Chemical Fingerprints for PM$_{2.5}$ in the Ambient Air near a Raw Material Storage Site for Iron Ore, Coal, Limestone, and Sinter

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ABSTRACT

To understand the contributions of a raw material storage site for iron ore, coal, limestone, and sinter to ambient air fine particulate matter (PM$_{2.5}$), the concentrations and chemical fingerprints for resuspended and ambient air PM$_{2.5}$ were compared. Investigations were done for 15 piles of raw materials, including 5 iron piles, 5 coal piles, 3 stone piles, and a single pile each for coke and sinter. Additionally, four sites, including A, B, C, and D, in the storage site surroundings were chosen to investigate the ambient air PM$_{2.5}$ concentrations. The concentrations, compositions, and i and j values for PM$_{2.5}$ varied significantly by season in the four sites under investigation. The chemical fingerprints of the PM$_{2.5}$ showed that water-soluble ions were the most important component in all sites. Specifically, SO$_2^-$ and NO$_3^-$ were the predominant water-soluble ions in winter and summer, respectively. The most dominant components in the iron ore, coal, limestone, coke, and sinter piles were iron, carbon, Ca$^{2+}$ and carbon, carbon and SO$_4^{2-}$, and Fe and Ca$^{2+}$, respectively. During the summer, PM$_{2.5}$ concentrations ranged from 13.7–18.0 µg m$^{-3}$, where the chemical composition of water-soluble ions, metals, carbon accounted for 54.2%, 5.7%, and 23.7% respectively. During winter, the concentrations ranged from 44.7–48.0 µg m$^{-3}$, where the water-soluble ions, metals, carbon components accounted for 49.2%, 8.1%, and 17.4% respectively. From the chemical mass balance, the main sources of PM$_{2.5}$ in sites B, C, and D were stationary sources, mobile sources, and secondary organic aerosols. To effectively address the air pollution threat associated with the surroundings of a raw material storage site, the environmental protection agency should formulate measures to effectively reduce the contribution of resuspended dust and other pollution sources to ambient air PM$_{2.5}$.

Keywords: PM$_{2.5}$, Chemical fingerprints, Raw materials, Re-suspension, Seasonal variation

1 INTRODUCTION

Deteriorating air quality caused by elevated concentrations of fine particulate matter (PM$_{2.5}$)
from a raw material storage site for iron ore, coal, limestone, and sinter is a great cause of concern to the residents in the southern region of Taiwan. In the steel production industry, PM$_{2.5}$ is emitted during manufacturing as well as during the resuspension process at raw material storage sites. The iron ore sintering process for steel plants requires several materials, including iron ore, coal, limestone, coke, and recycled steel (Fernández-González et al., 2017). Contingent on the particle size distribution and chemical compositions, the concentrations of PM$_{2.5}$ differ among piles of raw materials. Recently, existing plants have expanded their production due to increased demand for steel (Cullen et al., 2012). Consequently, information published by the Taiwan Emission database system (TEDS 9.0) shows that the steel industry is responsible for 15.9% of the total PM$_{2.5}$ emitted nationwide.

There are both natural and anthropogenic sources of PM$_{2.5}$. However, emissions from the latter are believed to impact human health more adversely (Artiñano et al., 2003; Weijers et al., 2011). Major anthropogenic sources of fine particulates are associated with modern industrialization activities such as combustion, construction, manufacturing processes, and road dust (de la Campa et al., 2010; Benchrif et al., 2018). A comparison study between the pollution contributions from long-range transportation from neighboring China and those from local sources in cities within Taiwan established that the later was more dominant (Lai and Brimblecombe, 2021).

Previous analyses of PM$_{2.5}$ in Taiwan indicate several components, including, organic and elemental carbon, metals, crust elements, and water-soluble ions (Wang et al., 2020). Metallic components in PM$_{2.5}$ from industrial processes are not easily removed by pollution control devices. Consequently, they end up being discharged into the atmosphere (Ahmad et al., 2018). Recent surveys on the concentrations of metal components in particulate matter from six air quality monitoring stations including Nanzi, Zuoying, Sanmin, Qianjing, Qianzheng, and Xiaogang found significant concentrations of Al, Ca, Fe, and K. Overall, Fe was the most abundant metal, and Xiaogang station had the highest concentration (Lin et al., 2008; She et al., 2020).

Research on PM$_{2.5}$ is mostly focused on four processes: emission, transformation, transportation, and deposition (Pöschl, 2005; Choi et al., 2007; Shen et al., 2020). The latter is critical to the human population since it directly impacts their health. Deposition can be divided into wet and dry deposition. In wet deposition, the constituents of PM$_{2.5}$ are washed by precipitation culminating on solid surfaces such as land and vegetation or in water masses such as lakes, rivers, and oceans (Zhao et al., 2018). Through dry deposition, fine particulates wind up on surfaces or in human or animal lungs during inhalation (Zhang et al., 2014). Researchers have established reliable methods for sample collection and quantification of particulate matter. Loo et al. (1975) describe the application of a dichotomous sampler to collect both fine and coarse particulate matter. Receptor models for connecting the ambient air PM$_{2.5}$ and their emission sources have been developed and applied in many studies (Houck, 1991; Pace, 1991; Wienke et al., 1994).

Inhalation of PM$_{2.5}$ has been linked to several adverse health effects on the respiratory system including inflammation, cancer, and exacerbation of pre-existing respiratory conditions (Chalupa et al., 2004; Feng et al., 2016). Additional adverse health effects to the rest of the human body include the destruction of the nervous and circulatory systems, aggravation of diabetes mellitus, and anomalies in newborns (Nogueira, 2009; Zhao et al., 2013). Several PM$_{2.5}$ components including, Ca, black carbon, V, and Zn are associated with cardiovascular hospitalization while road dust, sea salt, aluminium, calcium, chlorine, black carbon, nickel, silicon, titanium, and vanadium are linked to respiratory complications (Bell et al., 2014). According to a study by Dejmek et al., (1999), the odds ratio for intrauterine growth retardation upon exposure of pregnant mothers to PM$_{2.5}$ is 1.26. Additionally, women with high pre-pregnancy exposure to PM$_{2.5}$ experience increased incidences of gestational diabetes mellitus (Monshi and Asgarani, 1999).

In the study of emission sources of particulates, resuspension, which is the removal of initially deposited material through a mechanical process such as wind, traffic, and soil cultivation, is rarely covered. Resuspended PM$_{2.5}$ can be transported to residential areas and rise to levels coinciding with the average breathing zone hence posing an inhalation exposure risk to residents. Herein, the chemical fingerprints of resuspended PM$_{2.5}$ from a raw material storage site and its contribution to ambient PM$_{2.5}$ in the surrounding area were investigated. A receptor model called chemical mass balance (CMB) was applied to estimate the contribution of different pollution sources to ambient PM$_{2.5}$. To the authors’ knowledge, a study on the contribution of resuspended particles from a raw material storage site to ambient air PM$_{2.5}$ has not been carried out before.
Therefore, this study can fill this research gap and provide useful insights into developing pollution control strategies for affected emission sources. Gathering comprehensive knowledge of emission sources and the chemical composition of PM$_{2.5}$ in the surroundings of the storage site is an appropriate step towards understanding the overall air quality to create proper health guidelines for the residents in the neighborhood.

## 2 METHODOLOGY

### 2.1 Sample Collection at the Raw Material Storage Field and its Surroundings

The area under investigation was separated into four sites, herein referred to as sites A, B, C, and D, as shown in Fig. 1. Site A encompasses the raw material storage zone. Sites B and C are densely populated areas next to the raw material storage site. Site D is also densely populated but further from the raw material storage site compared to sites B and C. Samples were collected at the raw material storage site for three days in each season to represent summer and winter. The sampling dates for the two seasons were the 5th, 8th, and 11th of August for summer and the 9th, 15th, and 27th of December for winter. Sampling dates were synchronized with the dates of PM$_{2.5}$ manual inspections for the EPA monitoring station. Additionally, a peripheral airborne particulate sampler (BGI PQ200) was applied to collect samples continuously for 24 hours.

A total of 15 samples were collected to represent each of the following: 5 piles of irons ore (iron ore 1–5), 5 piles of coal (coal pile 1–5), 3 piles of limestones (limestone 1–3), 1 pile each of sinter and coke. Special care was taken to adhere to the soil sampling method (S102.63B) announced by the National Institute of Environmental Analysis (NIEA). The topsoil samples were divided into composites and grab samples. Samples from different sampling points on the horizontal surface were mixed to obtain the average concentration for that specific area. A virtual grid was applied to collect the samples at the intersections as well as within the grids.

![Fig. 1. A plan view of the raw material storage site in the area under investigations and the distribution of the sampling points for ambient air PM$_{2.5}$ including A, B, C, and D.](image-url)
The soil samples were obtained and put in separate plastic lock bags. Reagents used for the treatment of the soil samples included phosphate-free detergent, distilled water, acetone, and n-hexane. Decontamination of the sampling equipment was performed between the collection of consecutive samples. A metal brush was used to remove debris from the equipment; a non-phosphorus detergent was applied on a brush, after which the device was brushed thoroughly before rinsing with distilled water and then with organic solvents (acetone or n-ethane). Lastly, the sampling device was rinsed with distilled water.

2.2 Re-suspension Tests

After obtaining the coarse and fine particulate matters from the sample (PM_{10} and PM_{2.5}) a resuspension test was performed. The resuspension chamber was carefully constructed and set to accurately represent natural wind. A resuspension system was used to administer tests to determine both the vertical and lateral translocation of fine particulates within an enclosed space. The re-suspension test was performed for the portion of the total suspended solids (TSP) that passed through a 400-mesh screen (D < 38 µm) and reached the bottom plate. The powder was collected in a stainless-steel dish and then loaded into a dry powder generator. An accelerated airflow rate of 16.7 L min^{-1} was applied to simulate natural wind in the 1 m square horizontal cross-section and 2 m high re-suspension chamber. A dichotomous sampler for PM_{10} and PM_{2.5} was used to collect coarse and fine particulate matter. A dichotomous sampler was placed under the re-suspension chamber. The total time for re-suspension and sample collection was 5 minutes.

2.3 Weighing and Conditioning of the Samples

A microbalance was used for the 24 filter paper samples. The inspection was done for each sampling filter paper using a lamp and a magnifying glass to confirm the integrity of the filter paper. The following factors were considered to compromise the integrity of the filter paper: breakage, indentations, dents, scratches, fiber, and particle contamination. Care was taken to preserve the integrity of the filter paper during unpackaging, weighing, conditioning, and clipping. The filter paper was then placed in a conditioning environment for 12 hours. This was succeeded by fine weighing. The steps for the conditioning and fine weighing were repeated until the difference in weight was constant and less than 5 µg. The average of the weight before and after conditioning was taken as the weight after sampling. The formula for the calculation of PM_{2.5} concentration is as follows:

\[
PM_{2.5} \text{ Concentration} = \frac{W_f - W_i}{V} \tag{1}
\]

where PM_{2.5} concentration is in µg m^{-3}. W_i and W_f represent the masses of the filter paper in µg before and after sampling, respectively. V is the total volume of air in m^3.

To collect samples for ambient air PM_{2.5}, blank filter papers were exposed to the atmosphere for 24 hours with a sample volume of 24 m^3. The detection limit was 2 µg m^{-3}.

2.4 Analysis of Metal Compositions, Water-soluble Ion Components, and Carbon Content

Evaluating the chemical fingerprints for PM_{2.5}, involved establishing their metal components, water-soluble ion concentrations, and carbon content. A total of 15 samples were tested for 31 metal elements including Li, Be, Na, Mg, Al, K, Ca, Ti, V, Cr, Mn, Fe, Co, Cu, Zn, Ga, As, Se, Rb, Sr, Ag, Cd, In, Sn, Cs, Ba, Ti, Pb, U, Ni, and Si. Further analyses were done for 4 anionic species including SO_4^{2-}, NO_3^-, Cl^-, F^-, and 5 cationic species including Na^+, K^+, Ca^{2+}, Mg^{2+}, NH_4^+. The concentrations of elemental and organic carbons (EC and OC) were evaluated.

The metal composition analyses for PM_{2.5} were performed using the other half of the Teflon filter paper. The microwave digestion pretreatment process established by the Environmental Protection Administration (EPA) was applied. First, calibration was done using five standard solutions to produce a calibration line. The relative error value of the calibration line was set to be within ±10%. Otherwise, the operating condition of the instrument must be checked or
maintenance must be performed and another calibration line must be developed. After calibration, the samples were put into ICP/MS for analysis. The blank sample could not exceed double the MDL. A blank sample was prepared and analyzed after every 10 samples. Care was taken to ensure a recovery rate of 80–120%. Furthermore, one sample was tested twice in every batch of 10 samples. The relative difference between the two measurements could not exceed 20%. Quantification of the metal composition in the PM\textsubscript{2.5} samples was performed using high-resolution inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500A). The calibration line was checked using a standard solution whereby the acceptable absolute error was required to be less than 10%.

Sample pretreatment involved shaking half of each collected Teflon filter using ultrasonic vibrations for 90 minutes in 10 mL of deionized water. After extraction, the 10 mL was poured into a syringe and filtered with a 0.45 µm membrane to remove particles. Quantification of the water-soluble anions was performed using an Ion Chromatograph (IC) (Dionex, Model DX-120) series conductivity detector. Analysis of the anions was performed with the ASRS-ULTRA suppressor, Ion Pac AS4A-SC column, and Na\textsubscript{2}CO\textsubscript{3}/NaHCO\textsubscript{3} eluent. In addition, cations were evaluated using a CSRS-ULTRA suppressor, an Ion Pac CS12 column, and 0.1 M of H\textsubscript{2}SO\textsubscript{4} eluent. The concentrations in the blank samples fell below the limit of detection (LOD) for all ions. Therefore, deductions were unnecessary for the calibration data.

An elemental analyzer (Elementar Vario Mirco Cube), with the AS 200 type Autosampler and DP 700 integrator for were used to quantify both the EC and OC. The equipment’s precision and stability are shown in Table 1, and its operating conditions are provided in Table 2. A quartz filter paper was first placed in the thermal chamber and heated to 900°C to remove any available carbon before the chemical analysis. Analysis of the carbon content in PM\textsubscript{2.5} involved instantaneous dynamic oxidation. The sample’s chemical bonds were broken in the quartz reaction tube. It was rapidly oxidized to CO\textsubscript{2} after which it was reduced using Cu and then passed through the separation column for detection using a thermal conductivity detector (TCD). The organic carbon content was obtained based on the difference between the total carbon (TC) and EC.

2.5 Chemical Mass Balance
A chemical mass balance was applied for the nearby emission sources, including the raw material storage site, background soil dust, traffic dust, mobile sources, steel industry, various industrial boilers, electric arc furnaces, coke ovens, sintering furnaces, power plants, and incinerators, as shown in Table 1. Criteria for the selection of emission sources included a chi-square value of less than 4.0, press normalization R\textsuperscript{2} > 0.8, a t-statistic > 2.0, where the ratio of the calculated to the measured value was required to be 0.8–1.2, the residual and uncertainty ratio was required to be –2.0–2.0, and unquantified and similar clusters should preferably be absent.

The theory of the receptor model was based on the similarities between the chemical compositions of emission sources and those of the receptor sites. It was supported by the principle of conservation of mass and statistics. The pollutant concentration at the receptor point was the linear summation of the pollution sources. The main simplifying assumption is that

| Carrier gas | Accuracy | Precision |
|-------------|----------|-----------|
| Helium      | ≤ 0.3%   | ≤ 0.2%    |
| Argon       | ≤ 0.5%   | ≤ 0.4%    |

| Process                                      | Set conditions |
|----------------------------------------------|----------------|
| Oxidation furnace temperature (°C)           | 1020           |
| Reduction furnace temperature (°C)           | 500            |
| Carrier gas flow rate (mL min\textsuperscript{-1}) | 100            |
| Sample delay time (s)                        | 8–15           |
| Total execution time (min)                   | 9–12           |
there were no chemical reactions between different chemical species from the emission sources. A fingerprint database required for the chemical mass balance receptor model was established before the CMB8.2 receptor model was applied to estimate the respective percentage contributions of the PM$_{2.5}$ sources to ambient air PM$_{2.5}$.

3 RESULTS AND DISCUSSION

3.1 Ambient Air PM$_{2.5}$ Concentration during Summer and Winter

Kaohsiung has a tropical climate, where rainfall is experienced 5–9 months of the year. The average annual temperature is approximately 24.3°C while the annual rainfall is approximately 1738 mm (Chen and Chen, 2003). During summer, warm, and humid air masses flow from the southern side of the country, which is mostly ocean. Under rainfall and typhoon conditions, the relative humidity ranges between 69.85 and 74.76%. Wind flow over the Kaohsiung region leads to lower relative humidity in winter, which ranges between 67.51 and 70.60%.

From 2005 to 2017, the EPA compared manually calculated hourly concentrations for PM$_{2.5}$ to those from an automatic monitoring station. The PM$_{2.5}$ annual average concentration for 13 years is presented in Fig. 2. There has been a steady decrease in the concentration of PM$_{2.5}$ in Xiaogang

![Graph showing PM$_{2.5}$ annual average concentrations for 13 years.](image)

**Fig. 2.** Annual values for (a) ambient air PM$_{2.5}$ concentrations and (b) PM$_{2.5}$/PM$_{10}$ ratios in Daliao and Xiaogang stations for the years 2005 to 2017.
station from 55.2 μg m\(^{-3}\) in 2005 to 36.2 μg m\(^{-3}\) in 2017. Similarly, in Daliao, the concentrations were 50.4 and 3.02 μg m\(^{-3}\) in the respective years. The reduction over the period was 34.4% and 40.1% for Xiaogang and Daliao stations, respectively. The Taiwan EPA has put control measures to reduce the emission for air pollutants including PM\(_{2.5}\), PM\(_{10}\), SO\(_x\), NO\(_x\), and NMHC from both mobile and stationary sources. This could explain the trend for PM\(_{2.5}\) in Xiaogang and Daliao for the years 2005–2017. The variations in the PM\(_{2.5}\) concentration in winter and summer can be explained by the north and south winds, which are dominant during winter and summer, respectively (Tsai et al., 2003). During winter, the dominance of the north wind reduces the possibility of vertical diffusion of PM\(_{2.5}\) in the mixed layer.

The PM\(_{2.5}\) concentration in summer ranges from 13.7–18.0 μg m\(^{-3}\) as shown in Fig. 3. This is below the 24-hour standard in Taiwan. Water-soluble ions account for the largest proportion in the total PM\(_{2.5}\) mass, with an average of 7.4 and 22.7 μg m\(^{-3}\) in summer and winter respectively of the total PM\(_{2.5}\) mass. In summer, the carbon composition by mass exceeded the contribution from other chemicals in site C only. However, in winter, the concentration of other chemicals exceeded that of the carbon content in all sites. The average carbon content was 11.8% and 34.2% in summer and winter, respectively. The mass composition in winter exceeded that of summer because of lower wind recirculation, which limited dilution rates (Virtanen et al., 2006; Tseng et al., 2019). The average mass concentrations of EC and OC in the atmospheric PM\(_{2.5}\) for the summer and winter seasons for the four sites under investigation are shown in Fig. 4. The seasonal average OC content ranged between 1.2 and 3.0 μg m\(^{-3}\) in summer while the range for winter is between 5.6 to 6.2 μg m\(^{-3}\). The range of EC content was wider in the summer than in the winter. In summer, it ranged from 0.6 to 1.8 μg m\(^{-3}\) while in winter it ranged from 1.7 to 2.2 μg m\(^{-3}\). EC is emitted from the incomplete combustion of fuels while OC results from the degradation of carbonaceous materials. The dominance of either EC or OC indicates the major source of carbonaceous aerosols.

Carbonaceous aerosols form a major component of PM and play an important role in atmospheric chemistry. Investigations carried out in the past found that the OC/EC in the region ranged between 2.32 and 2.76 in periods when the sea and land breeze interacted (Fang et al., 2008). Herein, the OC/EC ratio ranged between 1.5 to 4.0. This indicates that the predominant sources of carbon content in PM\(_{2.5}\) are photochemical reactions, industrial, and combustion processes. This is in line with previous studies on carbon compositions in PM\(_{2.5}\) in the Kaohsiung.
Fig. 4. Mass of elemental and organic carbon in PM$_{2.5}$ and the ratios of OC/EC for different sites during summer and winter.

metropolitan area. For instance, Huang (1991) found that carbon content accounted for about 22.1% of the ambient air PM$_{2.5}$, on average, and the average OC/EC ratio was 2.6. The OC/EC ratio can be applied to determine the intensity of secondary organic aerosol formation. The critical value is OC/EC $=2.2$ or OC/TC $=0.67$. Carbon component analysis methods include thermal decomposition method (TM), solvent extraction (SEM), thermal manganese oxidation method (TMO), thermo-optical reflection method (TOR) optics, and thermo-optical transmission method (TOT). For sites A, B, and C, during summer the OC/EC value is less than 2.2 indicating a low potential for the formation of secondary organic aerosols (SOA).

In summer, the OC/EC for Site D greatly exceeded that found at the other sites. This is because site D is a rural area, and therefore, the degradation of carbon-containing products such as vehicle tires and vegetation, leads to more carbonaceous aerosol emissions compared to that derived from incomplete combustion of carbon-containing fuels.

At other times, the OC/EC ratio in all sites during winter exceeded 2.2, indicating a high potential for the formation of SOA (Pandis et al., 1992; Cao et al., 2008). Overall, carbonaceous PM$_{2.5}$ are mainly emitted from combustion sources upwind of the site under investigation, including mobile sources and fixed sources, such as the petrochemical industry, boilers, steelmaking processes, and secondary derived aerosols.

3.2 Chemical Speciation of Ambient Air PM$_{2.5}$

The concentrations of PM$_{2.5}$ and their speciation at the sites under investigation for two seasons are presented in Fig. 3. The concentration during winter exceeded that of summer for all sites. Further, it can be observed that metals had the lowest concentrations in all sites and for all seasons. The most important category of chemicals is water-soluble ions. Site B has the highest metal composition while site D had the lowest concentration of metals and water-soluble ions. This phenomenon can be attributed to the proximity of the respective sites to the raw material storage site and the dominant wind directions (Cohen et al., 2011).

3.3 Metal Composition

The profile for metals during summer is shown in Fig. 5(a), Site D had the highest concentrations
of Na and K. Al was lacking at all the sites except site A. Ca is the only metal of natural origin that was absent at all sites. Mn was present at site B only while Zn was present in all sites. Other metals of anthropogenic origin were absent at all the sites. During winter, all metals of natural origin were present at all sites except Al, which was absent at site A. Other than V, Mn, Zn, and Pb, other metals of anthropogenic origin were absent at all sites under investigation. The metallic elements in the PM$_{2.5}$ mostly exist in the solid phase and are distributed in the environment through atmospheric airflow or gravity sedimentation (Moreno et al., 2004). During summer, the metal composition accounted for an average of 5.7% of the total mass of PM$_{2.5}$ at all the sites under investigation as shown in Fig. 3. Specifically, Na had the highest contribution, which were 35.2%, 25.1%, 45.9%, and 50.1% for sites A, B, C, and D, respectively. The predominance of Na in summer can be attributed to the sea salt droplets occasionally blown by the westerly wind from the sea to the land surface. It is hypothesized that the main source of Fe is re-suspension from the iron ore piles in the raw material storage site. Besides, during steel production, ferromanganese (Fe-Mn), ferrosilicon (Fe-Si), and aluminium are added to remove excess oxygen. The background soil of surrounding areas is rich in crustal elements such as Si, K, Mn, and Mg. The northwest wind brings crustal elements and sea salt droplets. This

![Fig. 5. PM$_{2.5}$ metal composition for sites A, B, C, and D during (a) summer (b) winter.](image-url)
is believed to cause high concentrations of these crustal elements in PM$_{2.5}$. The proportion of Zn in the total mass of the metals ranged between 6.39% and 10.5%. The main source is believed to be the mobile sources in the sites under investigation (Lin et al., 2020; Shen et al., 2020).

### 3.4 Water-soluble Ions

In Fig. 6, the percentage composition of individual water-soluble ions is presented. During winter, SO$_4^{2-}$ was the most important component at all the sites. In summer, NO$_3^-$ was the most important composition. The percentage of SO$_4^{2-}$ was almost constant for both seasons. In all seasons and for all sites, SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ contributed about 90% of the total water-soluble ions. The percentage composition of NH$_4^+$ in summer exceeded that of winter for all sites. In winter, the water-soluble ionic component, SO$_4^{2-}$, accounted for about 70% of the total water-soluble ions by mass, followed by NO$_3^-$ with an average of 10%, and NO$_2^-$ with an average of 5%, ranking third, as shown in Fig. 3. The predominance of SO$_4^{2-}$ and NO$_3^-$ implies that anthropogenic sources have a higher contribution to ambient air PM$_{2.5}$ than natural sources. Information on the contribution of primary and secondary emission sources to ambient air PM$_{2.5}$ can be obtained from the ratios of SO$_4^{2-}$ to NO$_3^-$. Further, as seen in Table 4, the percentage composition of SO$_4^{2-}$ was significantly higher in pile 1 compared to the other piles. This can be used to paint a picture of the transformation phenomena of sulfate and nitrate ions (Seinfeld et al., 1984). Fine nitrate has been associated with aerosol acidity and moisture content. Additionally, the oxygen isotopic anomalies of SO$_4^{2-}$ and NO$_3^-$ in PM$_{2.5}$ are transferred to O$_3$ during the oxidation process of SO$_2$ and NO$_x$.

The I and J values for summer and winter for the four sites are presented in Fig. 7. For all sites, the I and J values exceeded the threshold during winter. On the other hand, they fell below the threshold during winter. The main constituents of secondary derived inorganic salts in the atmosphere NH$_4^+$, NO$_3^-$ and SO$_4^{2-}$, easily exist in the air as NH$_4$NO$_3$, NH$_4$HSO$_4$, (NH$_4$)$_2$SO$_4$, and other types of salts. Previously, it was pointed out that is the equivalent concentration ratio of ammonium and sulfate (Seinfeld et al., 1998). When the value of I ≥ 2, it implies that SO$_4^{2-}$ has been neutralized and mainly exists as (NH$_4$)$_2$SO$_4$. On the other hand, if I < 2 then only part of it exists as (NH$_4$)$_2$SO$_4$, and free sulfate ions still exist in the atmosphere.

![Fig. 6. Percentage of water-soluble ions in PM$_{2.5}$ for the seasons of winter and summer and at sites A, B, C, and D.](https://aaqr.org)
Fig. 7. I and J values for PM$_{2.5}$ and the thresholds in sites A, B, C, and D for summer and winter.

\[ I = \frac{NH_4^{+\text{eq}}}{SO_4^{2-\text{eq}}} \]  

(2)

The 24-hour ratio of NH$_4^+$ to NO$_3^-$ and SO$_4^{2-}$ ratio is represented by the J value (Chu, 2004). When $J < 1$, then NH$_4^+$ is insufficient to neutralize NO$_3^-$ and SO$_4^{2-}$ and therefore, they are acidic. On the other hand, if $J > 1$, NH$_4^+$ is in surplus, and NO$_3^-$ and SO$_4^{2-}$ are basic.

\[ J = \frac{NH_4^{+\text{eq}}}{2 \times SO_4^{2-\text{eq}} + NO_3^{eq}} \]  

(3)

Herein, the value of I ranged between 0.81 and 1.30 while the J value ranged between 0.37–0.60, $I < 2$, $J < 1$, indicating the presence of NO$_3^-$ and SO$_4^{2-}$ and an acidic atmosphere.

From Table 4, the percentage composition of SO$_4^{2-}$ was significantly higher in pile 1 compared to the other piles. Further, water-soluble ions formed the most important component in the limestone piles especially, SO$_4^{2-}$, Ca$^{2+}$. All these led to lower I and J ratios. While the percentage composition of SO$_4^{2-}$ in the total PM$_{2.5}$ coal pile 1 exceeded 50%, that in the other coal piles barely reached 10%. This suggests that the contribution of coal piles to secondary aerosols was the highest in coal pile 1.

3.5 Carbon Content

From Fig. 4, the mass of organic carbon exceeded that of elemental carbon at all the sites under investigation and during all seasons. The OC/EC ratios for the four sites are presented in Table 3, where sites A and D had the highest values during summer. Carbon was a minor component in the iron ore pile, as shown in Table 4. Organic carbon exceeded elemental carbon in all piles except the third iron pile and this might indicate contamination. Unlike in the iron ore piles, carbon was the major component in the coal piles. The percentage composition of elemental carbon exceeded that of organic carbon in all piles except the second pile. The percentage composition of elemental carbon was more than 20% except in the first coal pile, where it was approximately 2%. The percentage composition of carbon in the coal pile 1 was
attributed to high sulfate concentrations. The high sulphate content is hypothesized to originate from leaching of overlying steel slags.

As shown in Table 4, the carbon content was the second most dominant composition in the limestone piles with OC more abundant than EC (Tseng et al., 2019). Only three metal types were present in significant percentage compositions in the three stone piles including Mg, Ca, and Fe. The other metal compositions were either insignificant or non-existent. As seen in Table 4, EC formed a significant percentage composition of the sinter pile, at about 21%. The rest of the compositions comprised less than 5% of the percentage composition. In the coke pile, OC exceeded EC, unlike in the sinter pile. The carbon component accounted for 17.4% of the total

### Table 3. The concentrations of OC and EC and the OC/EC.

|          | Site A |           | Site B |           | Site C |           | Site D |           |
|----------|--------|-----------|--------|-----------|--------|-----------|--------|-----------|
|          | Summer | Winter    | Summer | Winter    | Summer | Winter    | Summer | Winter    |
| OC (µg m⁻³) | 3.00   | 6.20      | 1.20   | 5.80      | 2.30   | 5.60      | 2.30   | 5.90      |
| EC (µg m⁻³) | 1.80   | 1.70      | 0.80   | 2.30      | 1.20   | 2.20      | 0.60   | 1.80      |
| OC/EC    | 1.70   | 3.61      | 1.48   | 2.56      | 1.93   | 2.50      | 4.04   | 3.28      |

### Table 4. Percentage masses of individual components in each pile of raw material.

| Raw materials | Iron piles | Coal piles | Limestone piles | Coke pile | Sinter pile |
|---------------|------------|------------|-----------------|-----------|-------------|
|               | IP1 (%)    | IP2 (%)    | IP3 (%)         | IP4 (%)   | IP5 (%)     |
| Na            | 2.42       | 0.51       | 0.24            | 0.17      | 0.39        |
| Mg            | 0.82       | 0.21       | 0.21            | 0.50      | 0.27        |
| Al            | 7.54       | 0.46       | 1.39            | 2.07      | 2.06        |
| Si            | 2.85       | 0.85       | 1.79            | 1.72      | 1.84        |
| K             | 3.26       | 0.55       | 0.24            | 0.13      | 0.40        |
| Ca            | 1.05       | 0.89       | 0.14            | 0.22      | -           |
| Ti            | -          | -          | -               | -         | -           |
| V             | -          | -          | -               | -         | -           |
| Mn            | -          | -          | 0.42            | 0.20      | 0.17        |
| Cr            | -          | -          | -               | -         | -           |
| Fe            | 46.3       | 14.8       | 41.2            | 44.4      | 42.6        |
| Co            | -          | -          | -               | -         | -           |
| Ni            | -          | -          | -               | -         | -           |
| Cu            | -          | -          | -               | -         | -           |
| Zn            | -          | -          | -               | 0.50      | -           |
| As            | -          | -          | -               | -         | -           |
| Se            | -          | -          | -               | -         | -           |
| Sn            | 0.07       | 0.02       | 0.01            | 0.01      | 0.32        |
| Cs            | -          | -          | -               | -         | -           |
| Pb            | -          | -          | 0.17            | -         | -           |
| F⁻            | -          | -          | -               | -         | 0.11        |
| Cl⁻           | 2.27       | 0.65       | 0.51            | 0.48      | 0.48        |
| NO₂⁻          | 1.27       | 0.23       | 0.34            | 0.27      | 0.31        |
| SO₄²⁻         | 22.7       | 5.83       | 4.63            | 4.19      | 6.59        |
| Na⁺           | -          | -          | -               | -         | 0.11        |
| NH₄⁺          | -          | -          | -               | -         | 0.34        |
| K⁺            | -          | -          | -               | -         | 0.58        |
| Ca²⁺          | 2.39       | 1.20       | 0.40            | -         | -           |
| Mg²⁺          | -          | -          | -               | -         | 2.54        |
| OC            | 7.80       | 7.31       | 9.05            | 7.60      | 4.67        |
| EC            | 2.40       | 0.95       | 11.3            | 0.00      | 0.08        |

Note: IP represents Iron piles; CP represents Coal piles; LP represents Limestone piles.
mass, where OC exceeded EC. The pollution source may have come from organic carbon derived from photochemical reactions or pollution emissions such as industrial combustion (Hays et al., 2005).

3.6 Chemical Fingerprints for the PM$_{2.5}$ in the Raw Material Piles

The chemical profiles of different types of iron ore piles are presented in Table 4. As anticipated, Fe content in the iron pile ranged from 14.8 to 46.3% and hence was the most abundant species in all five iron piles. Coal pile 1 had a significant amount of Na, Al, Si, K, and Fe. The percentage composition of all metals was less than 10% in all five iron piles, except for Fe in core pile 1, which was 14.4%. The percentage composition of metals in the other piles was either non-existent or insignificant. Additionally, the iron piles had water-soluble ions and other crustal components. Among the water-soluble ions, SO$_4^{2-}$ was present in the highest concentration. In iron pile 1, it was 22% while in the other piles it ranged between 4% and 6%. Aluminium was present in the highest percentage in iron pile 1, while in the other piles, it was less than 2%. The crustal elements included Na, Mg, Al, K, and Si, while the water-soluble ions included four anionic species such as SO$_4^{2-}$, NO$_3^-$, Cl$^-$, F$^-$, and 5 cationic species including Na$^+$, K$^+$, Ca$^{2+}$, Mg$^{2+}$, NH$_4$+. Among the water-soluble ions, SO$_4^{2-}$ was the most abundant water-soluble ion accounting for (4.2–22%) in all piles. OC content was highest in iron pile 1 at 11.3% while in the other iron piles, it ranged between 4.7–9.1%. This was suspected to contribute to the ambient air PM$_{2.5}$ through re-suspension especially during the sea-land breeze associated with periods with no precipitation in winter (Cao et al., 2008).

The composition of coal pile 1 as presented in Table 4 differed from the other coal piles. Its main component was SO$_4^{2-}$, which contributed 55.3% of all water-soluble ions. The metal composition was 31.6% while that of carbon was 6.45%. The carbon content was high in the other four coal piles. In coal pile 2, organic carbon (28%) exceeded elemental carbon (20.1%) unlike in the other coal piles (Moreno et al., 2004; Fang et al., 2008). The elemental carbon ranged between 25.4–41.9% while the organic carbon ranged between 9.06–10.8%. Water-soluble ions accounted for between 11.4–12.0%, where SO$_4^{2-}$ was responsible for 9.62–11.4%. The metal composition was 6.32–8.51%, and the contribution from crustal elements was insignificant. As shown in Table 4, Ca$^{2+}$ and Fe comprised the highest percentage composition of the sinter pile, with about 20% each (Moreno et al., 2004; Fernández-González et al., 2017). One possible reason for the high Ca$^{2+}$ was the contamination of the sinter pile with dust from the limestone pile, which naturally has high concentrations of Ca$^{2+}$. Additionally, within an integrated steel plant, the presence of oxygen species during reduction and refining processes causes the expulsion of low molecular weight metals in the form of fine particulates. Further, the leaching of steel slags for long periods could also explain the high Ca$^{2+}$ concentration.

In the limestone pile, the three piles differed significantly in terms of chemical composition. In limestone pile 1, total carbon accounted for 35.9% while metals accounted for 17%. Specifically, elemental carbon was 10.6%, and organic carbon was 25.3%. For the metal composition, magnesium and calcium accounted for 9.86% and 4.19% respectively. Water-soluble ions accounted for 13.2%, where the individual contributions from Ca$^{2+}$, SO$_4^{2-}$, and Mg$^{2+}$ were 6.64%, 3.86%, and 2.54%, respectively. In limestone pile 2, water-soluble ions were the main chemical composition where Ca$^{2+}$ and SO$_4^{2-}$ accounted for 18.9% and 4.93% respectively. Metals and carbonate accounted for 16.4% and 15.6% respectively. Specifically, organic carbon accounted for 13.9%. Stone pile 3 had water-soluble ions as the main components, just like stone pile 2. They accounted for 47.7% of the total chemical composition, with SO$_4^{2-}$ accounting for 39.5%. Carbon accounted for 20.2%, and metals contributed 3.13%. As anticipated, Ca$^{2+}$ was among the major components in limestone and could have contributed Ca$^{2+}$ in the ambient air PM$_{2.5}$ after resuspension (Monshi and Asgarani, 1999).

The main chemical component in the coke pile was carbon, which accounted for 27.4%. Water-soluble ions and metal accounted for 21.7% and 11.6%, respectively. Specifically, SO$_4^{2-}$, K$^+$, and Ca$^{2+}$ contributed 11.4%, 6.58%, and 3.18% respectively whereas Fe, Si, and Al accounted for 6.07%, 4.19%, and 0.89%, respectively. In the case of the sinter pile, the metal component was the most important, accounting for 37.4%. Specifically, the contributions from Fe, Ca, K, Si, Al, Na, and Mg were 18.6%, 5.69, 2.51, 2.24, 2.16, 1.91, and 1.08%, respectively. PM$_{2.5}$ from the coal mine was less dense compared to the other raw material piles and hence had a wider range of diffusion. The PM$_{2.5}$ from limestone and iron ore were characterized by high density, which...
resulted in smaller diffusion distances from the raw material piles to adjacent locations (Moreno et al., 2004; Cohen et al., 2011).

### 3.7 Contribution of Various Pollution Sources from the Receptor Model (CMB)

The results for the seasonal variations and fingerprint data for pollution sources are summarized in Table 5. Based on the compositional analysis, the main sources of PM$_{2.5}$ in the neighborhood of the raw material storage field were hypothesized to be petrochemical plants, boilers, background soil dust, secondary sources of sulfates and nitrates, and combustion sources. During winter, the main pollution sources of PM$_{2.5}$ for sites A, B, and C was the petrochemical industry where it accounted for 20.6%, 22.8%, and 17.8% of the total PM$_{2.5}$ mass, respectively. In site D, secondary nitrates accounted for the highest PM$_{2.5}$ mass at (19.3%). Secondary nitrates (NO$_x$) were mostly from mobile sources and other combustion sources upstream (Yang et al., 2019). Ammonium released from fertilizers and animal feeds is also believed to have contributed to the high concentrations of nitrates and sulfates in PM$_{2.5}$ especially in site D. The petrochemical industry was dominant in sites A, B, and C because of their proximity to the petroleum product processing plant. In addition to nitrates and sulfates, the boiler and background soil were dominant ambient PM$_{2.5}$ sources in site D due to the dominating influence of the land-sea breeze and lack of rainfall (Lee et al., 2008; Yuan et al., 2018).

The average contributions of the major pollution sources during summer for site A included sulfate (28.70%), traffic sources (16.36%), and soil dust (13.38%). Those for site B included sulfate (43.47%) and traffic sources (13.74%). For site C, they were derived sulfate (36.95%), traffic sources (14.19%), and derived organic carbon (13.85%), and those for site D included sulfate (35.34%), soil dust (27.44%), and organic carbon (14.91%). Overall, secondary aerosols were the predominant form of PM$_{2.5}$ in the area under investigation. Site A was predominantly affected by the west and northwest winds. Therefore, the petrochemical plant located on the west side of the raw material storage site was an important source of PM$_{2.5}$. Through CMB, it was established that emissions from the petrochemical plant, boiler, and incinerator flow towards the raw material storage site led to an increase in the ambient air sulfate concentrations (Hu et al., 2019). At site B, during the summer, the station is affected by the northwest west wind, and south wind, bringing sea droplets to the station. Site C was greatly influenced by sea salt droplets under the influence of sea and land breezes. Site D had no obvious pollution source during summer except mobile sources, which led to amplified nitrate composition in the PM$_{2.5}$. At site D, the crustal composition, as well as the contribution from mobile sources, were the predominant components of PM$_{2.5}$. Additionally, this site was adjacent to the petrochemical plant and therefore, under the influence of pollutants transported by the west, southwest, and northeast winds as well as the land-sea breezes (Cohen et al., 2004; Fang et al., 2018). The dominance of sulfates and secondary OC in summer for all sites indicates active biogenic sources in the neighborhood.

### Table 5. Percentage contributions of PM$_{2.5}$ sources to the surrounding of the raw material storage site according to the chemical mass balance

| Pollution sources | I (%) | II (%) | III (%) | IV (%) | V (%) | VI (%) | VII (%) | VIII (%) | IX (%) | X (%) | XI (%) | XII (%) | XIII (%) | XIV (%) | XV (%) | XVI (%) | XVII (%) | XVIII (%) | IX (%) | XX (%) | Total (%) |
|-------------------|------|-------|---------|--------|------|-------|---------|----------|-------|------|-------|---------|----------|-------|-------|--------|----------|--------|-------|-----------|
| **Winter**        |      |       |         |        |      |       |         |          |       |      |       |         |          |       |      |        |          |        |       |           |
| Site A            | 3.2  | 0.0   | 0.0     | 0.0    | 20.6 | 0.0   | 0.0     | 0.0      | 6.7   | 5.8  | 4.2   | 0.0     | 0.0      | 0.0   | 0.0  | 0.0    | 2.3      | 2.2    | 12.4 | 18.5     | 0.0      | 0.9    | 23.3  | 100       |
| Site B            | 4.4  | 7.4   | 0.0     | 0.0    | 22.8 | 0.0   | 0.0     | 0.0      | 5.1   | 0.8  | 6.2   | 0.0     | 0.0      | 0.0   | 0.0  | 0.0    | 2.4      | 1.6    | 10.6 | 17.0     | 0.0      | 0.7    | 20.9  | 100       |
| Site C            | 1.5  | 2.8   | 2.8     | 0.0    | 17.8 | 0.0   | 0.9     | 4.9      | 2.5   | 2.5  | 3.2   | 0.0     | 0.0      | 0.0   | 0.0  | 0.0    | 3.2      | 3.6    | 13.9 | 17.4     | 0.0      | 0.0    | 25.6  | 100       |
| Site D            | 0.0  | 0.0   | 0.0     | 0.0    | 0.0  | 2.8   | 0.0     | 18.0     | 7.0   | 0.0  | 0.0   | 0.0     | 0.0      | 0.0   | 0.0  | 0.0    | 11.8     | 1.5    | 13.2 | 19.3     | 8.9      | 1.1    | 16.4  | 100       |
| Site A            | 0.3  | 3.1   | 0.0     | 9.6    | 0.0  | 0.0   | 16.4    | 3.6      | 0.0   | 0.0  | 0.0   | 0.0     | 0.0      | 13.4  | 3.8  | 28.7   | 2.6      | 9.2    | 0.0   | 9.4      | 100      |       |       |           |
| Site B            | 0.0  | 0.0   | 0.0     | 3.4    | 0.0  | 0.0   | 13.7    | 3.6      | 4.1   | 0.0  | 0.0   | 0.0     | 0.0      | 0.0   | 1.2  | 4.9    | 43.5     | 5.0    | 8.6   | 0.0      | 12.1     | 100    |       |           |
| Site C            | 0.0  | 0.0   | 0.0     | 0.0    | 0.0  | 0.0   | 14.2    | 0.0      | 0.0   | 0.0  | 0.0   | 0.0     | 0.0      | 0.0   | 18.9 | 6.8    | 36.9     | 3.0    | 13.8 | 0.0      | 6.4      | 100    |       |           |
| Site D            | 0.0  | 0.0   | 0.0     | 0.0    | 0.0  | 0.0   | 0.0     | 0.0      | 0.0   | 6.5  | 0.0   | 0.0     | 0.0      | 0.0   | 0.0  | 0.0    | 27.4     | 4.7    | 35.3 | 3.6      | 14.9     | 0.0    | 7.5   | 100       |

Note: I-Steel plant’s converter, II-Coke Furnace, III-Sinter Furnace, IV-Electric Arc Furnace, V-Petrochemical Industry, VI-Incinerator, VII- Boiler, VIII-Traffic source, IX-Iron ore, X-Coal ore, XI-Stone ore, XII-Coke ore, XIII-Sinter ore, XIV-Soil dust, XV-Marine aerosol, XVI-Secondary sulfate, XVII-Secondary nitrate, XVIII-Secondary OC, IXX-Secondary EC, XX-Others.
4 CONCLUSIONS

The main goal of these investigations was to establish the contribution of a raw material storage site to ambient air PM$_{2.5}$ in the surrounding area. The major chemical compositions of resuspended PM$_{2.5}$ from iron ore, coal, limestone, coke, and sinter piles were successfully investigated. The chemical composition differed among the various piles. Furthermore, there were slight variations among piles with the same key compositions. The variations in the concentration of PM$_{2.5}$ between the two seasons indicated that prevailing weather conditions, such as the dominant wind direction, temperature, and rainfall, significantly affected the PM$_{2.5}$ concentrations. Lower wind intensities in winter as compared to summer led to inadequate circulation of air masses and hence elevated concentrations of PM$_{2.5}$ within the site under investigation during winter. In the summer, the PM$_{2.5}$ concentration ranged from 13.7–18 µg m$^{-3}$, and water-soluble ions accounted for 54.2% of the total. The main ambient PM$_{2.5}$ components were SO$_4^{2-}$ (73.6%), NH$_4^+$ (13.7%) and NO$_3^-$ (7.45%). In winter, the PM$_{2.5}$ ranged between 44.7 and 48.0 µg m$^{-3}$, and water-soluble ions accounted for 49.2% of the PM$_{2.5}$ total mass. The main components were NO$_3^-$ (36.5%), SO$_4^{2-}$ (30.1%), and NH$_4^+$ (24.3%). In the winter, the relative humidity (above 70%) and the low-temperature environment led to high ambient air NO$_3^-$ concentrations. The site selected for material storage has a more obvious impact on the regions downwind. The OC/EC ratios indicated that the predominant sources for ambient air PM$_{2.5}$ were photochemical, industrial, and combustion processes. However, the surroundings of the raw material storage site are affected more by other notable pollution sources, including petrochemical plants, soil dust, derived sulfates and nitrates, and mobile sources. The dominance of secondary organic aerosols raises concerns about the importance of dealing with the emission of secondary aerosol precursors.

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