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Chemical Characterization and Behavior of Respirable Fractions of Indoor Dusts Collected Near a Landfill Facility

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ABSTRACT

The study aims to determine the inorganic and organic phases in airborne particulate matter (PM) collected near a landfill facility. The establishments within the vicinity of the landfill considered in the study were a junk shop, a school, and a money changer shop. From the elemental analysis using inductively-coupled plasma mass spectrometry (ICP-MS), lead and cadmium were discovered to be more abundant in the total suspended particulate (TSP) fraction, whereas copper was more abundant in the smaller $PM_{2.5}$. Manganese, arsenic, strontium, cadmium, and lead were more abundant in the PM_{10} fraction than in $PM_{2.5}$. The results of the chemical characterization were compiled and evaluated in a geochemical modelling code (PHREEQC) to determine the potential speciation of these chemical constituents. Solution complexes of As, Pb, Cd and phthalates, and metal species, such as H_2AsO^{3-} , Cd_2OH^{3+}, $Pb(OH)^{3-}$, were predicted to form by the PHREEQC simulation runs once the end-member components interact with water. The results contribute to the background information on the potential impacts from exposure to airborne PM at workplaces around landfill facilities. Moreover, the data gathered provide a baseline for the chemical characterization and behavior of chemical constituents of PM possibly present in this specific type of environment.

Keywords: Airborne particulate matter, landfill facility, indoor air quality, solution complexes, PHREEQC speciation, respirable fractions

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INTRODUCTION

Air pollution is almost always considered ambient. However, humans are estimated to spend 70-90% of their time indoors, thus they tend to be more exposed to pollutants present indoors (Raunemaa et al. 1989). The World Health Organization reports that 2 million premature deaths per year are attributed to exposure to indoor air pollution. Indoor air pollution also accounts for up to 4.0% of the burden of disease for low-income countries (World Health Organization [date unknown]). Pollutants may come from combustion sources indoors (e.g. cook stoves), building materials and furnishings, household consumer products, appliances, or outdoor sources (Lamuth 2008). In the Philippines, a number of studies regarding ambient air quality have been performed in the past (Bautista et al. 2014; Pabroa et al. 2011). However, local studies on the characterization of indoor air aerosols and their airborne behavior are still very limited. Published data regarding the exposure of Filipino workers to indoor particulate matter (PM), particularly in the inhalable and respirable mass fractions, are sparse.

One of the most abundant pollutants indoors is PM. PM is a mixture of small particles and liquid droplets. The small particles in PM may be made up of several components, such as acids, organic chemicals, dust particles, soil, or metals (US EPA [date unknown]). PM is generally classified based on its size, sampling methodology, and cut-off point of the sampler used for its collection. For example, PM with a diameter of 10 μm is called PM_{10}, whereas PM with a diameter of 2.5 μm is called $PM_{2.5}$. PM, especially the smaller size fractions, is a health concern because of its capability to enter the respiratory system and cause health issues, such as chronic obstructive pulmonary diseases and reduced lung function for children and adults (W.H.O. Regional Office for Europe 2003). Furthermore, toxic metals in PM have been proven to cause acute inflammatory responses. Water-soluble metals, on the other hand, have been observed to affect the cardiopulmonary system (Costa and Dreher 1997). Hence, it is of importance to be able to characterize and identify the toxic metals present in PM.

Some sites of concern with respect to indoor air pollution are plastic/electronic recycling/dismantling centers near landfill facilities. These facilities collect and sort recyclable wastes as profitable businesses that flourish as an unorganized sector. Due to unsupervised and uncontrolled practices, improper handling techniques may be prevalent in such facilities. These improper handling techniques can bring about serious health risks not only to workers in such facilities but also to local residents. Most of these facilities situated in Metro Manila lack proper equipment to control or diminish pollutant emissions. Hence, the characterization of pollutants in such complex microenvironments is a crucial first step in preparing a comprehensive methodology for defining acceptable indoor air quality in occupational settings in Metro Manila. The flourishing community near Payatas Dumpsite, which is the main controlled and organized waste disposal site of Quezon City, is an appropriate place to initiate the study.

The study aims to determine the inorganic and organic phases in airborne PM collected near a landfill facility. The results of the chemical characterization were compiled and evaluated in a geochemical modelling code to determine the potential speciation of these chemical constituents. Several establishments within the vicinity of the landfill site were considered in the study. The indoor areas of a junk shop, a school, and money changer shop were selected as the sampling sites. The study will provide data on the profiles of total suspended particulates, elemental species, and organic phases in airborne PM.

MATERIALS AND METHODS

Preliminary Studies

To assess the presence of toxic metals in indoor air, preliminary analyses of ordinary house dust using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and Scanning Electron Microscopy/Energy Dispersive X-ray Fluorescence (SEM/EDX) were performed. Samples were collected from tabletop surfaces and air conditioning systems in different residential and commercial establishments, digested using hot acid (HNO_3/H_2O_2 system), and then subjected to ICP-MS (CalEPA 2007). The remaining undigested samples were subjected to SEM/EDX.

Indoor Airborne PM Sampling

For the landfill samples, PM was collected on Teflon filters using the Airmetrics Minivol sampler with a flow rate of 5 L/min. Three fractions were separately collected ($PM_{2.5}$, PM_{10}, and TSP) due to the limitation of the sampler (i.e. only one fraction can be collected at a time). At least three trials per fraction and location were performed. No TSP fraction was collected from the money changer shop. The sampling was conducted for only three hours due to personnel security, from 10 am to 1 pm, and 1 pm to 4 pm. Sampling was performed in three sites, namely a junk shop, a school, and a money changer shop. The sites were chosen based on their 1) distances from the landfill and 2) sampler and personnel security. Field and instrument filter blanks were also obtained prior to each sampling. The filters were weighed using a Sartorius ME5-F microbalance with a minimum resolution of 0.001 mg. Mass concentrations are reported in $\mu g/m^3$, which was calculated using equation (1). After weighing, the filters were halved for inorganic and organic phase characterization.

$$\text{conc in } \mu g/m^3 = \frac{\text{weight of sample in } \mu g}{\text{volume of air sampled in filter, } m^3} \qquad (1)$$

Where volume of air sampled, m^3 = flow rate of Minivol length of sampling conversion factor

$$5\,\frac{L}{min} \times 3\text{ hr} \times 60\,\frac{min}{hr} \times \frac{0.001\text{ m}^3}{1\text{ L}} = 0.9\,m^3$$

Sample Analysis

Half of the Teflon filter was set aside for ICP-MS analysis, whereas the other half was further halved for other characterization techniques. One-fourth of the Teflon filter was subjected to ^{1}H-NMR characterization. The other one-fourth was stored for a morphological analysis using SEM-EDX (data will not be shown). Microwave-assisted acid digestion using Ethos closed-vessel microwave digester and 18.5% HNO_3 (4% final acid concentration after diluting to 50 mL and upon introduction to ICP-MS), following the method of Kulkarni et al. (2007), was employed as the sample preparation technique prior to elemental analysis using ICP-MS. Recovery studies by means of spiking 0.1, 0.2, 0.25, 0.5, 0.75, and 1 mL of 10 ppm multi-elemental standard into different Teflon filters and subjecting them to the same digestion method were performed as a check for the extraction method. Seven solutions containing 1 ppb of the multi-elemental standard were prepared and then subjected to ICP-MS analysis for the determination of the method detection limit. A 500-MHz Agilent NMR spectrometer was used for the ×^{1}H-NMR analysis. PM samples were dissolved in deuterated chloroform ($CDCl_3$) to obtain the spectra for the nonpolar compounds, while deuterated water (D_2O) was used to obtain the spectra for the polar compounds. However, due to the limitation of the NMR spectrometer, no quantification was performed for the organic components.

Geochemical Modelling

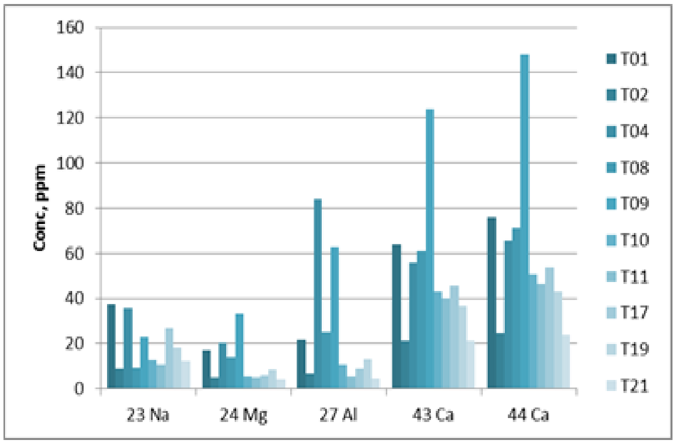
To model the possible speciation and reactions between water and toxic metals in PM, as well as the influence of organic phases on metal speciation, PHREEQC (pH-Redox-Equilibrium, written in C language), a program developed by the United States Geological Survey (USGS), was used. PHREEQC is designed to perform a wide variety of aqueous geochemical equations and is based on equilibrium chemistry of aqueous solutions interacting with minerals, gases, solid solutions, exchangers and sorption surfaces, and one-dimensional transport (Parkhurst and Appelo 2013). Model simulations performed in the study considered the formation of solution complexes and precipitation of solid phases. Measured parameters, such as pH, temperature, % humidity, and relative stoichiometry, were set as variables in different sets of reaction runs. All speciation calculations used the dissolved elemental concentrations (i.e. chemical analyses previously performed). Metals were indicated in their mineral phases and solution species form. Iso-phthalate containing aromatic and short aliphatic constituents was used as the representative for organic phases. The llnl.dat PHREEQC thermodynamic database supplied with the software was used for the analyses.

The first data block for the input statement describes a solution of water containing the elements Mn, As, Sr, Cd, and Pb. The temperature was also specified. Mole fractions of the elements, which were based on the total metal concentration measured with ICP-MS, were also included. The second data block describes the removal of water from the solution, in order to achieve the humidity conditions as measured. The last data block in the run simulates the reaction of an organic component, isophthalate, to the solution under the specified humidity condition. The concentration of isophthalate added was based on literature values since no quantification of the organic species was performed.

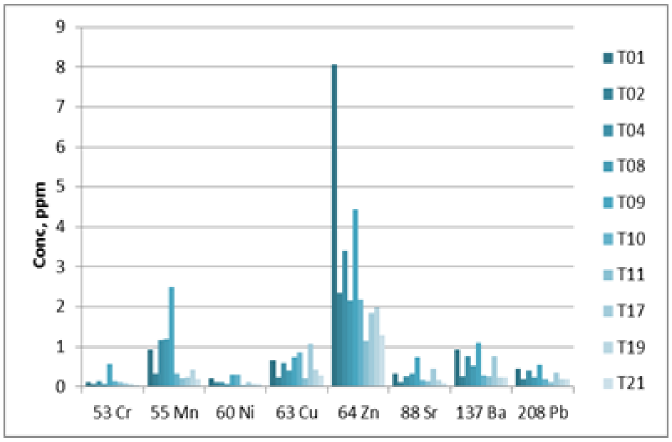
RESULTS AND DISCUSSION

Preliminary Studies

Preliminary results are shown in Figure 1. Among the elements found in the samples, the crustal/naturally found elements are Na, Mg, Al, Ca (Figure 1A), and Ba (Figure 1B). Meanwhile, the non-crustal elements, which are likely associated with anthropogenic activities, are Cr, Mn, Ni, Cu, Zn, Sr, and Pb (Figure 1B). Ca was the most abundant among the crustal elements. For the non-crustal elements, Zn concentration was the highest for all locations, followed by Mn, Cu, Ba, and Pb. Ni and Cr were present at low concentrations.



(A)



(B)

Figure 1. Total concentration of metals present in airborne PM. (A) Metals associated with crustal fraction and (B) trace metals. T01, T02, T04 – Residential-single-detached houses; T08, T09 – Laboratory; T10 – Restaurant; T11, T21 – Residential-Condominium; T17 – Dental Clinic; T19 – Residential.

No source apportionment was performed, thus the exact sources of the elements could be determined. However, high concentrations of Zn, in addition to the natural amounts present in the crust, are associated with the wear and tear of vulcanized vehicle tires and the corrosion of galvanic automobile parts (Wahab et al. 2012). Such could also the source of the Zn for the sampling sites since they were located within close range of some roads (Commonwealth Road and Katipunan Road). Similarly, Cu and Mn may originate from both natural and anthropogenic sources. Soil contains natural amounts of Mn and Cu, but anthropogenic sources, such as industrial processes (battery and electronics manufacturing, steel productions, welding and motor vehicle exhaust), that may be present nearby are also possible sources of these elements (Datta et al. 2012; Midander, 2006). In addition, Cu may also result from brake wear.

Pb may be part of the road dust as an element adsorbed and continuously resuspended in the upper layers of the soil. Anthropogenic sources of Pb, such as leaded gasoline, lead-based paint, and lead-arsenate pesticides, have long been eliminated from the industry; however, the presence of Pb in the environment may still be attributed to these sources due to the persistence of Pb. Other anthropogenic sources of Pb may also include the manufacture of lead-containing products, combustion of coal and oil, and waste incineration (ATSDR 2014). Ni, on the other hand, may also come from both natural and anthropogenic sources. The natural sources of Ni, which include the weathering of rocks and soils from volcanic eruptions, are unlikely reservoirs in the urban setting. Anthropogenic sources of Ni, such as industrial processes (combustion, incineration, metallurgical operations, Ni production, chemicals and catalyst manufacturing) may be more probable in the urban setting. In addition, mobile sources, such as engine wear and tear and impurities, and engine oil and fuel additives, may also be possible sources of Ni in an urban setting (Galbreath et al. 2003).

Possible sources of Cr may include stainless steel, paint pigments, and wood preservatives (Galarpe and Parilla 2014). Moreover, Cr is involved in many industrial processes, such as chromium plating and cement manufacture. It is also used as an additive to anti-corrosion coatings on vehicles (McSheehy 2008).

This preliminary study serves to measure the concentrations of metals in areas far from a landfill facility. Preliminary measurements show that metal concentration is significant even in locations distal from the landfill area. Based on these findings, analysis of the airborne PM at the landfill area is deemed important and necessary.

Description of the Main Sampling Sites

Payatas dumpsite, a 22-hectare open pit, is located in Quezon City, Philippines. It is the largest and oldest solid waste dumpsite in Metro Manila. Many material recovery facilities (junk shops) are also operational along the Payatas Road. A large community of families thrives near the dumpsite, thus facilities, such as schools, small stores, and small commercial establishments, are present within the vicinity of the site. The three chosen sampling sites are as follows: a junk shop, a school (around 1.6 km from the landfill), and a money changer shop (near Commonwealth road, a main highway in Quezon City). These sampling sites are situated along a busy road (i.e. which garbage trucks use to enter the landfill facility). Furnishing and fitting conditions, such as air-condition settings, room and ceiling-height sizes, and room content, were variable.

Variation in the Organic and Inorganic Components of PM

Table 1 summarizes the results obtained from the sampling, while Figure 2 shows a comparison of the weight distribution per PM fraction per location. From the data, it can be observed that the TSP fraction in the junk shop has the highest mass concentration, while the lowest mean for the $PM_{2.5}$ fraction was obtained from the elementary school. Moreover, the values obtained from the junk shop display a generally higher trend than the elementary school and money changer, possibly because it is not a fully enclosed (indoor) location. Thus, PM from outside sources (e.g. road dust, fugitive dusts) may have also contributed to the higher mass concentration. The mass concentration is directly proportional to size, because larger particles are heavier and have higher masses.

Numerical values obtained from the study, as shown in Table 1, are larger compared to literature values for indoor air in a classroom in Munich during winter time (19.8 $\mu g/m^3$ $PM_{2.5}$ and 91.5 $\mu g/m^3$ PM_{10}) (Fromme et al. 2007). Moreover, the measured parameters are higher than those measured in a typical residential environment in Athens, where the mean 24-hour indoor PM_{10} concentration is 35 $\mu g/m^3$ during the warm period and 31.8 $\mu g/m^3$ during the cold period (Diapouli et al. 2011). Furthermore, since no guideline values have been set for indoor air PM, 2005 ambient air guidelines set by the World Health Organization were used to compare the values. The guideline values are 10 $\mu g/m^3$ (annual mean) and 25 $\mu g/m^3$ (24-hour mean) for $PM_{2.5}$, and 20 $\mu g/m^3$ (annual mean) and 50 $\mu g/m^3$ (24-hour mean) for PM_{10}. It should be of immediate concern that the values obtained from the landfill site are well above the guideline values. However, it is recommended that a longer sampling time be carried out, in order to be able to fully quantify the PM mass concentration for the selected sampling locations.

Table 1. Three-hour averages for PM$_{2.5}$ and PM$_{10}$ in the three locations, namely the junk shop, elementary school, and the money changer shop.

	N	Mean	Min	Max
PM$_{2.5}$ (µg/m^3, 3-hour average, Junk shop)	3	70.19	57.78	76.67
PM$_{2.5}$ (µg/m^3, 3-hour average, Elem. School)	4	45.00	33.89	56.11
PM$_{2.5}$ (µg/m^3, 3-hour average, Money Changer)	4	77.08	60.00	102.22
PM$_{10}$ (µg/m^3, 3-hour average, Junk shop)	3	129.44	114.44	143.33
PM$_{10}$ (µg/m^3, 3-hour average, Elem. School)	4	126.25	98.89	176.11
PM$_{10}$ (µg/m^3, 3-hour average, Money Changer)	4	105.69	78.33	142.22
TSP (µg/m^3, 3-hour average, Junk shop)	4	209.72	187.78	247.22
TSP (µg/m^3, 3-hour average, Elem. School)	4	110.00	33.89	176.11

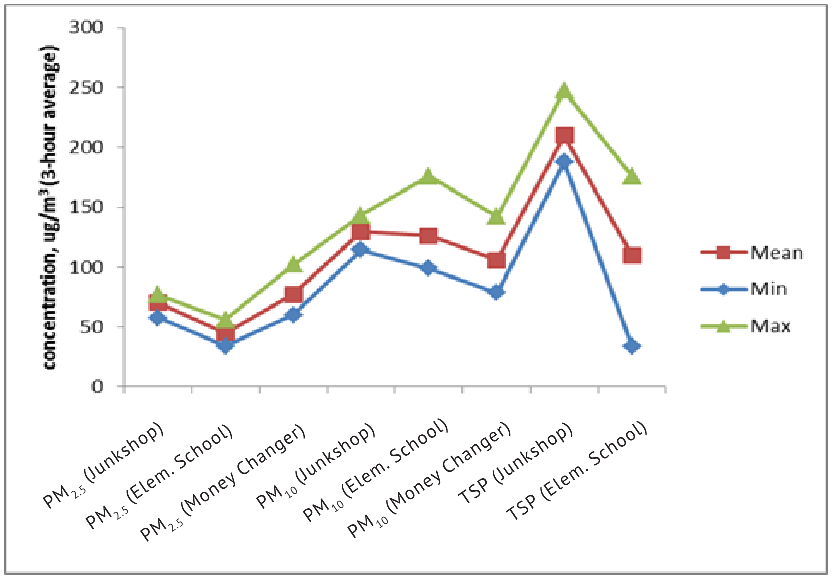


Figure 2. Mass concentration of PM fraction per location.

Method Validation (Closed-vessel microwave-assisted acid digestion)

To assess the accuracy of the method, recovery studies were performed. Percent recovery was calculated according to the following formula (US EPA 1994):

$$\% \text{ recovery} = (C_s - C)/S \times 100 \qquad (2)$$

where C_s = measured concentration of spiked sample

C = measured concentration of unspiked sample (background concentration)

S = theoretical concentration of the spiked sample

Upon the analysis of the spiked samples using ICP-MS, eight out of the 13 elements (manganese, cobalt, nickel, copper, arsenic, strontium, cadmium, and lead) analyzed have acceptable % recovery values (i.e. between 70-130%) for all the concentrations of the spike. To determine the method detection limit (MDL), equation (3) was used, and MDL values calculated for the elements were observed to have acceptable recoveries (US EPA 1994).

$$\text{MDL} = t \times S \qquad (3)$$

Where t = student's t-value at 99% confidence level (t_7=3.14)

S = standard deviation of the seven replicates

Inorganic and Organic Characterization of Airborne PM

Elemental analysis of the PM fractions allowed the determination of the dominant toxic metals on each fraction. Lead and cadmium were more abundant on the TSP fraction, whereas copper was more abundant on the smaller $PM_{2.5}$. On the other hand, manganese, arsenic, strontium, cadmium, and lead were also present on PM_{10} and were more abundant on this fraction than on $PM_{2.5}$. Figures 3-5 show the distribution of toxic metals for different size fractions of PM.

^{1}H-NMR analysis indicates the presence of organic constituents, such as hydrocarbons, from airborne PM. Spectra with similar peaks at 0.8-1.6 ppm and 7.0-7.5 ppm were obtained for all fractions and locations, signifying similar organic compositions for airborne PM present near landfill areas. Peaks found at 0.8-1.6 ppm indicate the presence of aliphatic groups for all the samples. An additional peak at around 4.7 ppm for the TSP fraction of the junk shop, which was also present in the PM_{10} fraction from the elementary school, may indicate the presence of some alcohols. Aromatic groups were also detected, as represented by peaks at 7.2-7.3 ppm (overlapping with the solvent peak, therefore not conclusive) (Figures 6A and 6B).

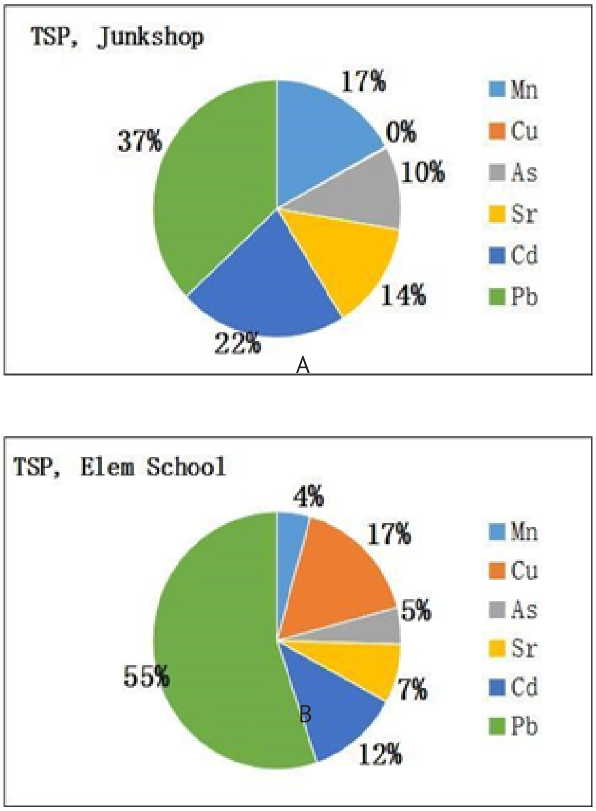
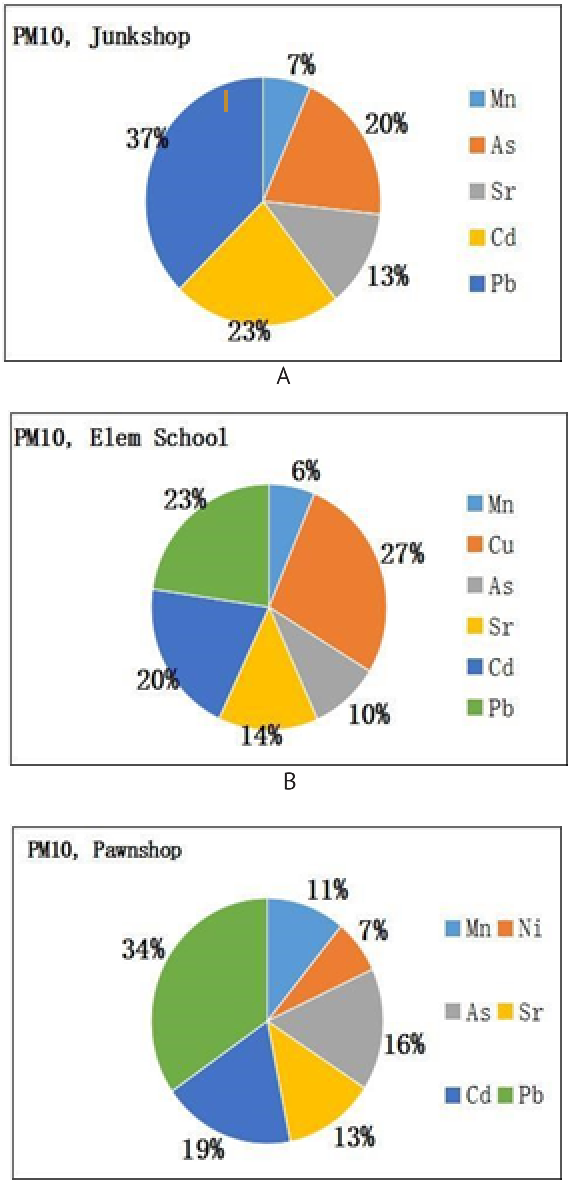


Figure 3. Metal concentrations in TSP fractions collected from sampling sites. (A) Junk shop and (B) elementary school. No TSP fractions were collected from the money changer shop.



A

B

C

Figure 4. Metal concentrations in PM_{10} fractions collected from sampling sites. (A) Junk shop, (B) elementary school, and (C) money changer shop.

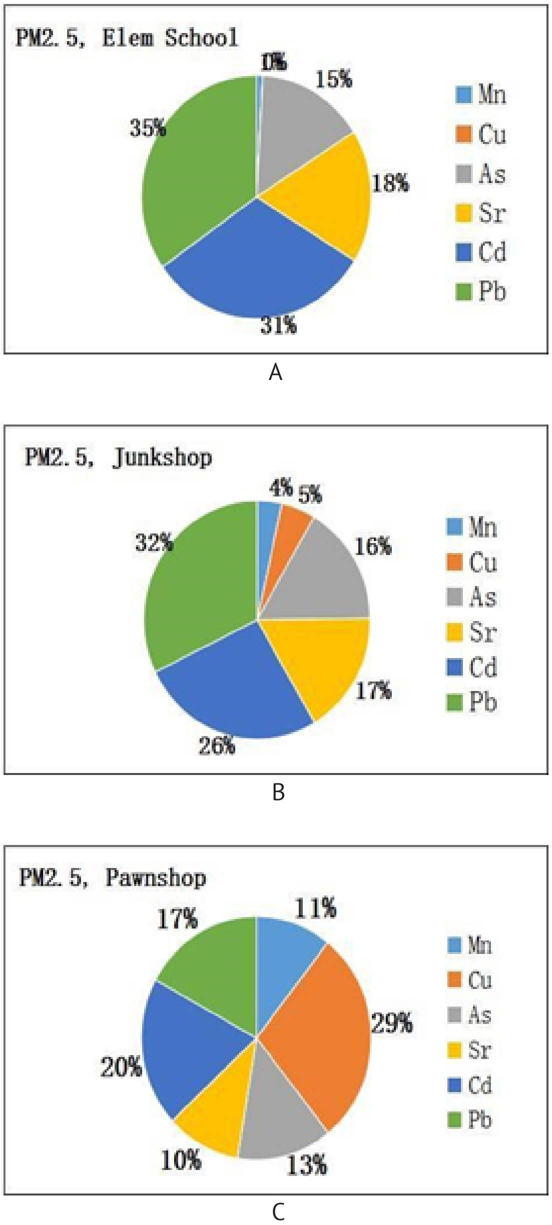
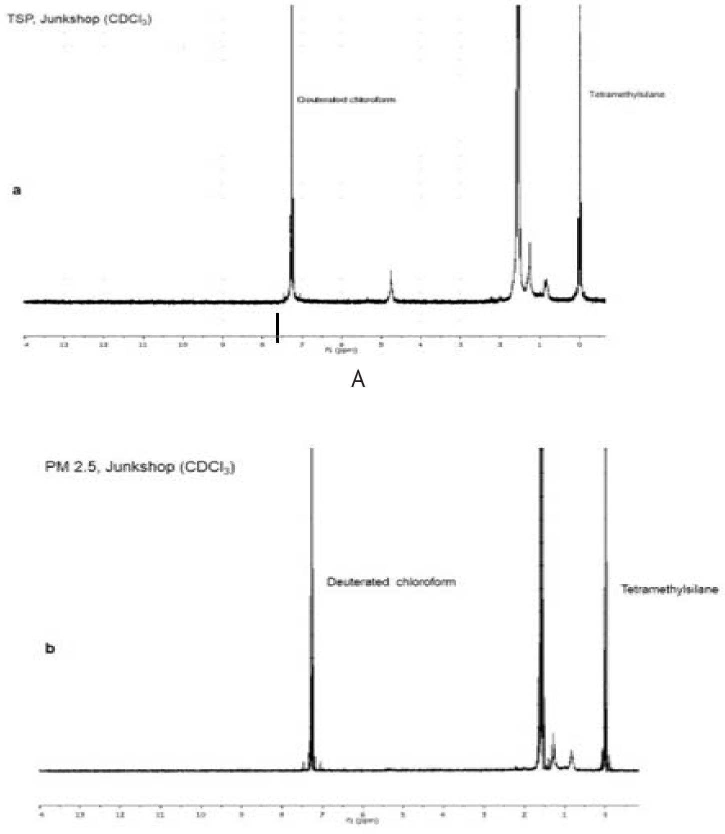


Figure 5. Metal concentrations in $PM_{2.5}$ fractions collected from sampling sites. (A) Junk shop, (B) elementary school, and (C) money changer shop.

Potential Speciation of Inorganic and Organic Constituents in PM

Simulation results show that solution complexes of H_2AsO^{3-}, $HAsO_3^{2-}$, AsO_3^{3-}, H_3AsO_3, $Cd(OH)_2$, $CdOH^+$, $Cd(OH)^{3-}$, Cd^{2+}, Cd_2OH^{3+}, $Cd(OH)_4^{2-}$, $Pb(OH)_4^{2-}$, $Pb(OH)^{3-}$, $Pb_3(OH)_4^{2+}$, $Pb(OH)_2$, Sr^{2+}, and $SrOH^+$ could potentially form when elemental components interact with the surrounding water vapor. Moreover, phases, such as $Cd(OH)_2$, $Cd_{(s)}$, litharge, massicot, monteponite, $Pb(OH)_2$, $Pb_2O(OH)_2$, $Pb_{(s)}$, and $PbO:0.3H_2O$, could possibly be precipitated after equilibration. Significant formation of isophthalate and protonated Pb-Isophthalate were also identified by the simulation runs. On the other hand, Cd-Isophthalate species were found at lower concentrations.



A

B

Figure 6. Small peaks detected for aliphatic and aromatic groups in all PM fractions collected. (A) TSP and (B) $PM_{2.5}$ fractions. Both fractions were collected from the junk shop.

CONCLUSIONS AND RECOMMENDATIONS

The results contribute to the discussion on local issues with indoor air quality in the workplace by providing background information on the potential impacts from exposure to airborne PM around landfill facilities. The gathered background information provides a baseline data on the chemical characterization and behavior of chemical constituents of PM possibly present in this specific type of environment. This places importance on occupational health in workplaces where Filipino workers (especially women and children) are exposed to environmental agents on respiratory health. Significant levels of metals were observed from all TSP and respirable fractions. Presence of such metals may be attributed to activities involving the production of such metals. However, since no topographical or meteorological parameters were taken in to account, attribution to anthropogenic sources cannot be made with certainty. Organic phases with aromatic and aliphatic characters were also detected in all airborne fractions. An additional peak for the TSP fraction may also be correlated to the presence of some alcohols.

The data gathered from this study will be used for further modeling studies on the speciation of the chemical constituents of PM. The results will also be utilized in developing methods to quantitatively determine the species formed during the simulations. Likewise, the information collected from the characterization of the respirable PM regarding indoor or workplace air quality is important in the assessment of the exposure of the workers in such occupational settings and the determination of its influence on nearby regions.

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