, crop residue and mostly organic household waste in their backyards. However, this practice has, since July 2015, expanded to urban areas especially in Beirut and Mount Lebanon in light of the solid waste management (SWM) crisis. This crisis began after Lebanon's main landfill, initially set up as a temporary dump site, was closed and the contract of the largest garbage-contracting company expired. Trash pick-ups were suspended leading to the accumulation of garbage piles at roadsides of commercial and residential areas. Although most attempts adopted by municipalities to manage the crisis were not environmentally sound, waste burning is considered one of worst approaches.

This study showcases the deterioration of the air quality in residential areas experiencing frequent waste burning episodes. Ambient concentrations of PM_{10}, $PM_{2.5}$, particle and gas phase PAHs and PCDD/Fs and particle bound metals were measured. The concentrations were compared either to concentrations taken before the crisis or to measurements on days that did not include waste burning activities at the same site. Inhalation cancer risk estimates from exposure to PAHs and PCDD/Fs and non-cancer hazard quotient from exposure to potentially toxic metals were calculated.

MATERIALS AND METHODS

Site description

Air sampling was conducted at the roof-top of a four story residential building near an open dump burning site in an area located east of Beirut, Lebanon. It is a residential area with small businesses and no heavy industrial activities. The sampling was conducted between October 2 and December 2, 2015 during a period that observed some days of street waste burning.

Sampling equipment

A DustTrak DRX Aerosol monitor Model 8533 from TSI was used to monitor real time PM_{10} and $PM_{2.5}$. The monitor was calibrated using two gravimetric samplers for PM_{10} and $PM_{2.5}$ (Thermo ChemComb 3500 speciation collection cartridges) to obtain the photometric and size calibration factors. The instrument was zero calibrated on a daily basis and data was logged every one minute.

Gas and particle phase organics (PAHs, PCDD/Fs) and particle bound metals were sampled using a DIGITEL DH77 high volume sampler (HVS) equipped with a PM_{10} single stage impactor. The HVS was operated at a flow rate of 500 L/min. The sampled air passed through the impactor head into a 150 mm filter holder loaded with a quartz filter (GE grade QM-A) used to capture the particulate phase organics and metals, then into a glass cartridge fitted with a Polyurethane Foam (PUF) used to capture gas phase organics. The quartz filter was prebaked at 400°C for 4 hours and the PUF plugs were pre-cleaned by soxhlet extraction for 16 hours. Sampling for PAHs and metals was conducted over a period of 24 hours while that of PCDD/Fs was done over a period of 48 hours. Sampling start time was at 8:00 pm.

Sample and data analysis

Data from the DustTrak was downloaded and checked for outliers. Ten-minute and 24-hour averages were computed and graphed using Igor wavemetrics software.

Sampling of dioxin, PAH and element content were conducted on different days. The analysis of each toxin family followed the procedures described below.

Dioxins

Seventeen 2,3,7,8-substituted PCDDs/F congeners were extracted from filters and PUFs and analyzed based on the US EPA TO-9 method. In brief, the quartz-fiber filter and the PUF were pre-cleaned with appropriate solvents and dried in a clean atmosphere. One of the PUFs was spiked with a surrogate $^{37}Cl_4$-2,3,7,8-TCDD standard, covered with Teflon® end caps, placed in a cleaned labeled shipping container, and tightly sealed with Teflon® tap to protect it form light until it is installed in the high-volume sampling. Post sampling, the filter and PUF are wrapped and brought back to the lab. Prior to extraction, both the filter and PUF were combined, spiked with $^{13}Cl_{12}$-labeled PCDD/PCDF internal standards, and Soxhlet extracted for 16 hours. The extract was subjected to an acid/base clean-up procedure followed by multitude clean-ups on silica gel, alumina, and carbon micro-columns. The extract was then concentrated for higher resolution gas chromatography-high resolution mass spectroscopy (HRGC-HRMS) analysis.

The loss during sampling is demonstrated by the determination of $^{37}Cl_4$-2,3,7,8-TCDD, which was spiked into one sample prior to deploying the trap and was found to be minimal. Quantification of the native 2,3,7,8-substituted analytes is based on isotopic dilution, which automatically corrects for variation in extraction efficiency and provides accurate values. Samples that had recoveries of labeled standards outside the acceptance limits were flagged. For all reported data, the signal-to-noise ratios were greater than 10:1 and detection limits were below the Method Reporting Limit (MRL). Detection limits are calculated for each analyte in each sample by measuring the height of the noise level for each quantitation ion and for the associated labeled standard. The concentration equivalent to 2.5 times the height of the noise is then calculated using the appropriate response factor and the weight of the sample.

Polycyclic aromatic hydrocarbons (PAHs)

Sixteen priority PAHs were extracted using a modified US EPA TO-13 method. In brief, the quartz filter and the PUF, which were cleaned as described in the previous paragraph, were extracted using 300 ml of diethylether/hexane (10 percent v/v) then refluxed for 18 hours using a soxhlet extractor setup. The extract was concentrated to 5 mL by rotary evaporator and afterwards further concentrated to 1mL under nitrogen flow. The concentrated extract was then loaded onto a preconditioned SPE cartridge (thermo scientific) and eluted with 10 mL of hexane. The eluent was concentrated to 1 mL and transferred to amber vials for gas chromatography-mass spectrometry (GC-MS) analysis.

A Thermo-Finnigan Trace GC-2000 Polaris QMS equipped with AI 3000 auto-sampler was used in this study. Chromatographic separation was achieved with a Thermo TG-5MS column (30m × 0.25mm, 0.25 µm film thickness) using helium gas as the carrier phase at a constant linear gas flow of 1 mL/min. Splitless injection of 1 µL volume was utilized and the injector temperature was set at 280 °C. The temperature program of the oven was as follows: 80 °C for 4 min, 10 °C/min to 170 °C hold for 1 min, 3 °C/min to 180 °C, 10 °C/min to 270 °C, and 3 °C/min to 300 °C hold for 3 min.

The mass spectrometer was operated under full scan mode. External calibration curve was used for the quantification of each of the 16 PAHs with standard mixtures of PAHs purchased from Absolute Standards. The overall recovery of each PAH ranged between 65 and 128%. Repeatability was determined by running six consecutive standard solutions of 2 ppm each during the same day. Results showed that the relative standard deviation was around 1.4 %. The limit of detection (LOD) of each PAH was calculated as three times the standard deviation of seven replicates of low concentration standards. The estimated LOD ranged between 8 and 38 pg/m^3.

Elements

Total elemental mass of the filter substrates was measured using a Thermo-Finnigan high-resolution magnetic sector and Inductively Coupled Plasma Mass Spectrometry (ICP-MS). A microwave-aided Teflon bomb digestion protocol using a mixture of 1 mL of 16 M nitric acid, 0.25 mL of 12 M hydrochloric acid, and 0.10 ml of hydrofluoric acid was used for extraction of total metals and elements (Herner *et al*., 2006).

50 elements and associated uncertainties were corrected for the filter-blank. Despite the fact that the quartz filter blanks for certain elements are high (as expected), the impact on the primary elements of concern was found to be minimal. In all cases, the filter blank uncertainty has been propagated with the analytical uncertainty to constitute the overall uncertainty in the reported data.

RESULTS AND DISCUSSION

PM measurements

Real time PM measurements were conducted between October 5 and 22, 2015. Daily averages for $PM_{2.5}$ (PM of aerodynamic diameter below 2.5 µm) ranged between 14.2 and 67.8 µg/m^3 (Fig. 1). The WHO set 24-hour guideline value of 25 µg/m^3 was exceeded on five days out of eight. Daily averages for PM_{10} (PM of aerodynamic diameter between 2.5 – 10 µm) ranged between 70.8 and 187.8 µg/m^3 (Fig. 1) exceeding the 24-hour WHO set guideline value of 50 µg/m^3 at all times (WHO, 2005). Short term exposure to such high concentrations of PM might be associated with increased hospital admission due to respiratory problems and premature mortality. An 8 year-long study in six U.S. cities have estimated a 1.5% increase in total mortality when the two-day $PM_{2.5}$ mean increase by 10 µg/m^3 (Schwartz *et al*., 1996). Also, long term exposure to high concentrations of PM has been associated with increase in lung cancer, cardiovascular diseases, cardiopulmonary diseases and morbidity. Pope et al. (2004) revealed that, for long term exposure, a 10 µg/m^3 elevation in $PM_{2.5}$ is associated with 8 to 18 % increase in mortality risk.

The ratio of PM_{10} to $PM_{2.5}$ ranged between 2.3 and 5 showing a prevalence of PM_{10} over $PM_{2.5}$. This shift towards higher particles in the PM size distribution confirms waste burning releases of white and black smoke that is enriched with bigger sized particles.

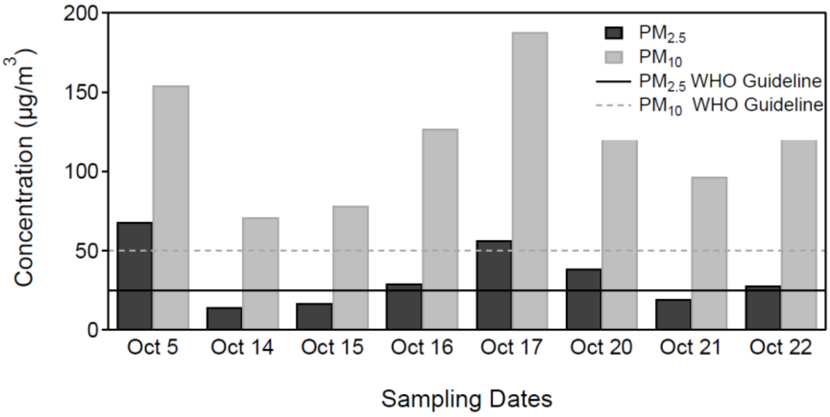

Figure 1. Twenty-four hour averages of $PM_{2.5}$ and PM_{10} during waste burning activities.

Diurnal variation in both $PM_{2.5}$ and PM_{10} showed similar behavior for all days of sampling except for October 5 and 17, 2015. During these days spikes in $PM_{2.5}$ and PM_{10} were recorded between 12:00 and 8:00 am. Ten minute averages of $PM_{2.5}$ and PM_{10} reached as high as 665 and 1126 µg/m^3, respectively, on October 5, and as high as 356 and 778 µg/m^3, respectively, on October 17, 2015 (Fig. 2 and 3).

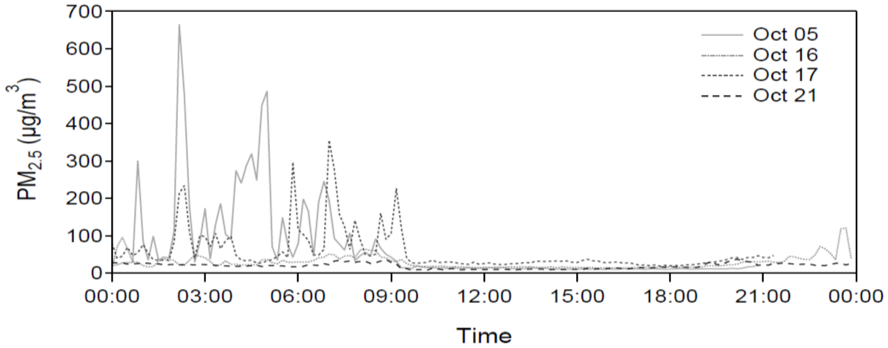

Figure 2. Diurnal variation of $PM_{2.5}$ mass concentration (µg/m^3) for designated days of waste burning.

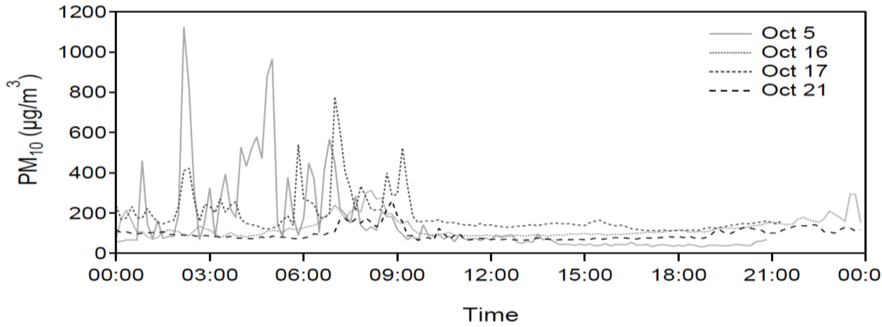

Figure 3. Diurnal variation of PM_{10} mass concentration (µg/m^3) for designated days of waste burning.

Polycyclic aromatic hydrocarbons

Four samples were collected for PAH analysis using HVS. The first three samples were taken on October 21, 30 and November 15, 2015, days of waste burning activities, while the sample taken on December 2, 2015, was after a rainy period and no waste burning activities. It is important to remind the reader here that the sampling on each day started at 8:00 pm and thus the bulk of the collected sample was actually taken on the day that follows. During waste burning, the total concentration of 16 measured PAHs averaged at 55 ± 19 ng/m^3 compared to a concentration of 24.1 ng/m^3 measured on December 2. On November 15, 2015, the sum of the 16 PAHs was 76.7 ng/m^3; a 218 % increase over the "no-burning" day. Individual PAH concentrations are shown in Fig. 4. Markers of incineration activities; Pyrene (PYR), Fluoranthene (FLT) and Phenanthrene (PHN), (Ravindra *et al*., 2006; Smith & Harrison, 1998) were prevalent during the waste burning days. Benzo[a]pyrene (BaP) and Dibenz[a,h]anthracene (DBA), which are considered as the most potent carcinogens among PAHs, averaged at 1.5 ± 0.9 ng/m^3 and 0.6 ± 0.5 ng/m^3, respectively. In comparison to the "no-burning" day, BaP measured concentration during waste burning days was 2.3 times higher and DBA was not detected when no waste burning activities were present.

Toxic Equivalents (TEQs) were used to report the toxicity-weighted masses of the 16 measured PAHs. The TEQs were calculated by multiplying the concentration of each PAH compound by its Toxic Equivalency Factor (TEF). BaP and DBA, which are the most toxic among the 16 PAHS, are assigned a TEF of 1. Other PAHs are assigned a factor ranging between 0.001 and 0.1 based on their toxicity relative to that of BaP. Endorsed by the World Health Organization (WHO), TEFs are widely used to facilitate risk assessment and regulatory control (Berg *et al*., 1998; Safe, 1998). Following, the concentration of PAH in air will be expressed in Bap equivalents calculated as the sum of TEQs.

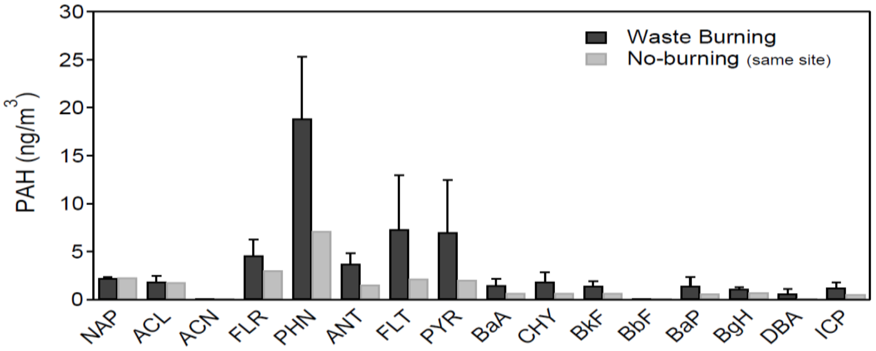

Figure 4. Concentrations of 16 PAHs during waste burning and during "no-burning" days. Error bars represent the standard deviation of 3 samples. NAP (naphthalene), ACL (Acenaphtylene), ACN (Acenaphtene), FLR (Fluorene), PHN (Phenanthrene), ANT (Anthracene), FLT (Fluoranthene), PYR (Pyrene), BaA (benzo[a]anthracene), CHY (Chrysene), BkF(Benzo[k]fluoranthene), BbF (Benzo[b]fluoranthene), BaP (Benzo[a]pyrene), BgH (Benzo[g,h,i]perylene), DBA (Dibenz[a,h]anthracene) and ICP (Indeno[1,2,3-cd]pyrene).

The inhalation dose ($Dose_{air}$) of PAHs was then calculated in accordance with the California Office of Environmental Health Hazard Assessment (OEHHA) air toxics hotspots program (OEHHA, 2015) based on equation 1 (Eq. 1).

$$Dose_{air} = C_{air} \times \{BR/BW\} \times A \times EF \times 10^{-6} \quad \text{(Eq. 1)}$$

Where,
C_{air}: Concentration of the compound of interest in ambient air (µg/m^3) (sum of TEQs)
BR/BW: daily Breathing Rate normalized to Body Weight (l/kg body weight *x* day)
A: inhalation Absorption factor (equal 1 assuming 100% absorption)
EF: Exposure Frequency (unitless) (350 days per 365 days for a resident)
10^{-6}: Factor used to convert µg to mg and l to m^3.

In reference to OEHHA, the daily breathing rates considered for different age groups were 225, 658, 535, 452, 210 and 185 l/kg day for 3rd trimester, 0-2 years, 2-9 years, 2-16 years, 16-30 years and 16-70 years, respectively.

Further, the residential inhalation cancer risk ($Risk_{inh\text{-}res}$) was calculated by multiplying the inhalation dose by the BaP cancer potency factor (CPF), the age sensitivity factor (ASF), the exposure duration (ED) averaged by 70 years of lifetime (AT), and the fraction of time spent at home (FAH) according to equation 2 (Eq. 2).

$$Risk_{inh-res} = Dose_{air} \times CPF \times ASF \times ED/AT \times FAH \qquad \text{(Eq.2)}$$

The CPF of BaP was considered to be 3.9 (kg day/mg) according to OEHHA recommendations (OEHHA, 2009). The age sensitivity factors used were 10 for 3rd trimester-2 years, 3 for 2-16 years and 1 for 16-70 years (OEHHA, 2009).

Inhalation cancer risks were calculated for each age group and were summed to estimate the cancer risk for 2 years of exposure ($Risk_{\text{3rd trimester}} + Risk_{\text{0-2 years}}$), 9 years of exposure ($Risk_{\text{3rd trimester}} + Risk_{\text{0-2 years}} + Risk_{\text{2-9 years}}$), 30 years of exposure ($Risk_{\text{3rd trimester}} + Risk_{\text{0-2 years}} + Risk_{\text{2-16 years}} + Risk_{\text{16-30years}}$) and 70 years of exposure ($Risk_{\text{3rd trimester}} + Risk_{\text{0-2 years}} + Risk_{\text{2-16 years}} + Risk_{\text{16-70years}}$). It is important to note that these calculations are associated with a degree of uncertainty especially upon extrapolation of long-period cancer risk estimates based on short-term measurements. Thus, inhalation cancer risks for shorter periods of exposure (0-2 years) are of higher confidence than those calculated for longer periods. Nevertheless, calculations for long periods were made to showcase the effect of exposure to these concentrations for longer periods. Moreover, these calculations represent low limit estimation because they do not account for any source or means of exposure other than inhalation.

By multiplying the $Risk_{inh\text{-}res}$ by 10^6, the estimated probability of risk can be converted to chances of developing cancer per million people. During waste burning days, the cancer risk estimates were highest on November 15 (Table 1).

TABLE 1

Per Million Cancer Risk Estimates from Exposure to PAHs

Population	**Sampling Days**			
	Oct 21	**Oct 30**	**Nov 15**	**No burning**
Per million cancer risk estimates (2 years)	1	1	2	1
Per million cancer risk estimates (9 years)	3	2	3	1
Per million cancer risk estimates (30 years)	3	3	5	1
Per million cancer risk estimates (70 years)	4	4	6	2

Polychlorinated dibenzo dioxins and furans

Atmospheric concentrations for 17 toxic congeners of 2,3,7,8-substituted, tetra- to octa-chlorinated dibenzo-p-dioxins (TCDD) and dibenzofurans (PCDD/Fs) were measured on October 2, 13, 15 and 19, 2015. The sum of the 17 PCDD/Fs ranged between 1.0 and 9.2 pg/m^3. The levels measured during this campaign were 3.2 to 53.5 times higher than previous measurements taken in an industrial zone in Lebanon (data not shown). It is worth noting that this is the first time that all 17 congeners are detected in all samples (except for 2,3,7,8, TCDD on October 15) and that most toxic PCDD, 2,3,7,8 TCDD, reached a concentration as high as 2.6 pg/m^3 on October 19 while it was never detected in previous measurements (Fig. 5).

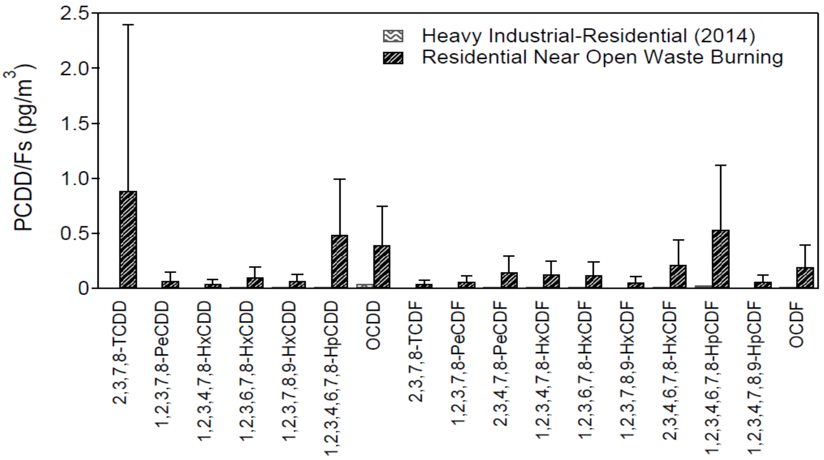

Figure 5. Concentrations of the 17 congeners of dioxins and furans during waste burning. Measurements are compared to measurements taken in 2014. Error bars represent the standard deviation of 4 samples.

Toxic equivalents, daily inhalation dose, and cancer risk estimates were calculated for PCDD/Fs following the same procedure used for PAHs using PCDD/Fs TEFs and a cancer slope factor of 1.3×10^5 (kg *x* day *x* mg^{-1}) for 2,3,7,8-TCDD (OEHHA, 2005). Calculated inhalation cancer risk estimates for 2, 9, 30 and 70 years of exposure are shown in Table 2.

TABLE 2

TEQs (pg×10^3/m^3), and Cancer Risk Estimates Calculated for 2, 9, 30 and 70 Years of Exposure to 17 PCDD/Fs

	Oct 2	**Oct 13**	**Oct 15**	**Oct 19**	**Industrial Site**
Σ TEQ (pg×10^3/m^3)	73.3	34.4	192.4	3133.8	6.5
Per million cancer risk estimates (2 years)	2	1	4	65	0
Per million cancer risk estimates (9 years)	3	1	7	110	0
Per million cancer risk estimates (30 years)	4	2	9	154	0
Per million cancer risk estimates (70 years)	4	2	11	182	0

Particle-bound metals

Three samples taken on Nov 19, 25 and 26 (days having waste burning activities) were analyzed for metal content and compared to concentrations measured on December 1, a day where no burning activities were present and after rain. A total of 50 metals and elements were measured using ICP-MS. Typically, crustal elements (Ca, Al, Fe, Mg and K), S and Na were the most prevalent elements in all samples. Both crustal metals and other metals showed an increase during waste burning (Fig. 6). This increase ranged between 65 and 1448%. A comparison between the levels of potentially toxic elements (e.g., Pb, Sn, Ni, Cr, V, and Ba) during days of waste burning and the "no-burning" day is presented in Fig. 6. The health impact of these potentially toxic metals was assessed by calculating each metal's Hazard Quotient (HQ) (Eq. 3).

$$\text{Hazard Quotient} = \frac{\text{Exposure concentration } (\mu g/m^3)}{\text{Inhalation REL } (\mu g/m^3)} \qquad \text{(Eq. 3)}$$

where REL is the reference exposure level adopted by OEHHA for each metal (OEHHA, 2014).

Calculated HQs for the different metals were below 1, indicating that the adverse non-cancer health effects of these concentrations were not alarming.

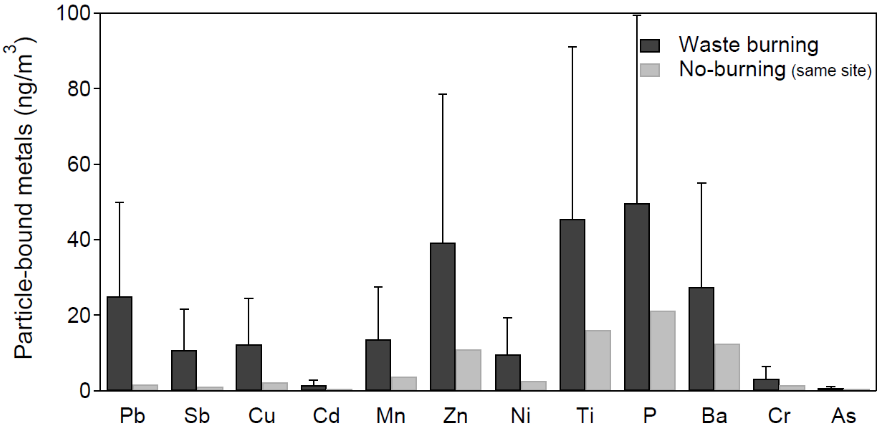

Figure 6. Average 24 hour concentrations of selected particle-bound metals during waste burning and no-burning days.

CONCLUSION

Open dump waste burning in the vicinity of highly populated areas has recently been practiced in Lebanon in the wake of the solid waste management crisis in the country. This is the first study in Lebanon that identifies the most harmful gas and particle bound chemical constituents. It involves PM and gas phase collection, chemical analysis and risk assessment of chemicals captured at a residence close to the waste burning fire.

Results show alarming levels of PM_{10} and $PM_{2.5}$. Spikes reaching up to 1126 and 665 µg/m^3 for PM_{10} and $PM_{2.5}$, respectively, were recorded on Oct 5 and 17. Particle and gas phase concentrations of PAHs and PCDD/Fs have also increased drastically during waste burning days. Combined short-term (2 years) incremental cancer risk from exposure to PAHs and PCDD/Fs averaged at 20 cancer cases per million people while it was calculated to be 1 cancer case per million people when no waste burning activities were carried out. Very alarming levels of PCDD/Fs were measured in the sample of October 19. This translated in an increase of short-term (2 years) incremental cancer risk to 65 cases per million people solely due to inhalation of PCDD/Fs. Particle-bound metals have also shown a strong increase during waste burning days but levels remain below alarming thresholds.

The presence of an immediate health risk was shared with the public via social media and local radios and televisions. In the absence of a governmental body able to properly manage solid waste, it was crucial to mobilize active citizens to stop waste burning in the highly populated residential areas.

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