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Image based analytical approaches for study of particulate matter (PM) in air

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Particulate matter (PM) stands as a predominant pollutant in developing countries, demanding effective source identification and remediation strategies. This review centers on the scanning electron microscopy (SEM) image-based methodology for PM analysis, particularly emphasizing the passive technique of utilizing plant leaves for PM capture. The SEM-image-based approach serves as a powerful tool for unraveling the morphological characteristics of PM, crucial for source identification. Additionally, SEM, when equipped with energy dispersive spectroscopy (EDS), enables chemical and mineralogical characterization, providing insights into the origin of PM. The first part of the review describes the plant as the best bio-sampler for PM. In this context, removal of PM from the environment through plant-based interventions is described. Subsequently, the application of SEM for size-based analysis using ImageJ and morphological analysis for source identification of PM is detailed. Following this, the PM chemical and mineralogical composition for source identification are described based on EDS analysis. Image-based techniques play a pivotal role in selecting the most effective plant species for PM removal from the air. The review comprehensively outlines the morphological, chemical, and mineralogical attributes utilized for PM source identification and their subsequent remediation by plants. Finally, the benefits of SEM-image-based techniques for PM analysis are elucidated. This review offers a holistic understanding of the SEM-EDS and plant-based approach, presenting a promising avenue for addressing PM pollution and enhancing environmental quality.

KEYWORDS

particulate matter (PM), SEM-EDS, source apportionment, elemental composition, image-J

1 Introduction

Ambient air pollution is projected to cause approximately four million premature deaths each year, with levels of air pollution continuing to rise in most locations (Neira and Prüss-Ustün, 2016; Shaddick et al., 2020). The correlation between human disease and poor air quality has been recognized since antiquity. The major health effect of air pollution entered the world in the 20th century (Speak et al., 2012). Among air pollutants, airborne particulate matter (PM) is made up of extremely small particles and liquid droplets containing soil and dust particles and many harmful substances like organic chemicals, acids, and metals United States Environmental Protection Agency. In PM, dust, dirt, and

soot are quite visible with the naked eye; however, some are very small and can only be seen with the help of microscopes. The presence of PM in ambient air, which comes in a variety of sizes and chemical and biological components, is a major scientific and policy issue (Ali et al., 2019). The composition of PM encompasses both inorganic and organic components. Inorganic PM may include minerals, metals, and salts, while organic PM consists of carbon containing compounds originating from different types of sources like biomass burning or industrial emissions (Jimenez et al., 2009). According to the World Health Organization (WHO), in 2021, approximately 58% of outdoor air pollution-related deaths were due to ischemic heart disease and stroke. Moreover, about 18% of deaths occurred due to cardiopulmonary and acute lower respiratory infection, and 6% of lung cancer deaths are attributed to PM globally (WHO, 2021).

Atmospheric PM play major contribution in shaping the impacts of air quality. These particles, suspended in air, exhibit diverse chemical compositions, originating from both natural and anthropogenic sources. PM can be broadly classified based on their origin, such as natural geogenic and anthropogenic. The PM derived from natural processes such as mineral dust, sea salt, aerosols, biogenic emissions, forest fires, dust storms, volcanoes, and aerosolized sea salt. In contrast anthropogenic sources contribute PM through industrial activities, combustion processes, tobacco smoke and vehicular emissions (Speak et al., 2012; Seinfeld and Pandis, 2016). PM in the atmosphere is generated through primary and secondary processes. Primary PMs are directly emitted in air, such as soot from combustion or soil erosion particles. Secondary PM are formed in the environment through complex chemical reaction between primary PM involving precursors gases leading to the formation of fine PM (Kanakidou et al., 2005).

In most developing countries, the PM level is higher than the permissible limit due to continuous increase in industrialization and the number of vehicles (Speak et al., 2012). PM is classified according to its aerodynamic diameter: (a) Total suspended particulate matter (TSPM), (b) coarse PM ($\leq 10 \mu\text{m}$), (c) fine PM ($\leq 2.5 \mu\text{m}$), and (d) ultra-fine PM ($\leq 1 \mu\text{m}$) (Muhammad et al., 2020; Xie et al., 2020). PM with a diameter larger than $10 \mu\text{m}$ has a somewhat small suspension half-life and is predominantly filtered by our nose and upper airways. In environmental air samples, the total number and total surface area of these PM increase exponentially as the diameter of the PM reduces. However, PM mass will also reduce with the diameter. According to size ranges, $<11 \mu\text{m}$ PM can enter the nasal passages of the human respiratory system, $7\text{--}4.7 \mu\text{m}$ can enter the pharynx, $4.7\text{--}3.3 \mu\text{m}$ can enter the trachea. PM with an aerodynamic diameter of $3.3\text{--}2.1 \mu\text{m}$ can enter the primary bronchi, $2.1\text{--}1.1 \mu\text{m}$ can enter the bronchi branches, $1.1\text{--}0.65 \mu\text{m}$ can enter the bronchiole, and $<0.65 \mu\text{m}$ particles can enter the alveoli regions (Londahl et al., 2006). Studies also showed an increase in illness and mortality related to PM exposure (Anderson et al., 2012; Chen et al., 2022; Liang et al., 2022; Liu et al., 2022). PM causes many health complications such as cardiovascular diseases, cancers, neurodegenerative diseases, respiratory diseases (asthma), and viral infections (Thangavel et al., 2022). Much evidence is recorded in China, which reveals PM can increase the mortality rate due to asthma (Liu et al., 2022). Epidemiological studies related to human mortality provide proof of the relationship between short-term ambient PM_{10} and $\text{PM}_{2.5}$

exposure with cardiac ischemic heart disease mortality. PM_{10} and $\text{PM}_{2.5}$ show a linear relationship with cardiac ischemic heart disease mortality (Liang et al., 2022). Various instruments are installed at various locations, especially in urban and industrial locations around the globe, for the continuous measurement of PM in different size fractions such as TSPM and respirable suspended PM (RSPM) and also in lower fractions. To control pollution, we need to have proper measurement of PM with respect to its concentration levels in various size ranges.

In recent decades, numerous research studies on PM and its associations have been published. To facilitate authentic data collection associated with PM, the Web of Science stands out as the best-built tool. A network visualization was conducted using the Web of Science's built-in tool and VOSviewer software. This analysis provides a clearer understanding of PM studies and global trends. Figure 1 illustrates the network visualization of terms associated with PM in the last 10 years (2012–2022) with at least 25 occurrences of associated keywords. Approximately 433 keywords meet the threshold of having a minimum of 25 occurrences. It depicts current trends in research and development regarding PM studies in the Web of Science. The word frequency in articles and its associations with other keywords are displayed on the cloud map. Each term in the network is represented by a circle, the size of which corresponds to the number of publications in which the term appears. The thickness of the lines indicates the strength of connections between topic areas or keywords, and the length of the curved lines indicates the approximate connection frequency. Each colour represents a group of terms grouped into clusters, showing connections between various topics.

The review work by Galvão et al. (2018) summarizes trends in analytical methods for the broad chemical characterization of PM from 1997 to 2018, and single-PM particle analysis has previously been addressed by Bzdek et al. (2012). Tiwari A. et al. (2022) discussed all possible methods for the study of PM based on the literature. It appears that the study of PM has primarily focused on its composition, particle size, and source apportionment. The morphology of PM unveils its origin, as each specific activity or source emitting PM exhibits distinct microscopic characteristics. (Liati et al., 2019; Zhang et al., 2022). SEM is an effective and easy tool for identifying the microscopic character of PM. The morphology of PM indicates the appearance, texture, and source of the particles. Consequently, SEM-based PM studies are crucial for accurately identifying particle characterization and composition (association with EDS). In context of SEM-EDS analysis, emphasis on mineral composition becomes paramount. Unlike other analytical technique like inductive coupled plasma (ICP), SEM-EDS provide insight into the morphological, elemental and mineralogical aspect of particles. This technique provides clear view on the source of PM. Mineral composition serves as a key indicator of their origin and generation processes (Fialová et al., 2017). However, a small number of studies have attempted to use SEM imaging to identify the PM shape. This review mainly focuses on the emerging approach of SEM-EDS based PM investigation and emphasizes how imaging of PM by SEM and chemical and mineralogical information by EDS is crucial for its source apportionment. SEM imaging methods for the study of PM have been shown as a schematic summary in Figure 2.

2 Plant leaves as a bio-sampler of PMs

Studies have shown that plants are key points for filtering the air by absorbing PM on their leaves and remediating PM pollution (Escobedo et al., 2008; Cavanagh et al., 2009; Maher et al., 2022; Kim et al., 2023). PM is deposited on plant leaf surfaces by sedimentation under gravity and diffusion (i.e., Brownian motion) (Freer-Smith et al., 2005; Maher et al., 2022; Kim et al., 2023). Plants have the competence to retain PM on their surface through their micromorphological characters of leaves, macro structure of vegetation, and environmental variables like wind and temperature (Freer-Smith et al., 2005; Mo et al., 2015; Chen et al., 2017; Tiwari et al., 2022). The arrangement of leaves also plays a major role in enhancing PM deposition (Chaturvedi et al., 2013; Mo et al., 2015; Tiwari et al., 2022; Tiwari et al., 2023). Leaves are the best bio-samplers for PM sampling because many factors are responsible. The leaf morphology, leaf surface, leaf shape, leaf size, and foliage pattern of the leaf are responsible for the capturing of PM (Terzaghi et al., 2013; Wang et al., 2015; Corada et al., 2021).

In leaf morphology, trichomes were the most frequently cited leaf surface trait for capturing PM. Trichomes or leaf hairs have different variations in size and morphology and on leaf distribution pattern. Mostly they are present on the abaxial side of the leaves. Plants with a dense number of trichomes or leaf hairs on their adaxial surface are more efficient in capturing PM compared to those plants whose leaves are either glabrous (hairless) or have trichomes present on the abaxial surface and low-density trichomes (Saebo et al., 2012; Chen et al., 2017; Zhang et al., 2017; Li et al., 2019). Leaf roughness also influences PM capture. Leaves with rough surfaces are more efficient, and furrowed, ridged, and wrinkled surfaces can retain coarse PM (PM_{10}) better than smooth leaves (Chaudhary and Rathore, 2018). Waxy coating also plays a major role in PM accumulation. Ultrafine particles become more enduringly absorbed in the leaf surface waxy coating (Hofman et al., 2014; Song, 2015). The shape and size of leaves also impact PM capture. Leaves with larger surface areas can intercept a greater volume of air, increasing the chances of PM deposition. Moreover, specific leaf shapes, such as compound leaves with multiple leaflets, can create additional surfaces for particle interception (Katoch, and Kulshrestha, 2022). Leaves have a special structure like stomata. Stomatal size, density, and guard cell area are special traits for PM capture. PM can be trapped in stomatal cavities and grooves, so large and dense stomata are more efficient at accumulating fine and ultra-fine PM (Chen et al., 2017; Zhang et al., 2017; Li et al., 2019). The entire leaf design of plants, including the arrangement and density of leaves, influences their ability to trap PM. Dense canopies with overlapping leaves can form a physical barrier that prevents airborne particles from passing directly through. Furthermore, differences in foliage density within a plant community might influence PM deposition patterns on leaves (Vardoulakis et al., 2003). Microscopic observation is commonly used to determine the amount of PM on leaves (Song et al., 2015; Sgrigna et al., 2016). It can quantify particles with original diameters of particles and by the shape and may indicate the source of particles (Stoffyn-Egli et al., 1997).

3 Scanning electron microscopy (SEM) based PM analysis

Microscopy can be used for measuring particle size distribution and the number of particles (Weber et al., 2014). SEM is commonly employed to visually quantify the PM captured by leaves and to identify the number, types, and sources of the PM (Deljanin et al., 2014; Manisha et al., 2016). The SEM images reveal the size, shape distribution, morphology, and chemistry of particles as small as a few nanometers. Consequently, it can provide information about a probable source that is not available through basic bulk chemical analysis (Tasić et al., 2006). Its capacity for greater magnification with appropriate resolution and a large depth of field makes it highly useful. SEM also has the ability to analyze the elemental composition of PM when combined with Energy Dispersive Spectroscopy (EDS) attached to the microscope (Kocic et al., 2014). However, the most commonly used method for SEM micrographs is to identify the size of PM and count PM using software-based analysis. Some studies have also utilized SEM micrographs in software for size distribution and quantification of particles. ImageJ software (Ottel   et al., 2010) was previously employed for quantifying PM in all size ranges (Gajbhiye et al., 2019; Tiwari et al., 2023).

3.1 Image based counting of PM by software

SEM images provide significant insights into the aggregation pattern and shape of PM. With the assistance of SEM images, the quantification of particles becomes possible. SEM micrographs are utilized for counting particles in different size fractions. Image-based software is employed for accurate measurement and counting of particles in various size fractions. The counting of PM_{10} , $PM_{2.5}$, and $PM_{0.2}$ is achievable through SEM micrographs with the aid of ImageJ.

ImageJ is an image-based technique. To use this software, the image should be in binary (black and white) form. An electron microscope is used to take images of the PM sampling surface, such as plant leaves, filter papers, or another exposed surface of PM. The SEM images were captured at various magnifications (e.g., 100 \times , 250 \times , and 500 \times). After collecting the images, the particles are automatically quantified by this software package (Ottel   et al., 2010). With the help of SEM micrographs of sampled leaves, PM density is measured by this software package. Particle counting in all size ranges like PM_{10} , $PM_{2.5}$, PM_1 , and $PM_{0.2}$ can be easily done (Song et al., 2015; Weerakkody et al., 2018b), and the average size of PM is also identified by this software (Zapata-Hernandez et al., 2020). For errorless data, an auto-threshold mechanism is applied to all the calculations (Ottel   et al., 2010). To explain any changes in PM accumulation due to variable edge effects, the leaf perimeter/leaf surface area ratio in each category was determined by this software. With the help of ImageJ software, we can identify the actual number of PMs in all size ranges (0.1–100 μ m) and how much space they occupy on the leaf surface (Song et al., 2015; Zhang et al., 2022).

ImageJ software has two types of thresholding mechanisms (manual and automatic) built-in. Automatic thresholding is used for PM counting because it is not influenced by the user, but in manual thresholding, the user arranges the pixels by adjusting itself, creating biasness in counting PM. The watershed function in ImageJ

was used to separate particles that were somewhat overlapping in a threshold image. Particles in this study, which are not circular, can still be counted (Ottel  et al., 2010). For better results, $\times 100$ magnification images are used for $>10 \mu\text{m}$ particles, $250\times$ is used for PM_{10} , and $500\times$ magnification image is used for $\text{PM}_{2.5}$. The magnifications of $250\times$ and $500\times$ were employed, respectively, 6.25 and 25 times, to make up for the loss in counting area caused by the zooming effect (Ottel  et al., 2010). The category of the particle (coarse, fine, or ultrafine) that is more prevalent on the surface of the leaves can also be determined by counting particles in various size ranges by ImageJ. After that, these results help in selecting those plants which are efficient in capturing fine and ultrafine particles (Shi et al., 2017). This software assists in recognizing the particle concentration on plant leaves, making it simple to correlate it with the atmospheric PM concentration of a specific area (Sternberg et al., 2010). Particle counting can also help identify the source of PM because smaller, and more numerous particles typically arise from industrial sources. If there are fewer, bigger particles present, they are likely to come from soil or other natural sources. Palma et al. (2017) studied the rural sites always had the least particles, whereas urban and industrial sites had the most. Another study which was based on SEM-image was able to reveal that PM concentration in the air is distinguishable between summer and winter seasons. As the summer season has more PM concentration in the air, it was evident from more accumulation on plant leaf surfaces as compared to the winter season when studied with SEM-ImageJ method (Kumar and Elumalai, 2018). So, it is sensitive enough to distinguish the seasonal variation.

The SEM-based imaging facility of PM on the plant leaf surface provides PM appearance with morphology and the pattern of accumulation on the leaf surface. ImageJ software analysis of SEM micrographs provided the distinct deposition pattern of PM on different leaf surfaces (adaxial Vs. abaxial) (Gajbhiye et al., 2019). Plant leaves have adaxial and abaxial surfaces, and both surfaces have different morphology with variable capture efficiency. For instance, in *Calotropis procera*, fine PM accumulation was higher at the abaxial surface, and larger PM accumulation was found at the adaxial surface (Gajbhiye et al., 2019). This pattern was observed by the SEM image of the sampled leaf of *C. Procera*, and counting was done by ImageJ software. This type of innovative method uses object-based image processing to automatically estimate PM on tree leaves from SEM micrographs. This method is very useful for the selection of plants for removing PM from the air (Li et al., 2021; Tiwari et al., 2023). Plant leaves can accumulate PM, and the SEM image of the plant leaves gives information about the accumulation pattern of PM in different size ranges. PM size, shape, and number were also identified if the SEM images are analyzed by ImageJ software. After the analysis (PM counting in different size ranges), the selection of better plants for removing PM in the respirable range from the air is possible. For instance, Tiwari et al. (2023) studied six plants for the selection of suitable plants for removing PM from the air. In this research, SEM images-based method was used incorporated with ImageJ and found that *Dalbergia sissoo* is the best-performing plant for removing air in the respirable range (Tiwari et al., 2023). As compared to leaves with rough surfaces and bigger trichomes, smooth leaves with smaller trichomes retain more fine and ultrafine particles (Tiwari et al., 2023). Smaller leaves, lobed shapes, hairy and rough leaf surface are more effective in

retaining the PM on its surface (Weerakkody et al., 2017; 2018b). Another study was done by Weerakkody et al. (2019) in which they used SEM-ImageJ method and demonstrated that the arrangement of plants may also affect the PM retention capability. Findings of this study showed that planting with heterogeneous topography should enhance the PM retention ability compared to homogenous topography planting design (Weerakkody et al., 2019). SEM-ImageJ based study also used for the characterization of PM which was emitted from pyrolysis (burning in the absence of O_2). For instance, pyrolysis of cashew nuts at 700°C emits smaller particles as compared to pyrolysis at 500°C (Kibet et al., 2017). Based on these findings and literature reviews, high-temperature cooking may potentially be a source of dangerous PM, molecular toxins, intermediates, and free radicals, which may be precursors to disease (Kibet et al., 2017; Chen et al., 2022; Liang et al., 2022).

4 Morphological analysis of PM by SEM images

SEM Microscopy can be used to observe the particle deposition pattern on plant leaves, and the capture of PM from the environment by plants is considered an important ecosystem service. The deposition of PM on plant leaves can occur in two ways. Firstly, plants have the ability to retain particles on the leaf surface through their micromorphological characteristics, such as stomata, trichomes, and epicuticular wax (Terzaghi et al., 2013). Secondly, particles deposited on the leaf surface may be temporary and can resuspend in the air when there is a change in air flow or an increase in wind speed. Thus, leaves possess special micromorphological characteristics to adhere particles to their surface (Nowak et al., 2014). Microscopy can be employed to measure the size and shape of PM. SEM is commonly used to visualize particles and identify types of PM. With this approach, significant information about the size and shape characteristics of the particles captured on leaves can be collected. The characteristics of particles, such as roundness, compactness, and shape index, provide insights into the source of the particles (Slezakova et al., 2008; Makkonen et al., 2010; McDonald and Biswas, 2012).

Morphological studies of PM facilitate comparative research on the PM capturing efficiency of different plant species and source apportionment of PM based on their morphological characteristics. This method can also provide insights for species selection for specific pollutant reduction (Yan et al., 2016). Particles can be divided into fifteen subcategories based on their shape, size, and general appearance (Freer-Smith et al., 1997). Smooth sphere, agglomerate round and ovoid, agglomerate flat, agglomerate free shape, nodules like, hollow sphere, and hollow irregular-shaped are considered inorganic particles. Cylindrical dehydrated spores, symmetrical spores, rough spherical, and sheet material often curled are considered organic particles (Freer-Smith et al., 1997). Hollow sphere particles are assumed to be incomplete combustion products. Probably inorganic materials are amorphous and free-shaped. Some particles are not distinguishable because they do not show similarity to the classified categories, so they are not assigned to any group. Most of the amorphous inorganic particles probably come from the soil, mainly containing silicon, iron, and aluminum

TABLE 1 Source identification of PM by their morphological character.

S No.	Morphology	Source	Origin	References
1	Rounded smooth surface	Thermal powerplant, fly ash, fossil fuel burning	Natural and anthropogenic	Zeb et al. (2018); Goel et al., 2020
2	Rough spherical	Pollen grains	Biogenic origin	Teloli et al. (2016)
3	Large irregular	Soil, local crustal, from combustion process	Natural	Goel et al. (2020)
4	Large crystal shape with sharp end	Vehicular emissions, road maintenance	Natural and anthropogenic	Bora et al. (2021)
5	Non spherical	Crustal, soil particles, silica powder, Ca and Fe rich	Natural and anthropogenic	Satsangi and Yadav. (2014)
6	Minute irregular shape	Soil erosion of rocks	Natural	Satsangi and Yadav. (2014)
7	Crystal shape	Rock particles, soil dust	Natural	Bora et al. (2021)
8	Rounded	Combustion process, industrial particle	Anthropogenic	Yin et al. (2013); Goel et al. (2020)
9	Spherical	Coal, power plants, soil resuspensions	Anthropogenic	Goel et al. (2020)
10	No distinct morphology	Soil, automobile emission	Natural and anthropogenic	Satsangi and Yadav. (2014)
11	Irregular	Soil, local crustal, from combustion process	Natural and anthropogenic	Satsangi and Yadav. (2014)
12	Small spherical	Vehicular emission, fuel type	Anthropogenic	Yin et al. (2013); Zeb et al. (2018); Goel et al. (2020)
23	Agglomerate free shape	Fuel, biomass burning, incomplete fossil fuel	Natural and anthropogenic	Bora et al. (2021); Rana et al. (2022)
14	Crystal shape	Rock particles, soil dust	Natural	Bora et al. (2021)

(Breed et al., 2002; Almeida et al., 2006; Castanheiro et al., 2020). Beckett et al. (2000) also categorized particles similarly to Freer-Smith et al. (1997). However, they divided inorganic amorphous particles based on their size (<2.5 μm and >2.5 μm), with inorganic and amorphous particles smaller than 2.5 μm assumed to be of anthropogenic origin (Guevara, 2016), as the majority of particles in this size range are present in the air.

Morphological analysis is always appreciated for the ability to link particles back to their origin. For instance, particles of hollow carbonaceous sphere shape are emitted from the burning of fossil fuels. It is difficult to assign the source of agglomerated particles and mass of mixed materials. They mostly come from anthropogenic sources. The average size of anthropogenic PM is less than 10 μm . It can also be observed that particles emitted by anthropogenic sources are generally smaller than those produced from natural sources (Beckett et al., 2000). According to Sagnotti et al. (2009), particles rich in iron, with a size range of 0.1–5 μm , of which 1–2 μm is more common. The iron-rich particles have variable shapes, ranging from rounded to irregular, and were found in high traffic squares in Rome metropolitan areas. These particles showed a distinctive rough, moss-like surface composed of adjoined or aggregated, sub-round particles typically about 50–60 nm in size (Sagnotti et al., 2009). Tasić et al. (2006) classified particles into two categories using SEM-based PM morphology in the urban area of Belgrade. First, materials of organic origin were among the particles of natural origin (pollens, bacteria, fungal spores). Suspended soil dust (mostly minerals) and angular-shaped objects were also included in this group. Second, particles from anthropogenic sources were defined by their special shape and smooth surface, which were usually emitted by high-

temperature combustion processes (Tasić et al., 2006). A large amount of PM is emitted from iron and steel-based industries. These types of industries emit two types of PM: stack PM and fugitive PM (Zhang et al., 2022). In recent research, it was found that fugitive PM emitted by the steel industry has five forms based on their microscopic morphology: spherical, irregular blocky, chain, lamellar, and flocculent particles (Zhang et al., 2022). Spherical particles are Si, Al, and Fe-rich and come from high-temperature combustion. Irregular blocky particles are Fe, Si, Ca, and Mg-rich and mostly produced by mechanical processes (crushing, belt conveying, unloading, and dropping) in iron industries. Chain and flocculent particles are composed of volatile minerals and lamellar particles are carbon-rich (Zhang et al., 2022). Airborne fiber PM has its special morphological characters and appearance, providing insights into the source of PM (Li et al., 2020). Microplastic fiber particles show spiral forms with a smooth surface, natural organic fibers show regular fibrous and elongated shapes. Man-made mineral fibers are regular fibers with a bar-shape, asbestos shows needle-like morphology, calcium sulfate fibers appear with a smooth strip-like surface, and metal fibers are regular to irregular fibrous shapes observed in SEM images (Li et al., 2020). Some possible PM morphology with sources is shown in Table 1.

5 Particle chemical characterization by SEM-EDS

For source identification, the characterization of PM and the identification of its chemical composition are required. The

elemental composition of airborne PM deposited on plant leaf surfaces has been shown to be strongly linked to pollution sources (Umbria et al., 2004; Canepari et al., 2008; Thorpe and Harrison, 2008; Sgrigna et al., 2016; Baldacchini et al., 2017; Baldacchini et al., 2019). SEM analysis combined with EDS can now offer a full characterization of PM deposited on plant leaves (Castanheiro et al., 2016; Weerakkody et al., 2018a; Castanheiro et al., 2020). These methods primarily provide information on the quantity, morphology, and elemental content of PM. SEM-EDS was used to investigate PM samples to determine their composition, making an effort to differentiate whether the PM came from anthropogenic or natural sources.

Based on SEM-EDS observations, particles can also be classified into anthropogenic, geogenic, and biogenic particles (Usman et al., 2022). Anthropogenic particles are mostly produced by industrial, vehicular, and fossil fuel combustion, among others. Carbonaceous particles are those with a content of C and O greater than 92% (Cong et al., 2009; Tumolva et al., 2010; Deka and Hoque, 2014; Bhuyan et al., 2018). The concentration of carbon in the air increases due to incomplete burning of biomass and fuels (Sahu et al., 2012). Sulfur in the PM indicates its origin from sulfur included in the fuel during the combustion process and is most often associated with secondary formation. (Pósfai et al., 2003; Jimenez et al., 2009; Agarwal et al., 2011; Seinfeld and Pandis, 2016). Biogenic particles were quantified using the technique used by Matthias-Maser and Jaenicke (Matthias-Maser and Jaenicke, 1994). Many scientists have discovered that particles of biological origin (living or dead) include tiny amounts of Na, Mg, K, P, Si, Fe, Cl, Al, and Ca in various sizes and shapes (Artaxo and Hansson, 1995; Matthias-Maser et al., 2000a; 2000b; Pósfai and Buseck, 2010; Kaur et al., 2022). The composition of biogenic PM is relatively different from others, showing O and C concentrations less than 75%, K more than 1%, and P and Cl less than 10%. These particles include plant debris, animal matter, bacteria, viruses, pollens, and spores (Iordanidis et al., 2008; Coz et al., 2010; Malli Mohan et al., 2019; Tiwari et al., 2022). Particles generated by natural crust are termed as geogenic particles. This particle mainly includes aluminosilicate, calcium-rich, and quartz (Al, Ca, C, Fe, Mg, O, K, Si, and Na). Aluminosilicate makes up to 72% of all chemical compounds found in the Earth's crust (Davidovits, 1994; Cong et al., 2009). Bioaerosol also identified by their chemical compositions. Bioaerosols mainly includes virus, bacteria, pollen grains, animal and plant debris, and spores. The concentration of O and C is more than 75% and Cl, Ca, S, P were present in minimum amount (e.g., Chen et al., 2012; Usman et al., 2022).

The composition of PM provides more information regarding pollution sources. Some particles have specific characteristics and compositions. Thus, using SEM-EDS, the elemental composition of particles was determined, and the origin of the particles can be better understood. It can be seen that the most abundant element found in the PM was Si, which has been found to be a marker of soil particle origin (Chow et al., 1996). Similarly, a high amount of S and P marks the presence of particles from anthropogenic combustion sources (Fung and Wong, 1995). Higher calcium concentration in PM is due to the presence of chalky soils (Beckett et al., 2000). Agglomerated particles with elements attributed to diesel (C, Na, Mg, K, Al, Si, P, and Cl), coal (Al, Si, Fe, Ca, S, Na) and coal ash (C, Al, Si, K, Ca) are generated by high traffic at roadside plants of industrial area (Searle,

2001; Tomasevic and Anicic, 2010). The C, O, Si, Ca, Na, and Mg in the PM could have derived from natural sources. Particles containing Pb, Br, Fe, Cd, Ni, Zn and Cl, on the other hand, could be the product of anthropogenic activity (Ottel  et al., 2010; Gonz lez et al., 2018). According to Tomasevic and Anicic, (2010), such PM with the composition Si, Al, Fe, Mg, N, S, Ca, K, and Cl are the soil dust. On the other hand, PM emitted from fuel burning were rich in Al, Si, Ca, Ni, Fe, V, and Pb. PM with an irregular shape and a high concentration of Fe, O, Si, Mg, and Al could have derived from soil dust resuspension, with a diameter of less than 3 μm (Song et al., 2015; Engelbrecht et al., 2016). PM with a diameter of around 10 μm and a spherical shape, as well as a significant concentration of Pb and Br, and presence of S, Ca, Na, Mg, and Cl could have come from vehicle exhaust (e.g., Nor et al., 2022). Fe particles with 86% of Fe weight originated from iron industries. PM including C, O, Si, Fe, and Ca came mostly from natural sources and to a lesser extent from human activities (Goudie, 2009; Wang et al., 2012). $\text{PM}_{2.5}$ (fine particles) were almost emitted from anthropogenic sources with a total content of C 50.18% and O 31.74% (accounting for 81.92%). The PM with Pb rich and composition with Fe, Zn, Ni, and Cu are the characteristics of PM emitted from local industrial processes (Tomasevic and Anicic, 2010; Daellenbach et al., 2020) and Fe rich with composition of Cu, Zn, Pb, Ni, and Cr are emitted from traffic sources (Moreno et al., 2003; Slezakova et al., 2008). The content of Mg, Al, Si, Cl, Ca, Fe, and Pb were higher, and these components were related to diesel fumes and coal dust (Ottel  et al., 2010). Some possible sources of elements are shown in Table 2. Recent studies related to PM source identification by its composition show different observations. For instance, condensable PM is a mixture of several gases (from coal industries), and its character shows both morphological and elemental composition evidence (Oroumijeh et al., 2022). Condensable PM exhibits a spherical morphology in SEM and is characterized by the presence of elements such as Hg, As, Se, and Sb in its composition. PM emitted by heavy traffic has some special characteristics, such as containing Ba, Cr, Cu, Mo, Pd, Zn, and Zr in both PM_{10} and $\text{PM}_{2.5}$; however, Fe, Li, Mn, Bi, Mo and Ti were mainly associated with $\text{PM}_{2.5}$ in traffic emissions (Oroumijeh et al., 2022; Alves et al., 2023). Some PM emitted from quarries, burning fossil fuels have a mixture of Ca, Si, Pb, Ca, Fe, Ti, and Al possible metals (Zapata-Hernandez et al., 2020).

6 Mineralogical composition of PM based on SEM-EDS

Atmospheric PM is composed of various solid and liquid substances. Classification of atmospheric PM according to their size, with respect to their mineralogical character, indicates their potential to affect human health and source identification (Pope and Dockery, 2006). Mineral dust PM plays a major part in atmospheric aerosols, contributing 35%–40% of global aerosols from different natural sources (Ramanathan et al., 2001). Additionally, mineral dust particles aid in reducing ambient ozone levels by approximately 5% (Soler et al., 2016). These particles, when carried by dust storms, can be transported over long distances, leading to impacts at regional and even global scales (Weinzierl et al., 2017). Therefore, understanding the dynamics of mineral dust aerosols

TABLE 2 Source identification of PM by their elemental composition.

S. No.	Sources	Elemental composition	References
1	Soil particle	Si, C, O, Si, Al	Chow et al. (1996); Song et al. (2015)
2	Anthropogenic combustion sources	P, S, As	Fung and Wong (1995)
3	Chalky soils	Ca	Beckett et al. (2000)
4	Road traffic attributable to diesel	Pb, C, Na, Mg, K, Al, Si, P, Cl	Searle (2001)
5	Coal	Al, Si, Fe, Ca, S, Na, Zn	Searle (2001)
6	Building construction dust	C, O, Ca	Song et al. (2015)
7	Vehicular exhaust	Pb, Br, S, Mn	Song et al. (2015); Galvez et al. (2022)
8	Iron steel manufacturing industries	86% Fe weight, Br, Cl	Song et al. (2015); Galvez et al. (2022)
9	Natural sources	C, O, Si, Ca, Na, Mg	Ottel� et al. (2010); Song et al. (2015)
10	Carbonaceous aerosol	O, N, C, S	Galvez et al. (2022)
11	Crustal and traffic emission	Si, Zn, Cu, Al, Na, Ba, K, Pb, Mn, O	Galvez et al. (2022)
12	Gold product and jewellery enterprises	Hg, Pb	Galvez et al. (2022)
13	Fly ash (Aluminosilicate)	Al, Si, O	Searle (2001)
14	Pharmaceutical waste and cosmetics	Sb, Fe	Bocca et al. (2014); Muritala and Adewole (2019)
15	Mining process	Tb, Zn, In	Genga et al. (2012)

is essential for comprehending their role in atmospheric processes and their potential environmental implications. Most of the mineral dust PM are found with irregular shapes or amorphous (Bora et al., 2021).

A recent work by G rka et al. (2020) focused on atmospheric PM characterization. They divided the PM according to their mineralogical composition into three groups. Group 1 primarily reflects industrial activities, showcasing a prevalence of Si/Al particles (58.2%), often containing toxic metals (34%). These particles are found across all size fractions (PM₁₀, PM_{2.5}, and PM_{0.2}), indicating widespread industrial emissions. Group 2, on the other hand, highlights urban influences, with terrigenous particles dominating the composition, along with sulfides and Ca/P/K spherules, originating from various urban sources like chemical plants and agricultural practices. Lastly, Group 3 predominantly consists of particles derived from natural geological processes, particularly from the local areas, featuring terrigenous phases like quartz (42.9%) alongside other mineral components (G rka et al., 2020). Particles with the composition of some elements (Al, Ca, Fe, Mg, Mn, Ni, and Si) showed the presence of alumina silicate, mica, quartz-like materials. This mineralogical composition denotes that PM generated from resuspension of dust from soil and other anthropogenic particles emitted from the burning of fossil fuels and biomass (Pipal et al., 2014; Bhardwaj et al., 2017; Sonwani and Kulshrestha, 2018). Teper (2009) also classified particles based on their origin of mineral components, which include tailing pond components (Pb, Zn, Fe sulfides; Pb, Zn carbonates; Fe sulfates), natural components from rock and soil erosion (aluminosilicates, quartz, Ca-Mg, Ca carbonates), and other pollutants from various airborne sources (aluminosilicate glass, Fe-Zn oxides, Fe oxides). However, overlap of some

components between groups occurs due to shared components like carbonates and aluminosilicates, and some pollutants can also originate from natural sources (Teper, 2009).

Mineral-rich atmospheric PM inhaled by humans is very toxic, causing silicosis, tuberculosis, lung cancer, and chronic bronchitis (Merget et al., 2002). Clay mineral-rich PM has been reported to have minimal or no toxic effect (Carretero et al., 2006). SEM-EDS results showed the mineralogical information of PM. The detection of elements on EDS like Al, Si, O, Mg, C, Ca, Na, and Fe has clearly confirmed the presence of quartz and aluminosilicate. The presence of high silicon and oxygen (Si + O = >50%) content in PM denotes the presence of quartz (SiO₂). These particles come into the environment by both natural and anthropogenic processes (Pachauri et al., 2013; Satsangi and Yadav, 2014). Silica is widely present in the earth's crust and a major component of granite and sandstone. Furthermore, silica is widely used in construction processes such as ceramics, cements, bricks, clays, and glass. So, this type of PM is emitted from the construction and renovation of buildings (Pachauri et al., 2013). The size range of silica particles is near about 0.2–0.5 µm (Usman et al., 2022). If in PM EDS analysis Al, Si, Ca, O present in higher percentage, it confirms the presence of biotite, almandine, and grossular minerals (Pachauri et al., 2013; Usman et al., 2022). Aluminosilicate is also widely present in the earth's crust (70%) (Cong et al., 2009). These particles are made up of oxides of Al and Si, with varying quantities of Fe, Mg, Na, Ca, and K. The average size of these particles ranges from 2.5 to 30 µm (Usman et al., 2022). These types of PM show sharp angular structures and have been identified as Na-feldspar (albite), K-feldspar (K aluminosilicate), Mg-Fe aluminosilicate, and Ca-Mg aluminosilicate (Satsangi and Yadav, 2014; Anake et al., 2016; Usman et al., 2022). By the help of elemental composition of

TABLE 3 Source identification of PM by their mineralogical composition.

S.N.	Mineral	Composition of minerals in EDS	Source	Reference
1	Aluminosilicate	Al, Si, and O	Natural and Combustion process	Searle (2001)
2	Sodium feldspar	Na, Si, Al, and O	Anthropogenic sources	Galvez et al. (2022)
3	Potassium feldspar	K, Al, Si, and O	Anthropogenic sources	Galvez et al. (2022)
4	Kaolinite	Al, Si, and O	Coal industries	Song et al. (2014)
5	Koktaite	N, Ca, S, and O	Crustal origin and waste dump	Song et al. (2014)
6	Mascagnite	S, O, and N	Coal Combustion	Song et al. (2014)
7	Gypsum	Ca, S, and O (>40%)	Coal ash, waste rock	Zheng et al. (2009); Song et al. (2014)
8	Boussingaultite	Mg, N, S, O	Crustal origin and waste dump	Song et al. (2014)
9	Quartz	Al, Ca, Fe, Mg, Mn, Ni, and Si (>50%)	Fossil fuel burning, Soil dust	Sonwani and Kulshrestha, (2018); Bhardwaj et al. (2017)
10	Mica	Al, Ca, Mn, Ni, and Si	Natural source, Soil	Sonwani and Kulshrestha, (2018); Bhardwaj et al. (2017)
11	biotite	Al, Si, Ca, O, Mg, and Fe	Crustal origin	Pachauri et al. (2013); Usman et al. (2022)
12	almandine	Al, Si, Fe, and O	Earth Crust	Pachauri et al. (2013); Usman et al. (2022)
13	grossular	Al, Si, Ca, and O	Crustal origin	Usman et al. (2022)
14	Halite, rutile	Na, Cl, Ti, O, and Si	Rare earth Crust	Bartz et al. (2021); Song et al. (2014)
15	Phosphate mineral (Apatite, hydroxyapatite, fluorapatite and chlorapatite)	Cl, F, O present in high amount. Ca, Si, and Al	Phosphate fertilizers, Mining	Macadam et al. (2003); Bartz et al. (2021)
16	Dolomite	Rich in Mg, Ca, O and C	Crustal origin, Mining	Bartz et al. (2021); Song et al. (2014); Macadam et al. (2003)
17	Carbonates (Calcite, Siderite)	Fe, C, Ca, and O (Fe and C rich)	Mining, Fossil fuel burning	Liu et al. (2023)

SEM-EDS analysis, some particles are nitrate-rich with irregular shapes (Matsuki et al., 2005). Nitrate particles are present in the form of NaNO_3 , HNO_3 , and other nitrogenous compounds (Teinilä et al., 2000). With the help of SEM-EDS, Lu et al. (2007) identified the mineral composition of PM present in the air in Beijing, China. They found different minerals such as illite, smectite, chlorite, quartz, feldspar, calcite, gypsum, kaolinite, present in the form of respirable PM (Lu et al., 2007). In a recent study by Bartz et al. (2021), PM was categorized by their mineral compositions into 11 different groups. Three groups were associated with natural sources, while others were emitted from anthropogenic sources like smelter industries. The presence of Si and Al elements in respirable PM indicates the involvement of anthropogenic activity (Bartz et al., 2021). Terrigenous minerals (quartz, feldspar, mica) and clay minerals are emitted from natural sources or the earth's crust (Bartz et al., 2021). Other minerals such as halite, rutile, amphibole, apatite, sulfides (Zn, Cu, Pb, Fe), carbonates (dolomite, calcite, siderite), and gypsum are emitted from most anthropogenic sources (Table 3) (Song et al., 2014; Bartz et al., 2021). Geogenic particles identified by their mineralogical compositions include aluminosilicates, quartz, fly ash, chloride particles, iron-silicon alloys, and calcium

carbonate particles. These particles originate from sources such as soil sediment, biomass burning, construction activities, and windblown dust, contributing to the composition of PM in the atmosphere (Ahmad et al., 2023).

The mineralogical composition of PM varies with the source of origin and the size of the PM. Hematite (Fe_2O_3) is associated with larger-sized PM emitted from industrial emissions from iron ore handling (Machemer, 2004). Magnetite (Fe_3O_4) and pyrite (FeS_2) are associated with coarse particles and emitted from coal combustion and steelmaking (Gürdal et al., 2015; Jabłońska and Janeczek, 2019). These PM were identified by EDS analysis to contain high amounts of Fe and O (>50%) (Trechera et al., 2020; Galvão et al., 2022). Metallic iron and iron silicate are also associated with coarse particles (Tugrul et al., 2009). Carbonaceous particles are associated with coal and coke emissions. Silicates decrease with particle size, and sulfates are more abundant in $\text{PM}_{2.5}$, with primary industrial emissions contributing significantly (Galvão et al., 2022). SEM-EDS-based analysis of PM mineralogical composition facilitates the identification of sources, whether they are anthropogenic, geogenic, or natural. This composition also reveals chemical alterations in primary pollutants and provides insight into secondary airborne pollutants. Once sources are identified, it

becomes easier to devise strategies for reducing PM emissions through source modifications.

7 Concluding remarks

Airborne PM poses a significant health risk due to its association with various harmful substances. The majority of PM is within the respirable range for humans (<10 μm), causing adverse effects on health. Numerous studies have linked ambient PM pollution to over four million premature deaths, contributing to a range of ailments from cardiovascular diseases and respiratory disorders to cancers and viral infections. Addressing this issue requires the development of effective techniques for source identification and remediation.

In this review, an SEM-image-based method is described for PM source apportionment and their counting on the leaf surface using ImageJ software. The review focuses on the use of SEM as an efficient tool for PM analysis, coupled with EDS, providing detailed information about the morphological and chemical and mineralogical composition of PM, thus aiding in source identification. Furthermore, the inclusion of network visualization provides a clear representation of terms associated with PM on a global research platform. The review also highlights the role of ImageJ software in the SEM-based micrograph study of PM, showcasing its importance.

The combined use of both techniques allows for accessing the PM accumulation pattern on different plant leaf surfaces, along with the counting of PM in various size ranges and source identification by their chemical and mineralogical composition. This approach also assists in selecting suitable plant species for the removal of PM from the air. In summary, the review emphasizes the multifaceted nature of the relationship between PM and human health, incorporating aspects of pollution measurement, plant-mediated remediation, advanced imaging techniques, and data analysis tools. As research in this field progresses, a holistic understanding of PM dynamics will be crucial for developing targeted interventions to combat the alarming health implications of ambient air pollution.

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