

Characterization and Elemental Analysis of Particulate Matter in a Chemical Digestion Laboratory

Yasmine Joy M. Labagnoy*, Edanjarlo J. Marquez, Sarah C. Johnson,
Astrid Korina S. Gabo-Gianan, Jervée M. Punzalan and
Migel Antonio P. Catalig

Department of Physical Sciences and Mathematics

University of the Philippines Manila

Manila City, 1000 Philippines

*yas.labagnoy@gmail.com

Date received: May 31, 2021

Revision accepted: November 16, 2021

Abstract

Particulate matter (PM) may constitute health-threatening substances that are classified as carcinogens. In laboratories, common PM-emitting activities include chemical processing and electronic equipment utilization. Thus, workers run the risk of inhaling possible contaminants into their respiratory systems and affecting their overall health. In this study, PM₁₀ and PM_{2.5} sampled from a chemical digestion laboratory were analyzed based on the size distribution and elemental composition. Samples were collected using a cascade impactor coupled with an air pump for 7 h per sampling day. Size distribution showed variation in the number of coarse particles, which was mainly attributable to activities that occurred during sampling. Elemental analysis by scanning electron microscope with energy dispersive X-ray identified 23 elements. Some of the highest obtained concentrations belonged to the elements O, Si, Na and Ba with 41, 21, 10 and 10% in weight percentages, respectively. Possible PM sources included indoor dust, laboratory reagents, mechanical abrasions of buildings, crustal material and combustion emission. Furthermore, six elements from the identified PM constituents, namely Ba, Cl, Al, F, Mn and Cr were determined as inhalation contaminants based on toxicological reports from the Agency for Toxic Substances and Disease Registry.

Keywords: elemental analysis, indoor air quality, particulate matter

1. Introduction

People spend approximately 90% of their time indoors (e.g., home, workplace). Thus, studies on particulate matter (PM) behavior in indoor settings are relevant to human health (Sanchez-Soberon *et al.*, 2019). Indoor air pollution (IAP) is responsible for more than one million annual deaths

worldwide (Junaid *et al.*, 2018). Furthermore, an estimated seven million people die yearly from diseases such as stroke, chronic obstructive pulmonary disease, pneumonia, heart disease and lung cancer caused by ambient and indoor PM (World Health Organization [WHO], 2018). In 2016, it was recorded that 84 deaths per 100,000 Filipinos were due to indoor air pollution, while 45 deaths per 100,000 Filipinos were linked to outdoor air pollution (Philippine Daily Inquirer, 2018). It was also reported that the Philippines ranked 57th out of 98 countries in terms of PM_{2.5} concentration in 2019 (Enano, 2020).

PM is made up of a heterogeneous mixture of solid and liquid components. It constitutes combinations of elemental, organic, inorganic, metallic and crustal and/or biological components. These particles can be emitted both from anthropogenic (e.g., agricultural byproducts and traffic exhaust) and natural sources (e.g., volcanic activity, forest fires and biological components). As particle size decreases, it becomes more penetrable to the respiratory system (Bae and Hong, 2018).

The coarse fraction with aerodynamic diameters ranging from 2.5 to 10 μm is primarily formed from road dust and constructions as well as the agglomeration of primary particles into secondary particles resulting in larger aerodynamic sizes (WHO, 2005; Falcon-Rodriguez *et al.*, 2016). They may also be byproducts of combustion (Kim *et al.*, 2015). Particles under this category often deposit into the tracheal region of the upper respiratory airways (WHO, 2005). On the other hand, the fine fraction (also called PM_{2.5}) with aerodynamic diameters less than 2.5 μm mainly originates from the combustion of wood and biomass burning or by aerosol formation through nucleation and condensation (WHO, 2005; Falcon-Rodriguez *et al.*, 2016). These smaller particles tend to accumulate in the bronchioles of the lower respiratory tract as shown in Figure 1a (Kim *et al.*, 2015). Both coarse and fine particles are collectively called PM₁₀. Figure 1b shows the PM sizes relative to human hair. Common chemical constituents in PM include inorganic ions (e.g., sulfates, nitrates and ammonium), organic compounds (e.g., polycyclic aromatic hydrocarbons), crustal materials (e.g., manganese [Mg], calcium [Ca] and potassium [K]), heavy metals (e.g., nickel [Ni], lead [Pb] and copper [Cu]), particle-bound water and biological components (e.g., allergens, microbes, pollen, mold spores, plant and insect parts) (Kinney, 2008). Indoor activities such as sweeping, dusting, cooking, walking and using electronics also emit particles (Kim *et al.*, 2015).

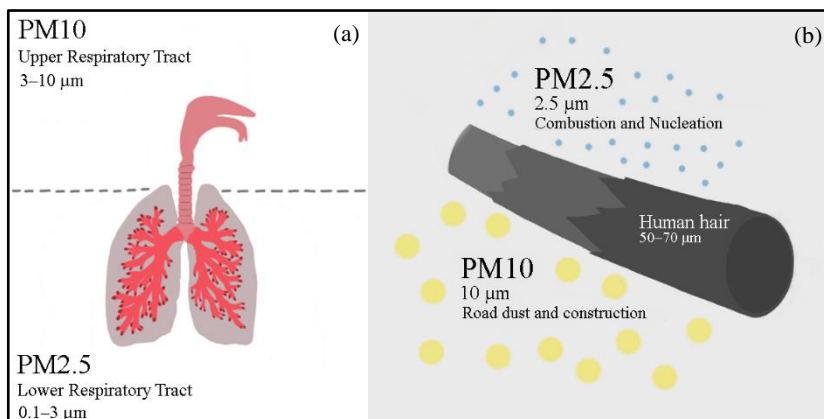


Figure 1. Characteristics of PM size: deposition potential of PM from the upper to lower respiratory tracts (a) and the size comparison of coarse and fine particles against human hair (b)

The International Agency for Research on Cancer (IARC) classifies PM as a group 1 (human carcinogen) contaminant. More specifically, some agents that may constitute PM are designated as carcinogens such as metals arsenic (As), beryllium (Be), cadmium (Cd) and chromium (Cr) (IARC-WHO, 2015). Furthermore, the United States Environmental Protection Agency (n.d.) also advocates executing health risk assessment defined as “the characterization of the potential adverse health effects of human exposures to environmental hazards.” This includes (1) hazard identification, which identifies health hazards associated with the contaminant and (2) dose-response assessment, which defines a quantitative relationship between the health hazard and the contaminant.

In the Philippine setting, some studies related to workplace PM have been conducted. Lomboy *et al.* (2015) sampled PM from a government tertiary care hospital, where they found that PM_{2.5} concentrations in the naturally ventilated Departments of Pediatrics and Medicine exceeded the prescribed WHO limit of 25 μg/m³. They also detected levels of Hg and Pb, which are potential carcinogens albeit in insignificant amounts. Lamorena-Lim and Rosales (2016) collected PM in establishments such as a junk shop, an elementary school, a pawnshop and a near landfill facility. Samples were analyzed for their constituting agents, which included metals Pb, Cd, Mn, As and strontium (Sr) in significant levels, as well as hydrocarbons. A study by Zali *et al.* (2015) sought to provide baseline data for indoor PM in Metro Manila offices. Concentration levels of CO₂ were examined, wherein values were above the

standard set by the American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE).

To the authors' knowledge, local studies on the risk due to PM inhalation from laboratory activity have not been explored. The frequent use of chemical substances, product processing through burning and heating, and the utilization of electronic equipment such as printers and computers are all possible PM emitting sources (Kim *et al.*, 2015). Particles that are accumulated indoors may eventually pose both carcinogenic and non-carcinogenic threats to human health. With this, the study aimed to investigate the characteristics and potential health risks of indoor PM in a laboratory setting. Specifically, the study characterized PM based on the size distribution and elemental composition and implemented health risk assessment by hazard identification.

2. Methodology

2.1 Study Area

A service laboratory within Metro Manila, Philippines was chosen for PM collection. It was a chemical digestion room that houses four fume hoods, hot plates, waste containers and a shower area. Fume hood 1 was used for flammable solvents, ammonium hydroxide (NH_4OH) and hazardous substances. Fume hood 2 was utilized for nitric acid (HNO_3) and sulfuric acid (H_2SO_4); fume hood 3 for phosphoric acid (H_3PO_4) and hydrochloric acid (HCl); and Fume hood 4 for acetic acid (CH_3COOH) and HCl . Each fume hood housed a hot plate and some chemical containers. Amber waste jugs for chemical compounds such as molybdovanadophosphoric acid (MVPA) were also present. The room was ventilated by two open windows. Directly below the windows was a roadside where vehicles often passed by the busy road. The location for the experimental setup was placed in an area where the workers would least be disrupted in their tasks. Therefore, the collection equipment was placed on a tabletop near the shower area. Table 1 shows a summary of the environmental aspects of the site, while Figure 2 displays photographs taken in the sampling site and its floorplan.

Table 1. Summary of conditions in the lab

Aspect	Details
Laboratory type	Chemical digestion room
Ventilation	Two open windows
Floor number	Second floor
Floor area	26.53 m ²
Occupancy	2-3 workers
Work hours	Weekdays (8 AM to 5 PM)

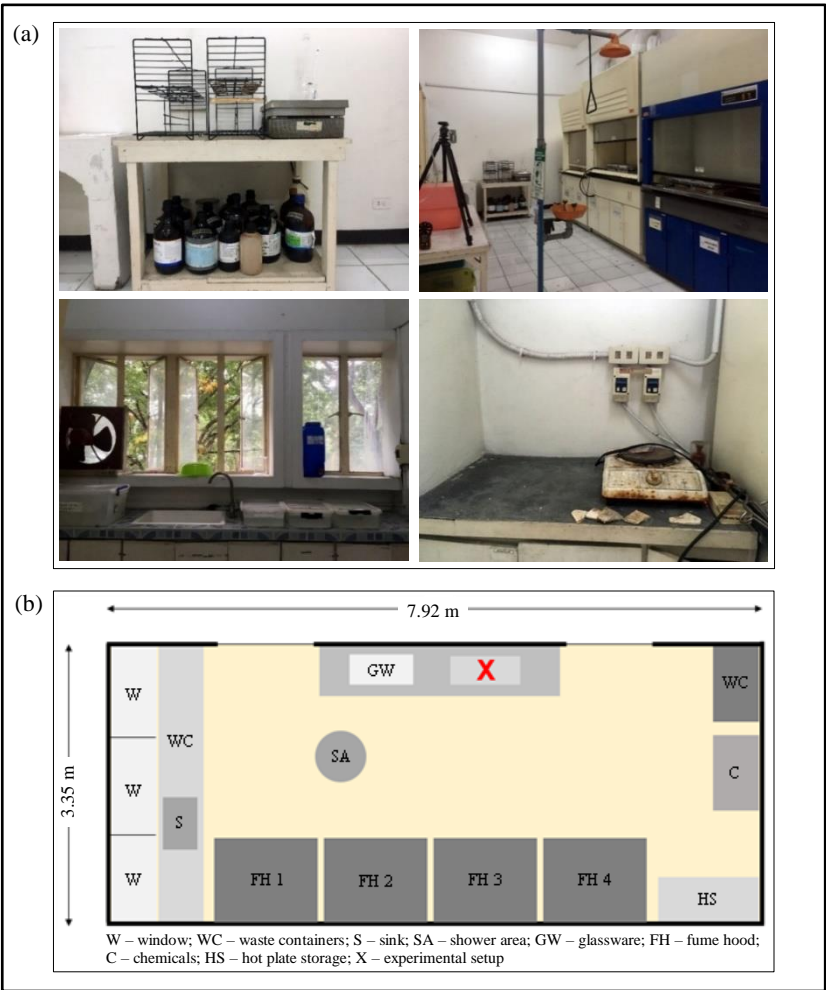


Figure 2. Chemical digestion laboratory as sampling site: photographs taken inside the lab including windows, fume hoods and the chemical digestion area (a) and the approximate dimensions and floor plan of the different components present in the lab (b)

2.2 Sample Collection

The personal cascade impactor (Series 290 Marple Style, Tisch Environmental, Inc., United States) is an instrument used to categorize PM into size fractions and measure PM mass concentrations. This was used to collect PM samples on eight glass fiber filters per sampling point along with the air pump (BUCK Libra™ L-4 Pump, A. P. Buck Inc., United States). Figure 3 shows images of each material used for the sampling.

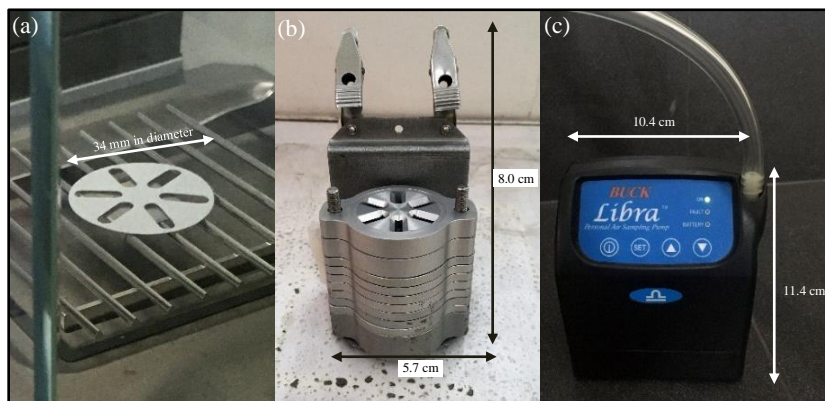


Figure 3. Equipment and materials used for sampling: glass fiber filter (a), cascade impactor (b) and air pump (c)

Data collection was carried out for two days (December 2 and December 9 in 2019). Each sampling day lasted for 7 h from 9 AM to 4 PM. The general activities conducted per day were also recorded. Each glass fiber filter was equilibrated in sealed petri dishes for at least 24 h before sampling. A glass fiber filter was placed in between each of the eight stages of the cascade impactor, which served as a substrate to collect PM. Table 2 shows the cut-point for each impactor stage lifted from Tisch Environmental, Inc. (2003). The air pump, connected to the cascade impactor via sorbent tubing, sucked air towards the first stage at a flow rate of 2 L/min.

Table 2. Cut-point of each impactor stage

Impactor stage	Cut-point (μm)	Impactor stage	Cut-point (μm)
1	21.3	5	3.5
2	14.8	6	1.55
3	9.8	7	0.93
4	6.0	8	0.52

The cascade impactor was attached to the head of a tripod placed on top of a table to prevent obstructions in the workplace (Figure 4a). The height from the tripod head to the floor was 1.55 m, which is the average breathing height of an adult Filipino (Lozada, 2020). After 7 h of sampling, the filters were placed back into the petri dishes for storage for future analysis.

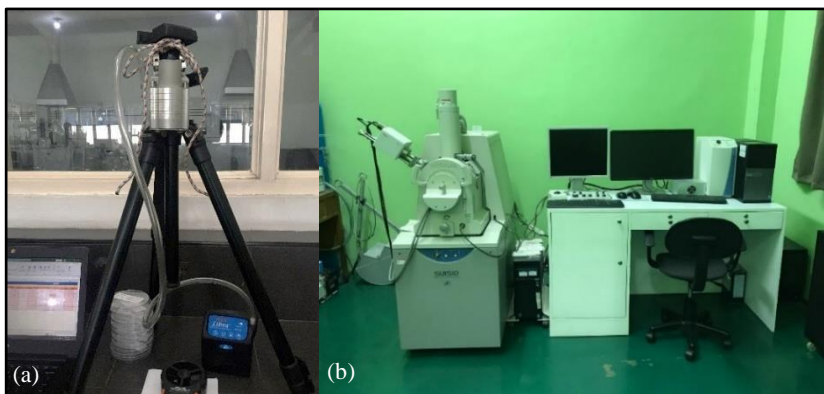


Figure 4. PM data collection and analysis: experimental setup for PM collection on a countertop (a) and SEM-EDX at GAEA Research Laboratory, University of the Philippines Manila (b)

2.3 Elemental and Morphological Characterization

The scanning electron microscope (SEM) (SU1510, Hitachi, Japan) coupled with the energy dispersive X-ray (EDX) detector (UltraDry, Thermo Fisher Scientific, Massachusetts, United States) was used for the analysis of the data obtained from the two sampling days to determine the size distribution and elemental composition (Figure 4b). Because the study focused on PM_{10} and $PM_{2.5}$, only particles collected via cascade impactor from substrates three to eight were analyzed – the cut-point for substrates one and two exceeded 10 μm as shown in Table 2.

For the size distribution, the SEM was used to measure the aerodynamic diameters of particles in microns. A third of the area of each analyzed substrate was examined per sampling day. Per substrate, 40 to 42 particles were counted and measured across the entire one-third of the filter to allow uniform distribution as much as possible, totaling 250 analyzed particles per sampling day. Individual particle diameters were assessed by manually drawing dimension marks on each particle using the data entry function of the SEM software. The assigned diameters were taken to be the longest dimension from each end of the particle for both spherical and non-spherical types. Data for

elemental composition was obtained using energy-dispersive X-ray spectroscopy. An accelerating voltage of 20 kV was utilized to detect heavy metals. The magnification power used ranged from 1,300x to 6,500x and no viewing modifications were applied.

From the elemental composition obtained from SEM-EDX analysis, contaminant identification was done based on toxicity information collated by the Agency for Toxic Substances and Disease Registry (ATSDR). The data from ATSDR was utilized to assess PM health risk by identifying and characterizing the health hazards of inhalable contaminants found in the environment.

2.4 Data Analysis

Correlation analyses of the weight percentages of relevant elemental pairs were performed to determine relative concentrations of each element and inter-element interactions, which were used as the basis of possible sources of the detected elements. Pearson Correlation was used via Microsoft Excel to derive Pearson's R-values. Significance levels or P-values were also calculated at $\alpha = 0.05$.

3. Results and Discussion

3.1 Size Distribution

Figures 5 and 6 exhibit the size distribution of coarse and fine particles on the two data points, December 2 and December 9, 2019, respectively. Table 3 shows a summary of activities that occurred during the two sampling days. A total of 250 particles per sampling day were analyzed. The results from December 2 showed an equal distribution of fine (50%) and coarse particles (50%) with 125 particles each. On the other hand, the data on December 9 displayed a distribution composed of 34% fine particles and 66% coarse particles.

The larger percentage of coarse particles on December 9 is attributed to a high human activity consisting of two occupants frequently entering and exiting the site and using a fume hood and a hot plate throughout the day. Increased human presence contributes to the agitation of settled particles (Kim *et al.*, 2015). In comparison, activities conducted on December 2 were minimal

consisting of one to two occupants entering to wash glassware or dispose of chemical waste. Additional particles can also be released from clothing movement (Fromme *et al.*, 2007). The use of the fume hood, apart from particle emission of electronic components, also creates vibratory movements that contribute to the resuspension of particles (Chatoutsidou *et al.*, 2015). Additionally, coarse particles may be emitted from high-heat chemical processes (e.g., hot plate use) or the accumulation of fine primary particles via agglomeration (Kim *et al.*, 2015; Falcon-Rodriguez *et al.*, 2016).

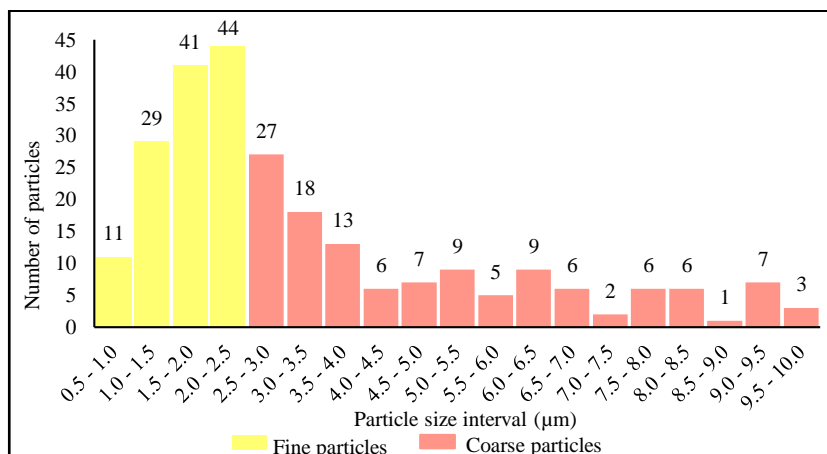


Figure 5. Size distribution of PM₁₀ for December 2: equal distribution for coarse and fine particles

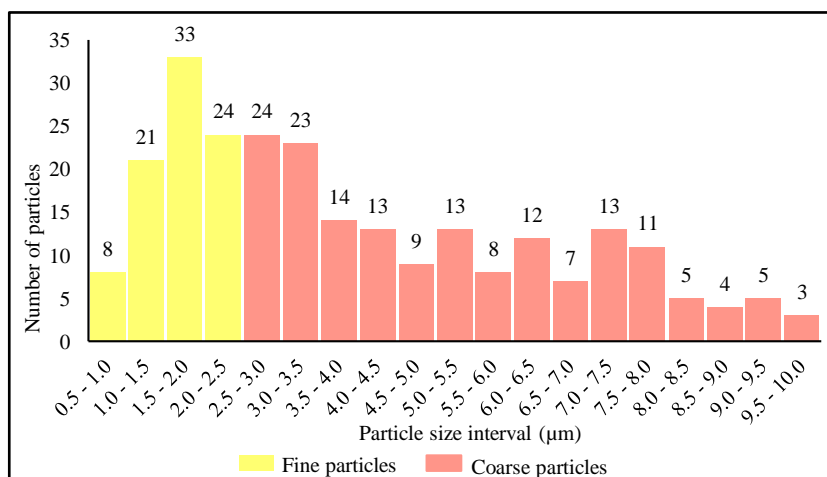


Figure 6. Size distribution of PM₁₀ for December 9: greater distribution for coarse particles compared with fine particles

Table 3. Summary of activities conducted in the lab during sampling

Day	Activities
December 2	Washing glassware Disposing chemical waste
December 9	Entering and exiting site Chemical digestion processes with use of the fume hood and hot plate

3.2 Elemental Composition

Twenty-three elements were identified using EDX spectroscopy. Upon identification, a summary of each element's weight percentage per particle was also generated. Table 4 displays the calculated average weight percentage per element detected. These values were obtained by averaging the weight percentages acquired from the 500 analyzed particles.

Table 4. Elemental composition of PM₁₀ in the chemical digestion laboratory

Element	Average weight (%)	Element	Average weight (%)
O	40.67	Zn	1.96
Si	20.69	K	1.64
Na	9.95	Fe	0.96
Ba	9.77	Mg	0.42
Cl	3.94	Ti	0.13
Al	2.99	S	0.13
Ca	3.31	P	0.03
C	2.29	N	0.02
F	2.07	Mo, Mn, Br, Cr, Tl, Cu	0.03

The identified sources of the elemental constituents were dependent on the type of ventilation and the nature of the laboratory. Oxygen (O) and silicon (Si), which are common components of dust, have the greatest values of weight percentage (Pateraki *et al.*, 2012). The metals Mg, K, Al, titanium (Ti), Cr, Mn, zinc (Zn), Br, and thallium (Tl) can be found in rusting building parts and mechanical abrasions of industrial materials (glass, cement and ceramics) (Karnae and Kuruvilla, 2019). The elements carbon (C), nitrogen (N), fluorine (F), sodium (Na), Ca, P, sulfur (S), Cl, iron (Fe), Cu, molybdenum (Mo), and barium (Ba) can be accounted to components and byproducts that are involved in chemical preparations (Matusiewicz, 2003; Bhargava *et al.*, 2006). It is noteworthy to mention that metals Mn, Cr and Cu, which can also be common agents associated with PM originating from traffic emissions and weathering

of buildings, were detected in insignificant levels (Hieu and Lee, 2010; Du *et al.*, 2013). This may be due to the environment, which is dominated by salt formation activities. However, more samples obtained from this type of laboratory may be needed to support this.

The activities that occurred in the laboratory included walking, using machinery and electronics, conducting chemical reactions and heating processes, which are all PM-emitting sources. In general, the chemical activities that take place in chemical digestion laboratories mainly involve the use of precipitating reagents processed at high temperatures ranging from 90 to 400 °C (Bhargava *et al.*, 2006).

The most common byproducts of chemical digestion processes are neutral salts. Wet oxidation is a process that removes organic compounds in liquid form by oxidizing them to carbon dioxide and water under the influence of high heat. Examples of neutral salts include sodium chloride (NaCl), ammonium sulfate ((NH₄)₂SO₄) and magnesium sulfate (MgSO₄) (Matusiewicz, 2003). Another method called acid digestion breaks down compounds into ions in an aqueous solution using heat and acid as the oxidizing agent. Common acid reagents include hydrochloric acid (HCl) and perchloric acid (HClO₄), which form chloride salts and perchloric salts, respectively (Matusiewicz, 2003). The chemical digestion room may still contain PM accumulated from other days apart from the sampling dates due to their lifetime, which ranges from minutes to weeks (Cheung *et al.*, 2011). Figure 7 shows the spectrum and micrograph of a fine particle that is high in Na and Cl.

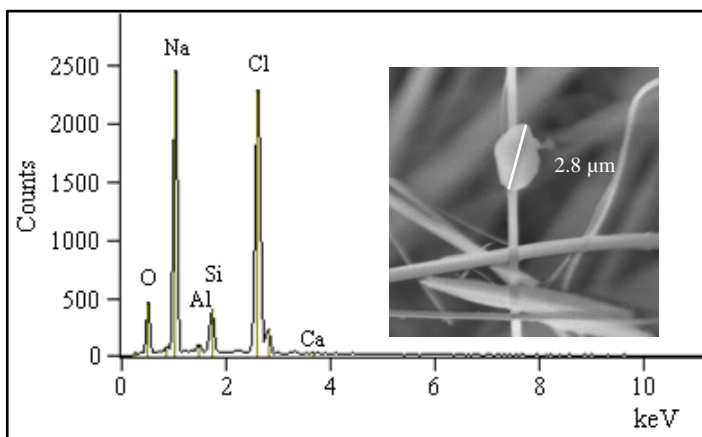


Figure 7. Na and Cl rich particle: particles often emitted from wet oxidation and acid digestion

Most of the particles analyzed also contained Na and Cl ranging from trace to major amounts. Table 4 also shows relatively high weight percentages with 10% (Na) and 4% (Cl).

Dust is a common source of PM in indoor spaces, which may contain high amounts of Si and O. Dust may also be industrial in origin such as from rusting windowsills, cement and alloys. These particles may contain Al, Mg and Fe (Karnae and Kuruvilla, 2019). Si and O may also be sourced from silica (SiO_2) originating from abundant glassware, ceramics and quartz materials present in the room (Pachauri, 2013). The presence of dust can be attributed to resuspended particles from clothing and human activity or the infiltration of outdoor road dust through the two open windows. Particles rich in Si and O are typically irregular in shape (Matusiewicz, 2003). Figure 8 shows the spectrum and micrograph of a coarse particle rich in Si and O. Relatively high amounts of P and Ca are also present in the spectrum. Phosphoric acid (H_3PO_4) is another common oxidizing agent used for acid digestion which decomposes into metaphosphoric acid (HPO_3). P can also be sourced from a common, water-soluble salt used as a buffering and emulsifying agent called tetrasodium pyrophosphate ($\text{Na}_4\text{P}_2\text{O}_7$) (Matusiewicz, 2003). Furthermore, Ca can be sourced from the chemical digestion of hydrochloric acid and acetic acid (CH_3COOH) into the salts calcium chloride (CaCl_2) and calcium acetate ($\text{Ca}(\text{C}_2\text{H}_3\text{O}_3)_2$), respectively (Hanusa, 2019).

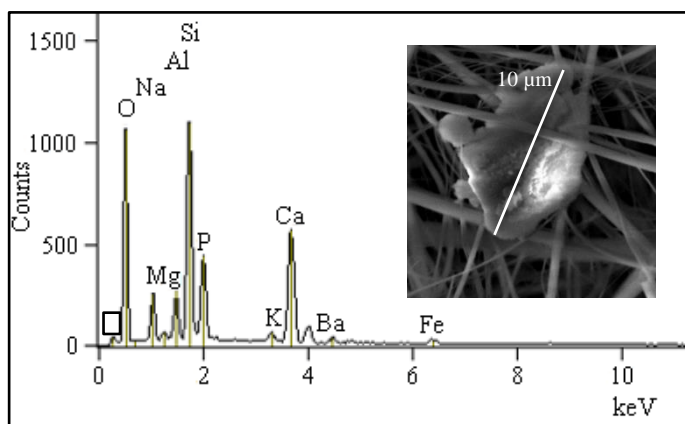


Figure 8. Si and O rich particle: common constituents of indoor dust

Table 4 shows a relatively high weight percentage for Ba (10%). Barium chloride (BaCl_2) is a common salt used for heat baths to precipitate soluble sulfates (Hanusa, 2018). It is also a salt product of a Ba and HCl chemical

reaction. Ba is also a component of construction materials such as glass and alloys (Karnae and Kuruvilla, 2019). Furthermore, barium fluoride (BaF_2) is a compound used in windows for infrared (IR) spectroscopy (Hanusa, 2018). The facility where the lab is located has a dedicated room on the same floor for IR spectroscopy analyses. Resuspension of dust particles through clothes and human activity may allow F-carrying particles to be transported into the lab. Hydrofluoric acid (HF) is also a common reagent used in acid digestion (Matusiewicz, 2003). Figure 9 exhibits the spectrum and micrograph of a fine particle rich in F, Cl and Ba, as well as Si and O. The trace amounts of K may come from chemical digestion reagents potassium hydroxide (KOH) and potassium permanganate (KMnO_4). Potassium salts such as potassium chloride (KCl) and potassium nitrate (KNO_3) may result from hydrochloric acid and nitric acid (HNO_3) reactions (Matusiewicz, 2003).

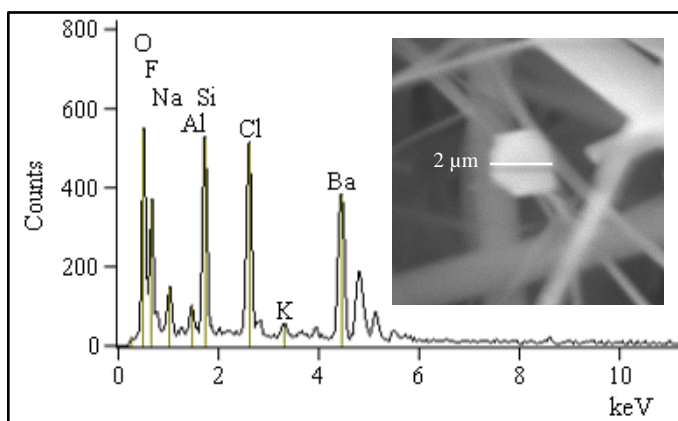


Figure 9. F, Cl and Ba rich particle: constituents may originate from chemical activity, resuspension, or construction material

Carbonaceous particles consist mainly of C and varying amounts of O. These particles tend to come from high-temperature processes such as precipitation by heating and diesel combustion (Mazzei *et al.*, 2008). Both are possible sources for the particle because (1) the site has two open windows above a roadside for the outdoor PM to infiltrate into and (2) the chemical digestion room houses hot plates used for chemical preparations. These particles are usually spherical (Bautista *et al.*, 2014). Figure 10 shows the spectrum and micrograph of a spherical C-rich particle. The spectrum also displays small amounts of S and Zn. S may come from the wet digestion of the common oxidizing agent sulfuric acid (H_2SO_4), which can form the byproduct sodium sulfate (Na_2SO_4). On the other hand, Zn can be found in building materials but

can also be used as an oxidizing agent such as in zinc nitrate ($(\text{Zn}(\text{NO}_3)_2)$ (Matusiewicz, 2003; Hieu and Lee, 2010). Common Zn salts also include zinc acetate ($\text{Zn}(\text{CH}_3\text{CO}_2)_2$) and zinc carbonate (ZnCO_3) (Matusiewicz, 2003). Furthermore, the presence of Si and Al can be attributed to SiO_2 -Al compounds originating from glass, cement and ceramics (Karnae and Kuruvilla, 2019).

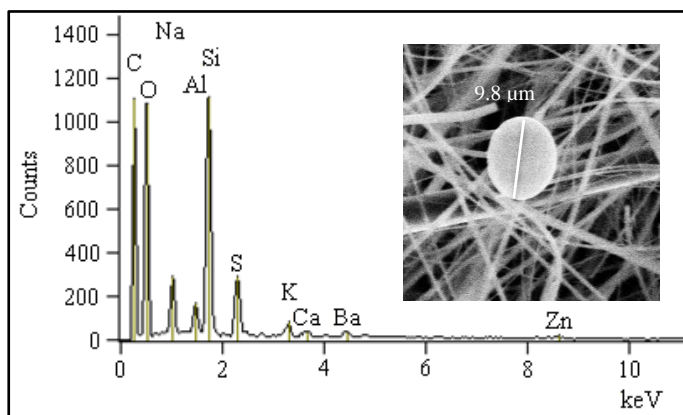


Figure 10. C rich particle: particles that may originate from high-heat processes or by infiltration from the outdoors

3.3 Correlation Result

Figure 11 displays the scatter plots of elemental pairings that exhibit strong correlation ($R \geq 0.60$). F and Ba obtained the highest value at $R = 0.82$. Given the nature of the site, it is likely that the source of F may come from HF since it is a common agent used for the analysis of environmental and biological samples (e.g., water and marine sediment samples) (Matusiewicz, 2003). This may be further supported by the strongly negative coefficients of F with most detected elements e.g., $R = -0.70$ with O.

The correlation between Si and K ($R = 0.72$) may suggest that the presence of K did not originate from K salts, but from crustal or pavement material because of its association with Si (Kim *et al.*, 2015). Furthermore, the association of K with Zn ($R = 0.69$) supports the two elements as the result of industrial or building material abrasion (Karnae and Kuruvilla, 2019). Additionally, they may also be related to traffic emission since K and Zn may originate from road dust and tire wear, respectively (Kim *et al.*, 2015). All P-

values of the previously mentioned pairings also suggested good associations between each pair ($p \leq 0.05$).

Figure 12 shows the scatter plots involving Cl. The correlation coefficient between Na and Cl ($R = 0.70$) reinforces the claim that their presence may be attributed to NaCl as a byproduct of various chemical digestion methods (Matusiewicz, 2003). Furthermore, it is noteworthy to mention that there was a negative correlation between Na and Cl for the lower values of weight percentages. This may be attributed to the association of Cl with Ba, albeit weakly correlated ($R = 0.24$), for these values as displayed in Figure 12b. Moreover, the possible source for Cl and Ba may be the common salt product $BaCl_2$ used for the detection of sulfates in solid/liquid fuels and lubricants (Hanusa, 2018). This is also supported by the negative correlations of Ba with the other detected elements e.g., $R = -0.80$ with O as well as the P-values for these elemental pairs.

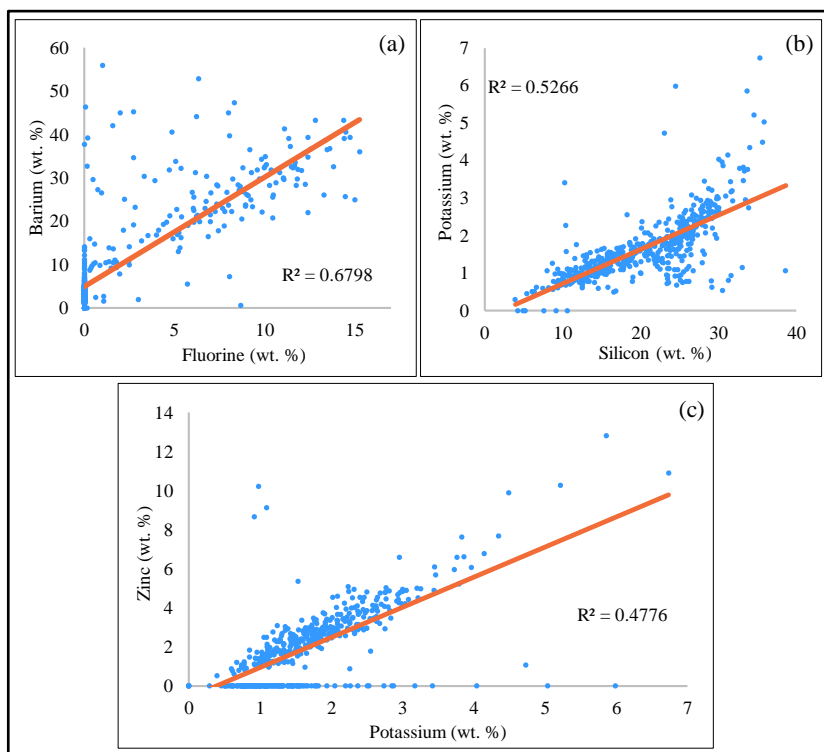


Figure 11. Strong correlation plots: F and Ba (a), Si and P (b) and K and Zn (c)

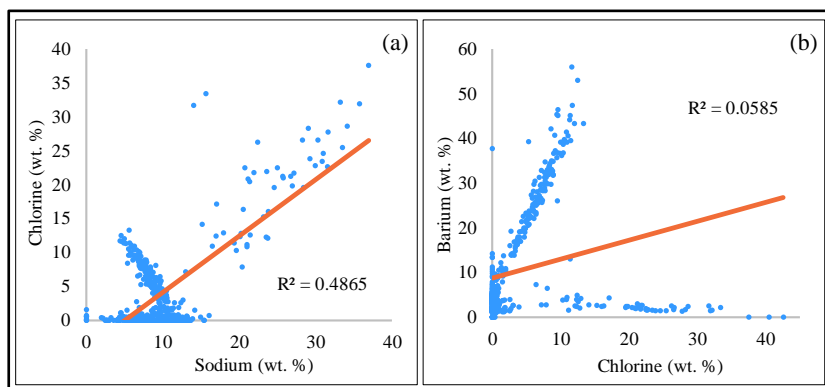


Figure 12. Correlation plots of Cl: Na and Cl (a) and Cl and Ba (b)

Figure 13 presents the correlation plots of elemental pairings with moderate correlations ($R = 0.40$ - 0.59). Silicon correlated with zinc ($R = 0.57$) further supports the latter being a byproduct of traffic exhaust due to the former's association with pavement material (Kim *et al.*, 2015).

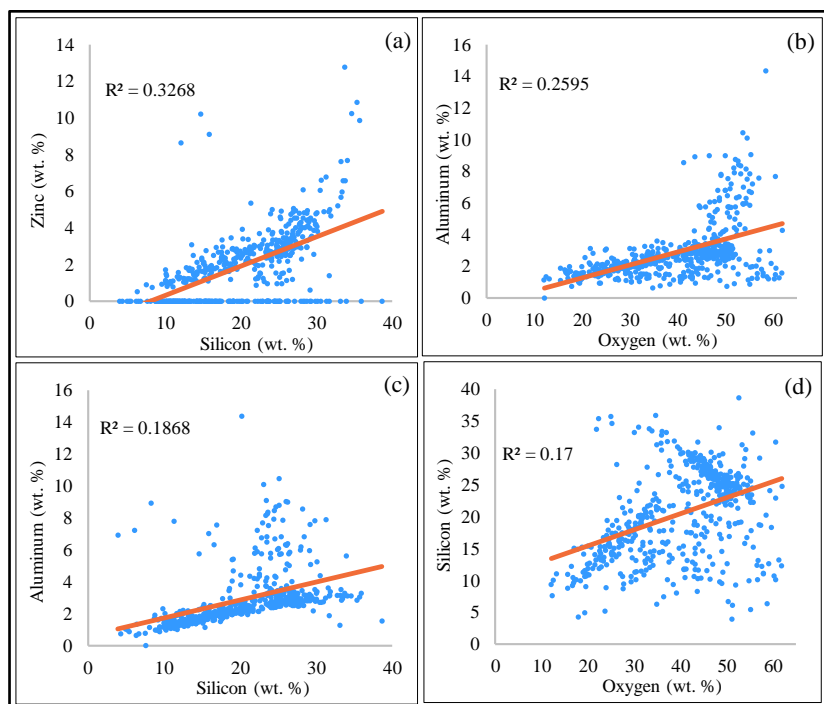


Figure 13. Moderate correlation plots: Si and Zn (a), O and Al (b), Si and Al (c) and O and Si (d)

Figures 13b, 13c and 13d show the moderate correlation plots among O, Al, and Si with R-values ranging from 0.41-0.51. Their association with each other is indicative of indoor dust with the main constituents O and Si, and Al is an additional component if the dust is industrial in origin (e.g., from rusting parts) (Karnae and Kuruvilla, 2019). The corresponding P-values for the moderate correlation pairings also support these associations.

Other correlations that may not be classified as strong or moderate ($R = 0.10$ - 0.40) but are still noteworthy are shown in Table 5. These pairings were considered based on previously mentioned possible sources, namely crustal material, mechanical abrasion of buildings and products of chemical digestion.

Table 5. Correlation values of noteworthy elemental pairs and their possible sources

Elemental pair	R-value	P-value	Possible source
Ca and O	0.35	1.52×10^{-15}	Crustal material (Pachauri <i>et al.</i> , 2013) Chemical digestion e.g., $\text{Ca}(\text{C}_2\text{H}_3\text{O}_3)_2$
Mg and O	0.29	1.51×10^{-11}	Crustal/building material (Pachauri <i>et al.</i> , 2013)
Fe and Mg	0.25	7.48×10^{-9}	Crustal/building material (Pachauri <i>et al.</i> , 2013)
Ba and Cl	0.24	4.30×10^{-8}	Chemical digestion e.g., BaCl_2
S and N	0.16	5.04×10^{-4}	Chemical digestion e.g., $((\text{NH}_4)_2\text{SO}_4)$
Mn and Cr	0.15	1.12×10^{-3}	Building/industrial material (Hieu and Lee, 2010)
S and Mg	0.14	1.99×10^{-3}	Chemical digestion e.g., (MgSO_4)
K and O	0.11	1.85×10^{-3}	Crustal material (Pachauri <i>et al.</i> , 2013) Chemical digestion e.g., KOH

3.4 Hazard Identification

The 23 detected elements were categorized into two groups: air contaminants and non-contaminants. The identification of contaminants was based on toxicology reports from the Tox Profile Index of the Agency for Toxic Substances and Disease Registry (ATSDR, 2021). Figure 14a displays the

percentage of the six identified contaminants out of 23 elements (19%) that pose potential health risks, mainly involving the respiratory system (ATSDR, 2003, 2007, 2008, 2010, 2012a, 2012b). These are F, Al, Cl, Cr, Mn and Ba. The 19% is further broken down into its constituents in Figure 14b. Although some of the data present very low levels of the weight percentage (e.g., Mn and Cr), the presence of these elements in PM was significant due to their toxic properties and ability to interact with the human body by respiratory airway absorption or reactive oxygen species (ROS) production (Falcon-Rodriguez *et al.*, 2016).

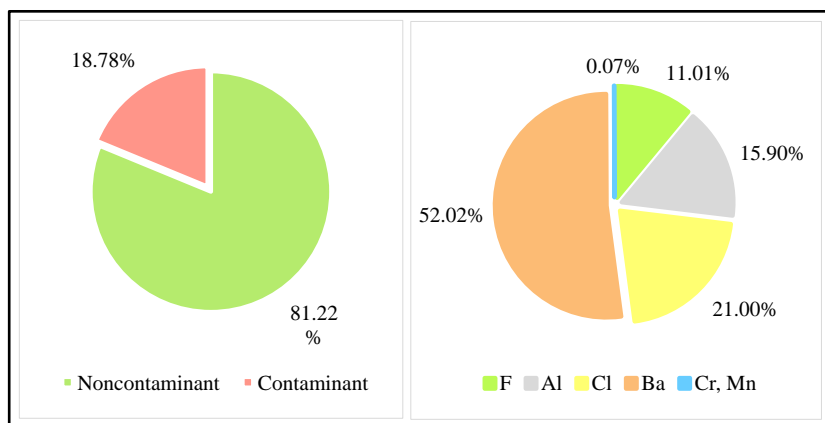


Figure 14. Identified air contaminants: aggregate weight percentages of detected contaminants versus noncontaminants on December 2 and December 9 (a) and percentage contribution of the detected air contaminants (b)

Table 6 exhibits the minimum, maximum and median weight percentages of contaminants detected among all 500 analyzed particles. It demonstrates that a contaminant can take up a large portion of the weight of a single particle. For example, Ba takes up 56% of a particle collected on December 9 within the 2.5-3 μm interval. However, these values were based only on detected elements, which means biological components were not accounted for.

The effects of toxins in PM on human health are dependent on an individual's age, health and lifestyle. Ailments resulting from PM exposure can range from mild problems (chest pain, shortness of breath and coughing) to life-threatening illnesses (lung damage, pre-term deliveries, diabetes and cancer) (Kim *et al.*, 2015). Furthermore, a toxin's likeliness to cause non-carcinogenic effects in humans over a specific period of exposure is defined by minimal risk levels (MRLs) (ATSDR, 2003).

Table 6: Elemental contaminants and their minimum, maximum and median weight percentage in a particle

Contaminant	Minimum weight (%)	Maximum weight (%)	Median weight (%)
F	0.04	15.26	7.00
Al	0.63	14.36	2.47
Cl	0.13	42.58	5.23
Cr	0.00	3.22	3.22
Mn	0.37	1.68	1.08
Ba	0.65	55.99	4.19

Specific ailments that may be induced by Al inhalation include impaired lung function and asthma. Accumulated studies related to occupational exposure to Al are currently insufficient to designate it as an inhalation MRL (ATSDR, 2008). High Mn exposure has been related to nervous system effects such as slow and clumsy movements and respiratory irritation. Due to these cases, $0.3 \mu\text{m}/\text{m}^3$ for respirable dust has been derived as its MRL for chronic inhalation exposure (365 days or more) (ATSDR, 2012b). On Cl, its toxicological profile contains reports on nose and throat irritation and lung injury. No inhalation MRL for particulate Cl has been derived so far but acute inhalation (14 days or less) of chlorine gas has a designated MRL of 0.06 ppm (ATSDR, 2010). The toxicological report on F cites instances of eye, skin, and lung irritation. For this, an MRL of 0.01 ppm has been assigned for acute exposure to gas, and no values for particulate exposure currently exist (ATSDR, 2003). Ba-related health effects included respiratory complications as well as increased blood pressure levels. Related investigations are still insufficient to assign Ba an MRL (ATSDR, 2007).

These identified elements react in varied ways when they enter the human body. Metals (e.g., Mn and Al) present in PM, when inhaled and end up in the bloodstream, induce the production of reactive oxygen species. The release of ROS results in tissue damage and inflammation (Falcon-Rodriguez *et al.*, 2016). Cl and F compounds are highly water-soluble and are irreversibly reactive in the respiratory tract (ATSDR, 2003, 2010). Likewise, Ba and its compounds are mainly absorbed in the respiratory airways causing alterations in blood levels and damage in tissues (ATSDR, 2007).

Despite these health implications, toxicological information (dose-response relationships and clinical reports on Mn, Al, Cl, F and Ba) are insufficient to be classified as potential carcinogens (ATSDR, 2003, 2007, 2008, 2010, 2012b). On the other hand, Cr is classified as a known human carcinogen

based on extensive studies of its interaction with DNA, cells and proteins when it enters the human body. The release of chromium-generated ROS induces the conversion and proliferation of cancer cells. Furthermore, particulate Cr(VI) compounds are assigned an inhalation MRL of 0.3 μg chromium(VI)/ m^3 for intermediate exposure (15-364 days) (ATSDR, 2012a).

4. Conclusion and Recommendation

The results of the study highlighted the relevance of PM characterization and health risk assessment in indoor spaces. The size distribution for coarse particles (aerodynamic diameters from 2.5 μm to 10 μm) was greater on December 9 (66%) compared with December 2 (50%). The results were dependent on the activities that occurred during sampling like chemical processes, which resulted in the emission of particles, conducted on December 9. Particle analysis by SEM-EDX resulted in the identification of 23 elements. Si and O, which are characteristic of dust, were the most abundant. High amounts of Na, Cl, Ca, Ba and F, which are common components of neutral salts formed by wet oxidation and acid digestion, were also detected. Carbon, found in relatively high amounts, was mainly attributed to PM emission by high heat processes. Also, the resulting R-values of some elemental pairs suggest possible association to chemical digestion processes, weathered crustal material, traffic exhaust and building or industrial material abrasion. Additionally, six of the 23 identified elements were classified as inhalation-contaminants based on toxicological reports from the ATSDR. Based on previous studies, the elements, namely Al, Ba, Cl, F and Mn, were found to induce mainly respiratory ailments through airway absorption and ROS production. However, studies are still insufficient to classify them as carcinogenic contaminants. On the other hand, Cr is classified as a group 1 human carcinogen causing the proliferation of mutated body cells.

For future studies related to indoor laboratory PM, the collection of “clean” samples (e.g., samples from days with little to no conducted activities) is suggested to allow additional points of comparison.

5. Acknowledgement

The authors would like to express their gratitude to the Department of Science and Technology – Industrial Technology Development Institute (DOST-ITDI) for allowing them to use its facilities.

6. References

Agency for Toxic Substances and Disease Registry (ATSDR). (2021). Toxicological profiles. Retrieved from <https://www.atsdr.cdc.gov/toxprofiledocs/index.html>

Agency for Toxic Substances and Disease Registry (ATSDR). (2008). Toxicological profile for aluminum. Retrieved from <https://www.atsdr.cdc.gov/ToxProfiles/tp22.pdf>

Agency for Toxic Substances and Disease Registry (ATSDR). (2007). Toxicological profile for barium. Retrieved from <https://www.atsdr.cdc.gov/toxprofiles/tp24.pdf>

Agency for Toxic Substances and Disease Registry (ATSDR). (2010). Toxicological profile for chlorine. Retrieved from <https://www.atsdr.cdc.gov/ToxProfiles/tp172.pdf>

Agency for Toxic Substances and Disease Registry (ATSDR). (2003). Toxicological profile for fluorides, hydrogen fluoride, and fluorine. Retrieved from <https://www.atsdr.cdc.gov/ToxProfiles/tp11.pdf>

Agency for Toxic Substances and Disease Registry (ATSDR). (2012a). Toxicological profile for chromium. Retrieved from <https://www.atsdr.cdc.gov/ToxProfiles/tp7.pdf>

Agency for Toxic Substances and Disease Registry (ATSDR). (2012b). Toxicological profile for manganese. Retrieved from <https://www.atsdr.cdc.gov/ToxProfiles/tp151.pdf>

Bae, S., & Hong, Y.C. (2018). Health effects of particulate matter. *Journal of the Korean Medical Association*, 61(12), 749-755. <https://doi.org/10.5124/jkma.2018.61.12.749>

Bautista, A.T., VII, Pabroa, P.C.B., Santos, F.L., Racho, J.M.D., & Quirit, L.L. (2014). Carbonaceous particulate matter characterization in an urban and a rural site in the Philippines. *Atmospheric Pollution Research*, 5(2), 245-252. <https://doi.org/10.5094/APR.2014.030>

Bhargava, S.K., Tardio, J., Prasad, J., Foger, K., Akolekar, D.B., & Grocott, S.C. (2006). Wet oxidation and catalytic wet oxidation. *Industrial and Engineering Chemistry Research*, 45(4), 1221-1258. <https://doi.org/10.1021/ie051059n>

Chatoutsidou, S.E., Ondracek, J., Tesar, O., Torseth, K., Zdímal, V., & Lazaridis M. (2015). Indoor/outdoor particulate matter number and mass concentration in modern

of offices. *Building and Environment*, 92, 462-474. <https://doi.org/10.1016/j.buildenv.2015.05.023>

Cheung, K., Daher, N., Kam, W., Shafer, M.M., Ning, Z., Schauer, J.J., & Sioutas, C. (2011). Spatial and temporal variation of chemical composition and mass closure of ambient coarse particulate matter (PM_{10-2.5}) in the Los Angeles area. *Atmospheric Environment*, 45(16), 2651-2662. <https://doi.org/10.1016/j.atmosenv.2011.02.066>

Du, Y., Gao, B., Zhou, H., Ju, X., Hao, H., & Yin, S. (2013). Health risk assessment of heavy metals in road dusts in urban parks of Beijing, China. *Procedia Environmental Sciences*, 18, 299-309. <https://doi.org/10.1016/j.proenv.2013.04.039>

Enano, J.O. (2020, February 26). Filipinos are breathing dirty, unsafe air – Report. INQUIRER.net. Retrieved from newsinfo.inquirer.net/1233395/filipinos-breathing-dirty-unsafe-air-report

Falcon-Rodriguez, C.I., Osornio-Vargas, A.R., Sada-Ovalle, I., & Segura-Medina, P. (2016). Aeroparticles, composition, and lung diseases. *Frontiers in Immunology*, 7(3). <https://doi.org/10.3389/fimmu.2016.00003>

Fromme, H., Twardella, D., Dietrich, S., Heitmann, D., Schierl, R., Liebl, B., & Ruden, H. (2007). Particulate matter and ultrafine particles in indoor air. *Atmospheric Environment*, 41(4), 854-866. <https://doi.org/10.1016/j.atmosenv.2006.08.053>

Hanusa, T.P. (2018). Barium. Retrieved from www.britannica.com/science/barium

Hanusa, T.P. (2019). Calcium. Retrieved from www.britannica.com/science/calcium

Hieu, N.T., & Lee, B. K. (2010). Characteristics of particulate matter and metals in the ambient air from a residential area in the largest industrial city in Korea. *Atmospheric Research*, 98(2-4), 526-537. <https://doi.org/10.1016/j.atmosres.2010.08.019>

International Agency for Research on Cancer – World Health Organization (IARC-WHO). (2015). Outdoor air pollution: IARC monographs on the evaluation of carcinogenic risks to humans. Retrieved from <https://publications.iarc.fr/538>

Junaid, M., Syed, J.H., Abbasi, N.A., Hashmi, M.Z., Malik, R.N., & Pei, D.S. (2018). Status of indoor air pollution (IAP) through particulate matter (PM) emissions and associated health concerns in South Asia. *Chemosphere*, 191, 651-663. <https://doi.org/10.1016/j.chemosphere.2017.10.097>

Karnae, S., & Kuruvilla, J. (2019). Source apportionment of PM_{2.5} measured in South Texas near U.S.A. – Mexico border. *Atmospheric Pollution Research*, 10(5), 1663-1676. <https://doi.org/10.1016/j.apr.2019.06.007>

Kim, K.H., Kabir, E., & Kabir, S. (2015). A review on the human health impact of airborne particulate matter. *Environment International*, 74, 136-143. <https://doi.org/10.1016/j.envint.2014.10.005>

Kinney, P.L. (2008). Climate change, air quality, and human health. *American Journal of Preventive Medicine*, 35(5), 459-467. [10.1016/j.amepre.2008.08.025](https://doi.org/10.1016/j.amepre.2008.08.025)

Lamorena-Lim, R.B., & Rosales, C.M.F. (2016). Chemical characterization and behavior of respirable fractions of indoor dusts collected near a landfill facility. *Science Diliman*, 28(1), 17-33.

Lombay, M.F.T.C., Quirit, L.L., Molina, V.B., Dalmacion, G.V., Schwartz, J.D., Suh, H.H., & Baja, E.S. (2015). Characterization of particulate matter 2.5 in an urban tertiary care hospital in the Philippines. *Building and Environment*, 92, 432-439. <https://doi.org/10.1016/j.buildenv.2015.05.018>

Lozada, B. (2020, April 25). Filipinos second-shortest in Southeast Asia. INQUIRER.net. Retrieved from globalnation.inquirer.net/102688/filipinos-second-shortest-in-southeast-asia

Matusiewicz, H. (2003). Wet digestion methods. Retrieved from http://www.pg.gda.pl/chem/CEEAM/Dokumenty/CEEAM_ksiazka/Chapters/chapter13.pdf

Mazzei, F., D'Alessandro, A., Lucarelli, F., Nava, S., Prati, P., Valli, G., & Vecchi, R. (2008). Characterization of particulate matter sources in an urban environment. *The Science of the Total Environment*, 401(1-3), 81-89. <https://doi.org/10.1016/j.scitotenv.2008.03.008>

Pachauri, T. (2013). SEM-EDX characterization of individual coarse particles in Agra, India. *Aerosol and Air Quality Research*, 13(2), 523-536. <https://doi.org/10.4209/aaqr.2012.04.0095>

Pateraki, S., Asimakopoulos, D.N., Flocas, H.A., Maggos, T., & Vasilakos, C. (2012). The role of meteorology on different sized aerosol fractions (PM₁₀, PM_{2.5}, PM_{2.5-10}). *Science of the Total Environment*, 419, 124-135. <https://doi.org/10.1016/j.scitotenv.2011.12.064>

Philippine Daily Inquirer. (2018, May 3). PH ranks 2nd in Asia-Pacific in deaths due to household pollution. INQUIRER.net. Retrieved from newsinfo.inquirer.net/987262/ph-ranks-2nd-in-asia-pacific-in-kinneydeaths-due-to-household-pollution

Sanchez-Soberon, F., Rovira, J., Sierra, J., Mari, M., Domingo, J.L., & Schuhmacher, M. (2019). Seasonal characterization and dosimetry-assisted risk assessment of indoor particulate matter (PM_{10-2.5}, PM_{2.5-0.25}, and PM_{0.25}) collected in different schools. *Environmental Research*, 175(3), 287-296. <https://doi.org/10.1016/j.envres.2019.05.035>

Tisch Environmental, Inc. (2003). Marple style personal cascade impactors 290 series operators manual. Retrieved from <https://tisch-env.com/wp-content/uploads/2015/06/TE-290-Marple-personal-impactor.pdf>

United States Environmental Protection Agency. (n.d.). Basic information about the integrated risk information system. Retrieved from www.epa.gov/iris/basic-information-about-integrated-risk-information-system

World Health Organization (WHO). (2005). WHO air quality guidelines for particulate matter, ozone, nitrogen dioxide, and sulfur dioxide. Retrieved from apps.who.int/iris/bitstream/handle/10665/69477/WHO_SDE_PHE_OEH_06.02_eng.pdf?sequence=1

World Health Organization (WHO). (2018). 9 out of 10 people worldwide breathe polluted air, but more countries are taking action. Retrieved from www.who.int/news-room/detail/02-05-2018-9-out-of-10-people-worldwide-breathe-polluted-air-but-more-countries-are-taking-action

Zali, M., Belino, M.C., Bosshard, H.F.R., & Dela Cruz, E.G. (2015). Establishment of baseline data of the indoor air quality of offices in Metro Manila. Proceedings of the the 13th Asia Pacific Conference on Built Environment, Hong Kong, 239-246.