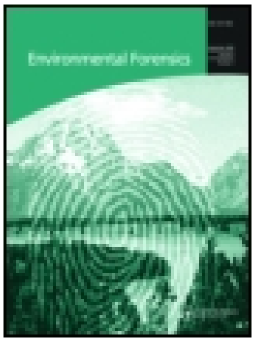

Environmental Forensics

ISSN: 1527-5922 (Print) 1527-5930 (Online) Journal homepage: https://www.tandfonline.com/loi/uenf20

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To cite this article: Muhammed Siddik Abdul Samad, Prem Mohan, George K. Varghese, Irfan Khursheed Shah & Babu J. Alappat (2019): Environmental forensic investigation of the air pollution from a cement manufacturing unit, Environmental Forensics, DOI: 10.1080/15275922.2019.1694094

To link to this article: https://doi.org/10.1080/15275922.2019.1694094

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ENVIRONMENTAL FORENSICS
https://doi.org/10.1080/15275922.2019.1694094

Environmental forensic investigation of the air pollution from a cement manufacturing unit

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ABSTRACT

Cement manufacturing is a process that results in the emission of significant quantities of suspended particulate matter (SPM) to the ambient air. An environmental forensic investigation was carried out in the surroundings of a major cement manufacturing unit at a place called Coimbatore in the southern Indian state of Tamil Nadu. The investigation was carried out to identify the contribution of the cement manufacturing unit to the SPM concentration of the surrounding air environment. The sampling points' selection and sample collection were done following the principles outlined in the INTERPOL Manual for Pollution Crime Forensic Investigation. On-site monitoring of the air samples was carried out using Mini Laser Aerosol Spectrometer (GRIMM, Mini-LAS Model 11R). The instrument was capable of measuring particles ranging from 0.25 to 32 μm and classifying them into 31 size channels. The test results at majority of the monitoring locations were well above the limits specified in the National Ambient Air Quality Standards of India. Microscopic studies of the dust samples were carried out for surface texture and particle shape. The spatial distribution of particles was analysed using geographic information system (GIS) for the visual identification of the extent of the pollution by keeping the cement factory as the focal point. The results from the GIS and microscopic analysis established the role of the cement factory in the particulate matter pollution of its surroundings, specifically in the areas North-West of the factory. The successfully adopted procedure can serve as a guideline for the environmental forensic investigation of similar pollution incidences.

KEYWORDS
Air pollution; environmental forensics; forensic investigation guideline; cement factory; suspended particulate matter; GIS

Introduction

Residents in the vicinity of industries are always under the threat of pollution of various natures resulting from industrial activities. Air pollution is one. Although sources of air pollution can be natural, like dust storm (Middleton et al., 2008; Shen et al., 2009; Zhang et al., 2011; Cesur et al., 2016; Al-Thani et al., 2018), volcanoes (Hansell and Oppenheimer, 2004; Tam et al., 2016), etc., it is the anthropogenic sources that attract the attention of regulatory agencies. Apart from industrial emissions, the major manmade source of air pollution is automobiles (Schauer et al., 2002; Hasunuma et al., 2014; Bang et al., 2018; Park and Sener, 2019). As part of air pollution abatement and control activities, regulatory agencies are often required to identify the source(s) of the pollution. In this context, there is a high demand for appropriate tools and methodologies that help easy decision-making and that are in conformity with existing standards and guidelines (Abril et al., 2016).

Among the various industrial activities, the cement plants are one of the major contributors of air pollution (Lei et al., 2011; Xu et al., 2014; Chen et al., 2015), especially particular matters (PMs) (Abdul-Wahab, 2006; Baroutian et al., 2006; Mousavi et al., 2014; Rovira et al., 2018). The dust particle from the cement factories spread over a large area through agents like wind, rain, etc. (Demir et al., 2005; Işıklı et al., 2006; Bilen et al., 2019). Because of this, the area surrounding cement factories are highly prone to air pollution. Many researchers have studied the impact of emissions from the cement factory on the surrounding environment (Abu-Allaban and Abu-Qudais, 2011; Tartakovsky et al., 2013; Liu et al., 2018). Emissions

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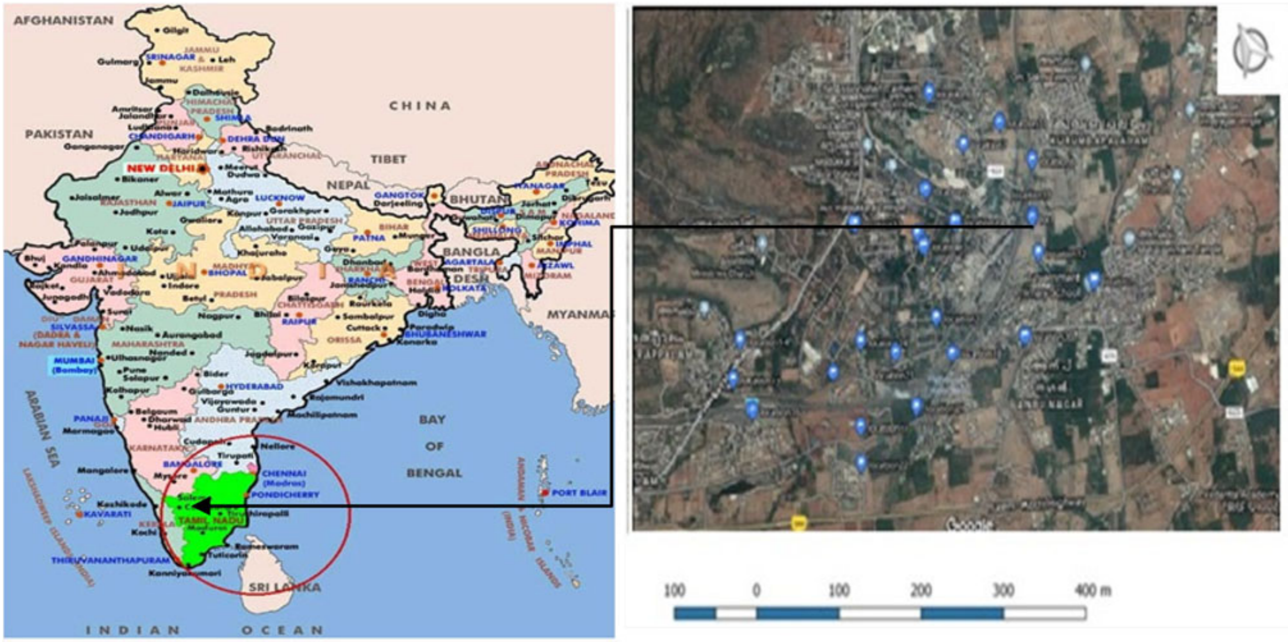

Figure 1. (a) Study area, (b) Sample locations (prepared from Google maps).

from the cement manufacturing facility consist mainly of particles with size ranging from 5 µm (clay size) to 50 µm (silt size) (United States Environmental Protection Agency [USEPA], 1993). Among particles of different sizes in the air, those ranging between 0.1 and 10 µm have significant health impacts (World Health Organization [WHO], 2005) and need to be controlled. In order to ensure clean air in the surroundings of a cement factory through proper management actions, the role of the factory in contributing pollutants in the air needs to be identified and established clearly. An environmental forensic investigation carried out at the site will accomplish this objective.

Environmental forensics involves applying scientific principles to deal with the incidents of pollution with an aim to bring the culprits before the law of the land and ensure that he pays according to the provisions of the law, for the damages he has done to the environment. Environmental forensic tools are now increasingly being used to identify potentially responsible parties in the event of environmental pollution (Morrison, 2000; Varghese and Alappat, 2012; Varghese et al., 2015), Raghunath S/o Rakhamji Lokhane vs. MPWPB & Ors (Original Application No. 11/2013(THC) (WZ)); Janardan Pharande vs. MoEF and Ors (Original Application No. 7/2014 (THC) (WZ)) and Vinesh Madanyya Kalwal vs. State of Maharashtra Ors. (Original Application No. 30(THC)/ 2013(WZ)). Recognising the importance of forensic environmental investigations in legally establishing the responsibility of pollution, the International Police Organization (INTERPOL) has published a manual for environmental forensic investigation (INTERPOL, 2014). Though the manual gives very useful guidelines for collecting environmental samples for forensic investigation, it does not address the process of environmental forensic investigation in its entirety.

This study investigated the influence of a cement factory on the surrounding air environment from a forensic perspective. Following the basic forensic principle of adopting multiple independent methodologies and tools to arrive at the same conclusion to prevent the "domino effect" (Morrison, 2000), investigations were carried out using different independent approaches. The air samples were tested for the identification of PMs of various sizes using portable laser aerosol spectrometer. The dust deposited on the instrument filter and overground points and tree leaves in the areas surrounding the cement factory were subjected to scanning electron microscope (SEM) analysis for identification of the surface texture, shape, etc. of particulates. These were then compared with SEM images of the cement particles from the alleged source. A spatial analysis of the PM concentrations vis-a-vis the wind pattern in the area was also undertaken to arrive at conclusions regarding source.

Description of study area

The study was carried out in the area surrounding a cement factory at *Madukkarai panchayat* in Coimbatore district, Tamil Nadu, India (Figure 1a). The study area is bounded by the Western Ghats mountain ranges on the west and north and is situated in the Palakkad Gap (a low, but wide, mountain pass in the Western Ghats connecting the Coimbatore district of Tamil Nadu and the Palakkad district of Kerala). The area is popular for its waste management initiative named "green friends", where the household wastes are collected by the members of the community itself and disposed of in a sustainable way. This initiative is supported by the cement factory located in this area. Even though the company is known to manage its surrounding environment in a sustainable way, due to the huge annual production and handling of cement (approx. 1.18 million tonnes of cement), there is a possibility of particulate matter spreading in the area.

Methodology

An environmental forensic investigation is initiated when there is a suspected or alleged incident of pollution and there is a party who has an interest in the incident who is ready to fund the investigation. The party could be any private entity or a government agency. Very often, it is in the interest of the party funding the investigation to know if the benefits of investigation outweigh the costs of the investigation. Thus, it is always recommended to do a preliminary investigation before deciding to go for a costly detailed investigation. A general methodology reflecting this approach and its explanation are given as supplementary material. The methodology adopted specifically for this investigation, conforming to the general methodology, is shown in Figure 2.

The methodology adopted makes use of the following three-tier approach: 1) Carry out air monitoring in the study area for particulate matter of different sizes, analyse the data statistically, and compare it with various existing air quality standards to establish pollution of the area. Also, use the data to prepare a hypothesis on possible source(s), 2) Use SEM images of the samples and controls to further confirm the source of pollutants, and 3) Use the geographical representation of PM concentration distribution around the alleged source to corroborate the findings from previous steps.

Sampling and analysis

The sampling locations were identified based on the principles laid by INTERPOL Pollution Crime Forensic Investigation Manual – Vol. 1 (INTERPOL, 2014). GRIMM Mini-LAS Model 11 R (Germany) was used to find the particulate matter concentration at 25 different sampling points that were chosen at various distances surrounding the industry. These sampling stations were numbered *Location 1* to *Location 25* (Figure 1b). The particulate matter concentrations (PM_{10}, $PM_{2.5}$, and PM_1) were found out at these locations. The instrument was pre-calibrated and readings were obtained at 6-second interval. The instrument was kept at a height of 1.5 m from the ground level. Sampling points were selected such that there were no obstructions to air flow. The sampling was done when there was no heavy wind blowing. Temperature, humidity, and wind speeds were also taken to confirm the uniformity in atmospheric conditions. GPS coordinates were recorded at each sampling location. Background readings at a location far from the industry were also taken.

For understanding the physical nature of the dust particles deposited from the atmosphere, dislodgeable dust was collected from plant leaves and analysed using SEM (Hitachi SU6600 Field-Emission SEM, Japan). The leaves plucked carefully from trees/plants at a location were tapped using a glass rod to dislodge the deposited dust to a watch glass. The dust from watch glass, representative of the deposition at that locality, was transferred to polythene bags for transporting to lab for SEM analysis. PM trapped in the filter of GRIMM Mini-LAS Model 11R was also collected carefully and analysed using SEM. The cement sample and soil from the location were used as the background/control samples.

Spatial analysis

Traditionally GIS is used for spatial analysis of the pollution (Jensen, 1998; Briggs 2005; Gulliver and Briggs, 2011; Ketzel et al., 2011). Spatial interpolation (Briggs et al., 1997) of the pollution measurements made in the monitoring stations was done to get concentrations at places where readings were not taken. A wide range of spatial interpolation tools is available (Burrough, 1986). Several studies compared the performance of the various interpolation methods (Van Kuilenburg et al., 1982; Weber and Englund, 1992; Knotters, et al., 1995), but it was observed that none was optimal. The results mainly depended on the data collected. The Inverse Distance Weighting (IDW) interpolation method integrated into QGIS software was used in the current study. The spatial distribution in the air of three different types of particulate matter (PM_{10}, $PM_{2.5}$, and PM_1) was analysed using GIS.

Results and discussion

Particulate matter concentration

The PM_{10}, $PM_{2.5}$, and PM_1 concentrations ($\mu g/m^3$) measured at the sampling locations are tabulated in Table 1.

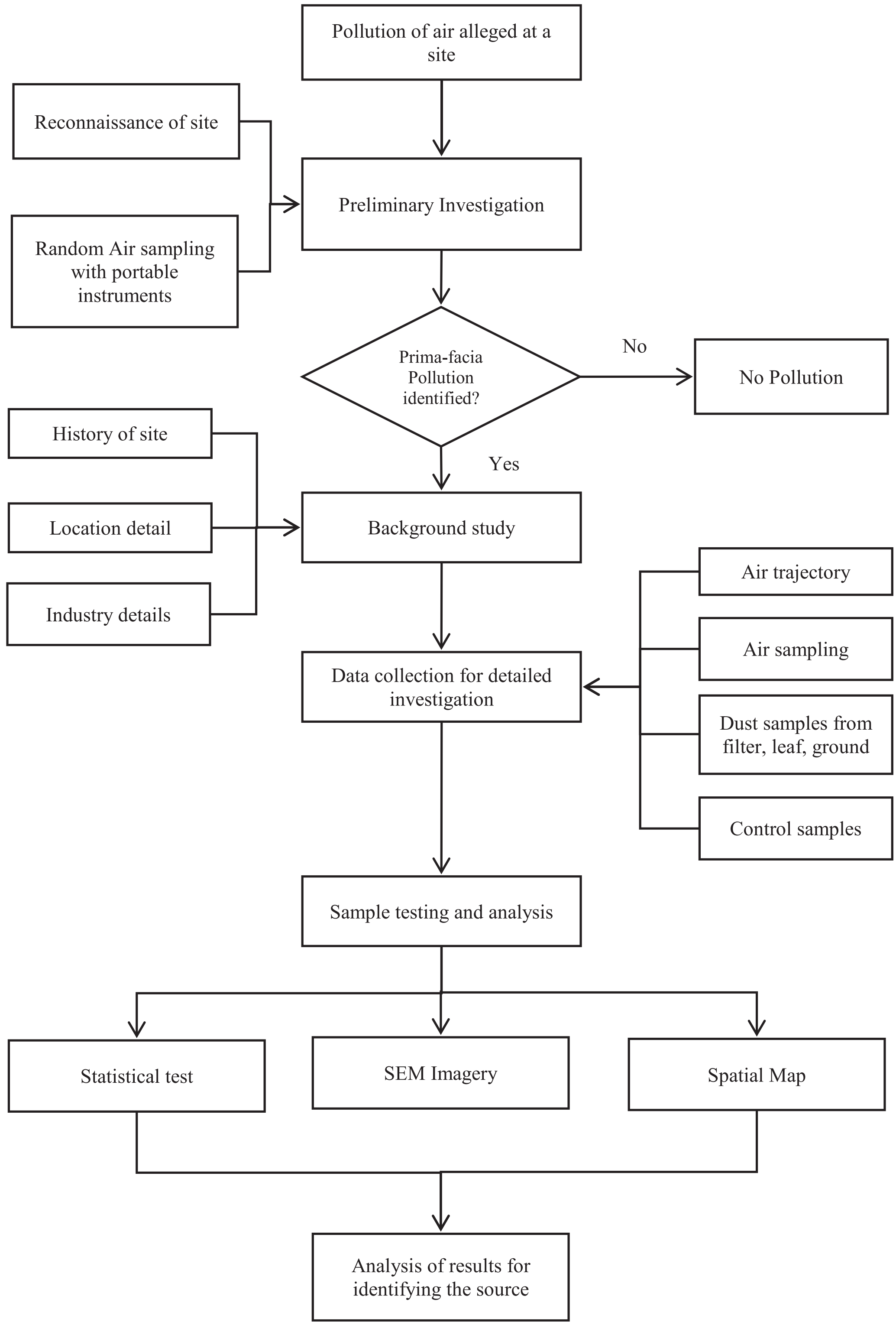

Figure 2. Case-specific methodology.

Temporal variation of particulate matters

The average of maximum and minimum values of PM_{10}, $PM_{2.5}$, and PM_1 were 299.86, 90.44, 40.4 and 56.01, 35.19, 37.4 µg/m^3, respectively. The mean values of PM_{10}, $PM_{2.5}$, and PM_1 were 108.28, 45.76, and 39.10 µg/m^3, respectively. The average values of maximum PM_{10} and $PM_{2.5}$ concentrations were, respectively, 3 and 1.5 times higher than the average value stipulated by National Ambient Air Quality Standards: Central Pollution Control Board. Also, they are,

Table 1. Concentration of PM_1, $PM_{2.5}$, and PM_{10} in the study area (μg/m^3).

	Minimum			Maximum			Mean			Std. Dev.			Variation Coeff.			Median		
Locations	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}
BG	32.2	35.6	46.7	35.3	44	83.1	34.1	39.2	59.9	0.7	2	8.7	0	0.1	0.1	34.2	39.4	58.5
1	36.3	39.5	46	52.3	204.7	828.4	39.2	52.1	105.3	3	19.3	89.7	0.1	0.4	0.9	38.2	47	84.3
2	28.8	33.1	57.5	115.1	174.6	542.8	33.7	48.8	130.9	15.1	28.8	107.6	0.4	0.6	0.8	30.4	39.6	95.6
3	29.3	33.4	66.4	32.2	73.3	414.9	30.6	46.7	145.4	0.8	8.7	69.9	0	0.2	0.5	30.6	46.7	133
4	28	34.7	65.8	33	66.6	232.7	29.9	49.4	149	1.3	9.2	47	0	0.2	0.3	29.8	49	151.5
5	28.9	31	48.4	57.7	133.1	1263.1	32.4	44.1	185.7	5.3	19.5	250.6	0.2	0.4	1.3	30.9	39.2	119.4
6	27.8	31.4	49.2	59.6	192.4	385.8	32.7	60.3	175.5	7.1	35.5	94	0.2	0.6	0.5	30.5	44.9	139.8
7	29	32.5	45.5	31.5	45.7	98.7	30.2	38.1	71.9	0.7	3.2	13.8	0	0.1	0.2	30.1	37.6	70.5
8	30.9	33	43.4	34.1	50.5	148	31.8	39	67.7	0.7	3.5	23.3	0	0.1	0.3	31.9	38.6	61.5
9	28.9	33	56.7	31.5	48.9	107.7	30.6	38.7	75.7	0.6	4	13.7	0	0.1	0.2	30.6	37.7	73.4
10	29.7	32.1	47.8	43.5	301.3	378.3	31.6	48.7	92.3	2.7	55.2	75.6	0.1	1.1	0.8	31.3	37.1	65.3
11	21.4	23.9	31.9	25.4	37.1	61	23.9	28.2	45.7	1	2.7	8.6	0	0.1	0.2	24.2	28.1	45.2
12	21.1	23.4	32.3	34.7	50.3	134.8	23.2	29.2	56.1	2.8	5.6	26.5	0.1	0.2	0.5	22.4	27.7	48.8
13	23.5	27.3	122.5	26.5	61.3	357.6	24.9	44.4	189.3	0.7	7	46.8	0	0.2	0.2	24.8	45	179
14	23.6	31.6	50.2	55.2	78.4	194.8	32.6	48.2	128.9	7.2	11.8	37.2	0.2	0.2	0.3	30.9	44.1	136.1
15	22.2	25.2	50	28.5	50.1	320.5	24.3	35	119.1	1.6	6.2	59.5	0.1	0.2	0.5	24.1	33.1	103.3
16	22.8	28.4	48	52.3	63	185.8	27.3	35.6	82.8	7.9	8.6	32.1	0.3	0.2	0.4	24.7	32.9	73.3
17	24	25.6	36.4	29.1	43.6	81	26.4	32.1	55.2	1.3	3.4	10.9	0	0.1	0.2	26.9	31.8	53.7
18	25.9	28.5	45.7	37	83	247.7	28.5	41.5	102.4	2.9	13.5	51.4	0.1	0.3	0.5	27.8	37	91.6
19	44.9	49.5	68.3	64.3	117.2	535.3	50.4	63.7	141.5	4.6	14.8	89.6	0.1	0.2	0.6	49.5	58.6	116.6
20	49.5	54.4	65.5	52.3	63	117.8	51.1	58.2	79.5	0.7	2.3	11.9	0	0	0.2	51.2	57.9	77.1
21	44.6	47.9	60	55.7	66.5	97.3	47.1	53.4	81.7	3.5	5.2	10.7	0.1	0.1	0.1	45.8	51.8	84
22	42.4	46	56.1	45.7	55.8	88.4	44	49.8	70.9	0.8	2.8	9.8	0	0.1	0.1	43.8	49.3	70.9
23	42.8	48.9	77.9	49.2	94	434.1	44.9	64.4	177.2	0.9	7.5	48.7	0	0.1	0.3	45	62.5	169.5
24	36.9	42.7	62.1	40.7	50.9	105.6	38.8	46.2	80.3	1.1	2.4	14.9	0	0.1	0.2	38.5	45.8	76.8
25	37.4	42.8	66.8	40.4	55.7	134.5	39.1	48.4	95.7	0.8	3.2	20.6	0	0.1	0.2	39.2	48.2	91.5

BG: Background location.

respectively, 2 and 2.5 times higher than the daily USEPA standards (PM_{10}= 150 μg/m^3 and $PM_{2.5}$ = 35μg/m^3) (USEPA, 2012). These values are approximately 15 and 9 times higher than the annual limits set by WHO (PM_{10} = 20 μg/m^3 and $PM_{2.5}$ = 10 μg/m^3) (WHO, 2005) and 7.5 and 3.5 times higher than the limits of European Union Air Quality Annual Standards (PM_{10} = 40 μg/m^3 and $PM_{2.5}$ = 27μg/m^3), respectively.

The mean concentrations of PM at the sampling locations are shown in Figure 3. The PM_{10} concentrations were higher compared to the background value taken from an area not affected by the industrial activities (approximately 3 km away from the industry, in the upwind direction). The $PM_{2.5}$ and PM_1 concentrations at all the sampling locations were close to the background value, which indicated that the PM_1 and $PM_{2.5}$ were distributed wider in the atmosphere. Studies have shown that particulate emission from cement industries contains mostly particles above 5 μm (USEPA, 1993; Gupta et al., 2012). Thus, the higher PM_{10} noticed in the vicinity of the cement plant was attributable to the plant.

Wind direction

The wind direction at the study area was obtained from HYSPLIT Model. The HYSPLIT model is a complete system for computing the air trajectories for complex dispersion and deposition simulation (Draxler, 2018). Figure 4 shows the trajectory frequency of the study area at the time of sampling. The trajectory frequency archived by the HYSPLIT model shows that the flow of air was towards the North-West direction. The wind direction and velocity were measured locally at the time of sampling using wind wane and anemometer (Amprobe TMA40-A). The wind direction noticed was conforming to that obtained from HYSPLIT model archive.

Scanning electron microscope analysis

SEM analyses were done on the dust particles collected from the leaf of the plants near the industry and on the sample collected from the filter paper in MiniLAS. SEM images of the control samples (soil and cement) were taken. The images were of 20k magnification. Figures 5–9 show the SEM images.

From the SEM images and the details of the image developed using ImageJ software (Schneider et al., 2012) shown in Table 2, it can be inferred that the dust deposited on leaf 1 (Figure 6) matches to the soil sample (Figure 8) collected from the surroundings of the industry, and the SEM images of leaf 2 (Figure 7) and dust particles in the filter paper (Figure 5) are similar to the SEM images of cement particles

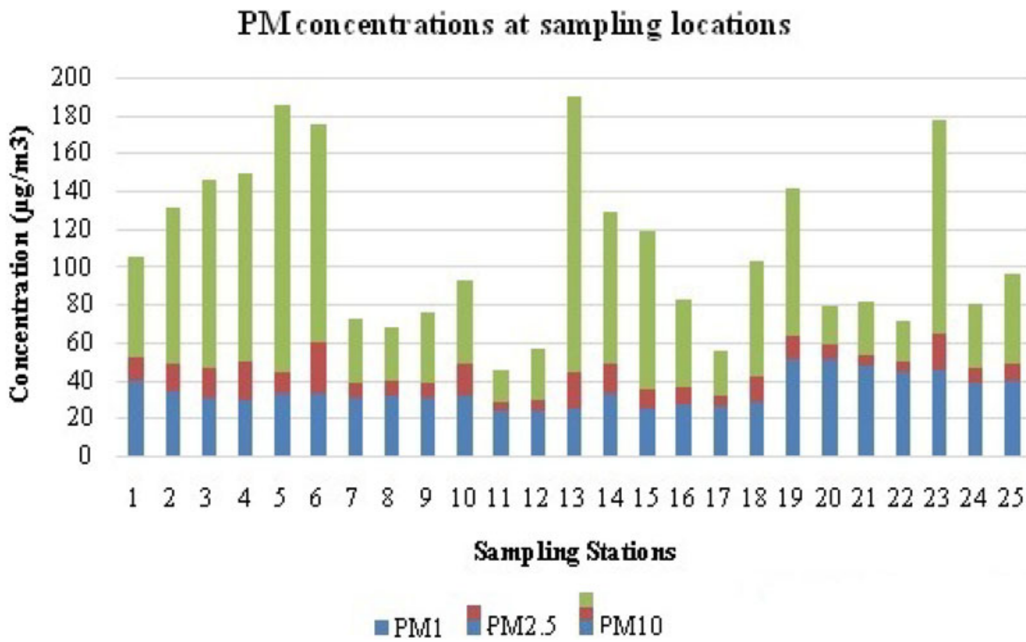

Figure 3. Distribution of PM in the sampling locations.

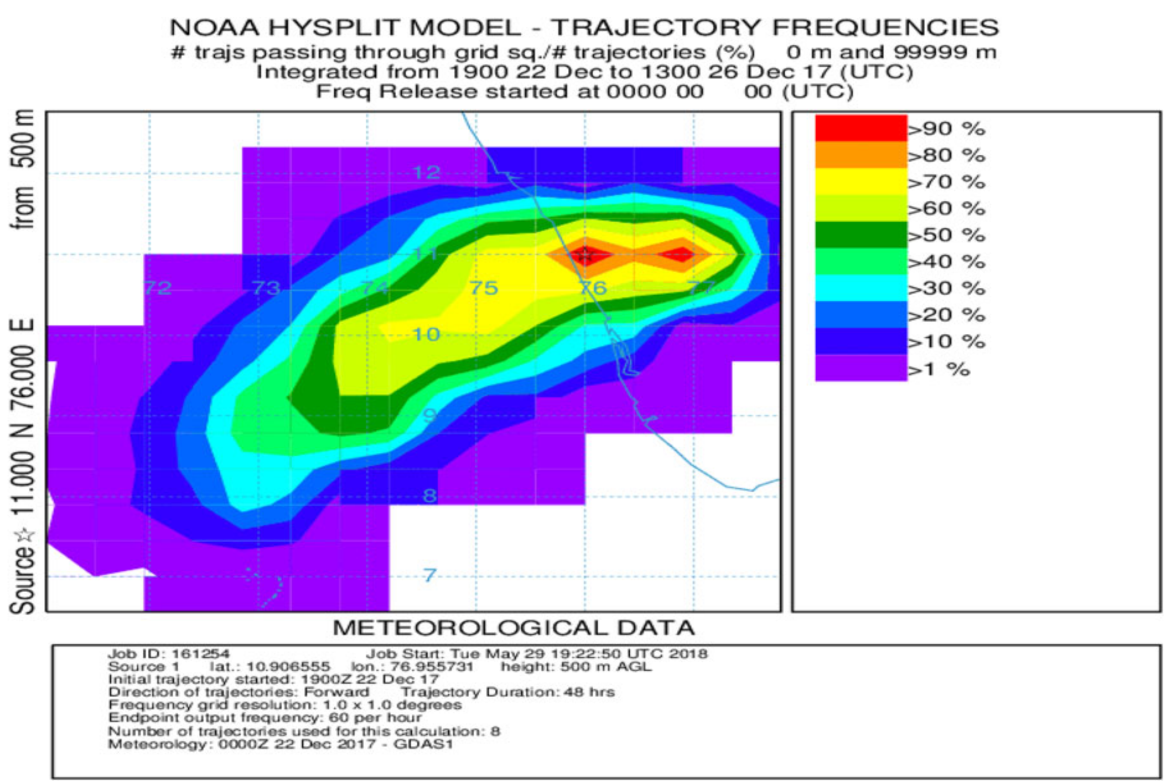

Figure 4. Air trajectory at the study area (Courtesy: HYSPLIT MODEL).

(Figure 9). From these observations, it can be concluded that the particulate matter concentration in the surrounding area is mostly contributed by the industry. In addition to the industrial influence, the wind and traffic in the area resulting in the wake of dust from the ground have a possible role in the contribution of particulate matter in the atmosphere.

GIS analysis

The spatial distribution of the PM in the study area was developed using the Quantum GIS software package (QGIS Development Team, n.d.). The output images show that the spread of the PM_{10} was prominent towards the North-West direction of the factory area (Figure 10).

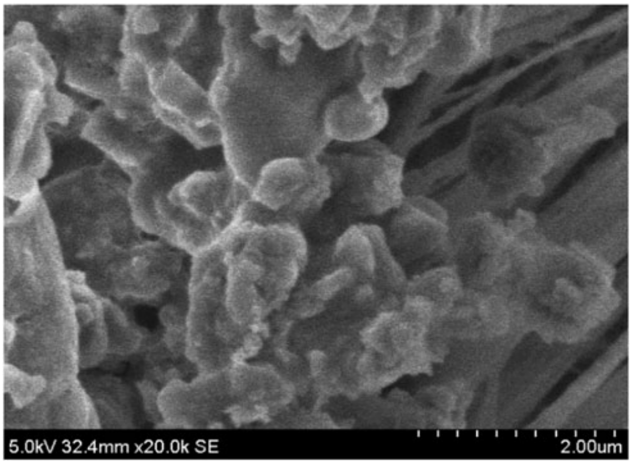

Figure 5. Particles on filter paper.

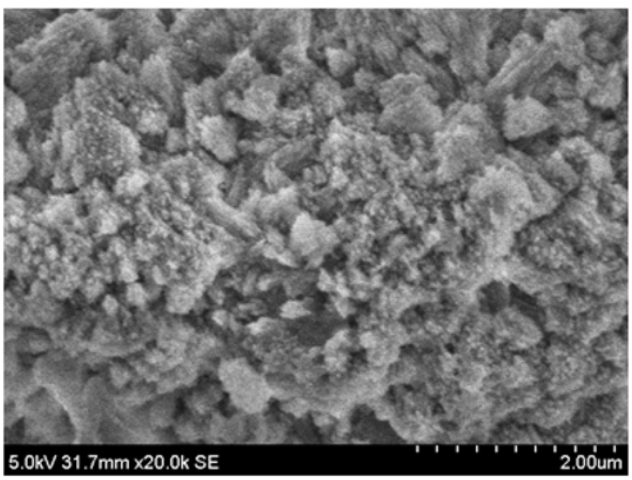

Figure 6. Leaf sample 1.

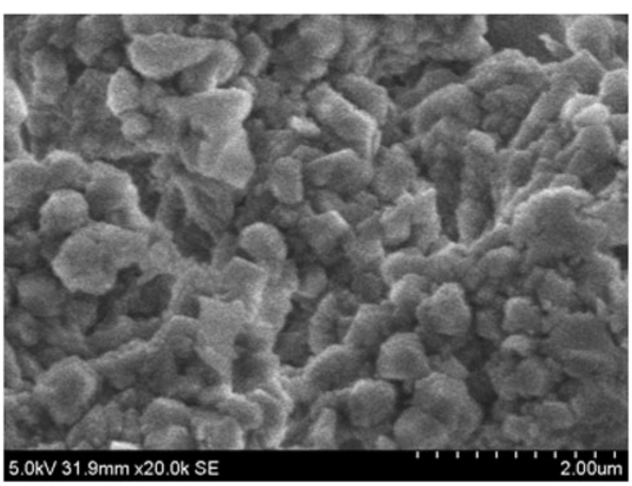

Figure 7. Leaf sample 2.

The direction of the wind was towards the North-West of the cement factory. $PM_{2.5}$ and PM_1 are somewhat uniformly distributed around the factory. (Figures 11 & 12) Also, it is observed that the PM_1 and $PM_{2.5}$ distribution has no clear centres unlike the case of PM_{10}.

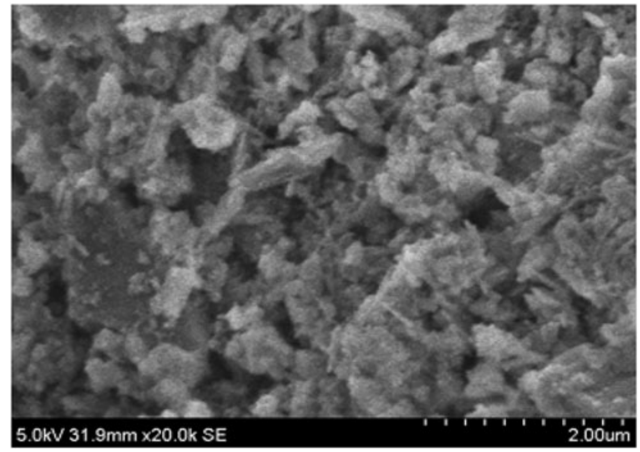

Figure 8. Soil sample.

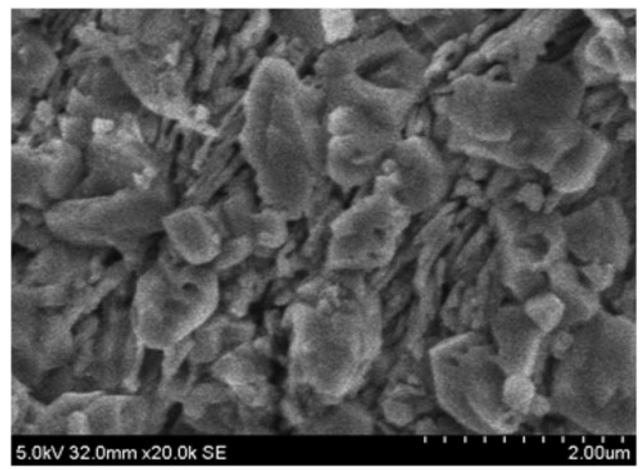

Figure 9. Cement sample.

Table 2. SEM image details developed using ImageJ software.

Sample	Area	Mean	Std. Dev.	Min.	Max.	Median
Leaf 1	119002	113.838	47.263	0	255	115
Leaf 2	117315	119.605	44.862	0	255	119
Soil	104144	114.253	46.724	0	255	115
Cement	102729	105.453	44.89	0	255	105
Filter paper	148630	109.207	45.157	0	255	108

Conclusion

Air pollution contributed by industries is a concern. In this regard, the cement industries are one of the lead contributors, especially in the case of particulate matter in the air. The study analysed the air quality of the area surrounding a major cement factory, by testing for the presence of PM in the air. The test results were analysed in three different ways: statistical analysis, using SEM images, and spatial analysis using GIS. It was found that the average values of the PM_{10} and $PM_{2.5}$ were much higher than the air quality standards stipulated by various regulatory agencies.

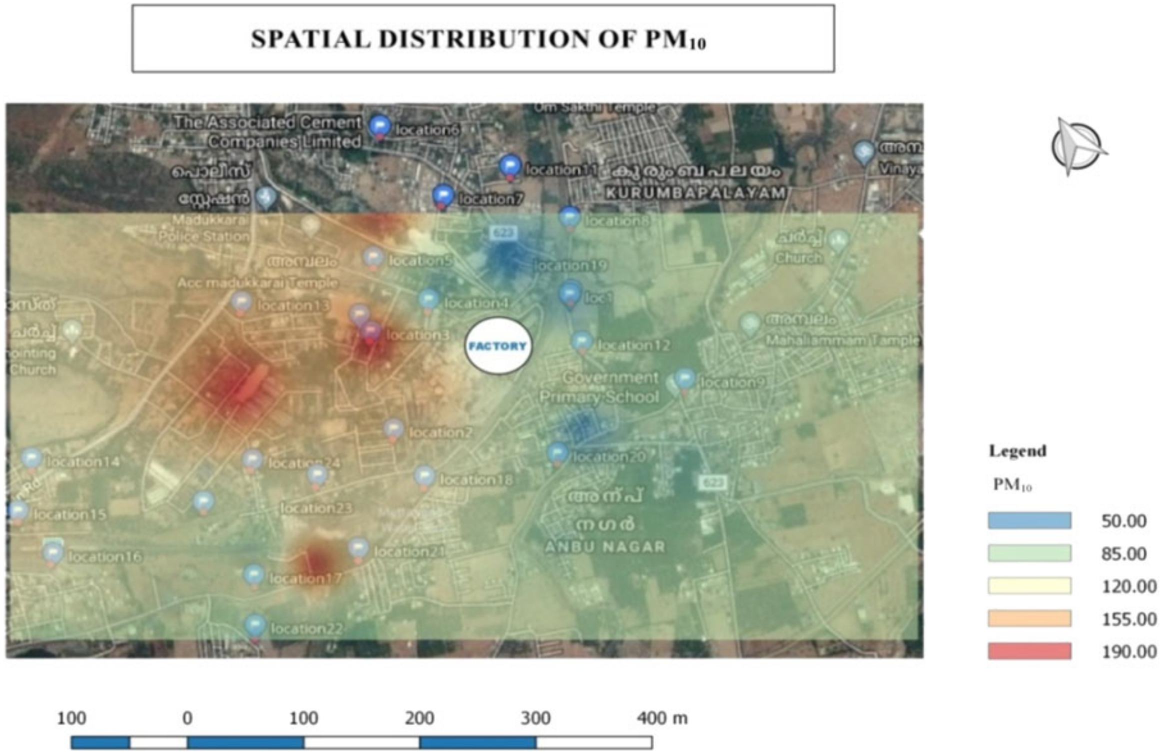

Figure 10. Spatial distribution of PM_{10}.

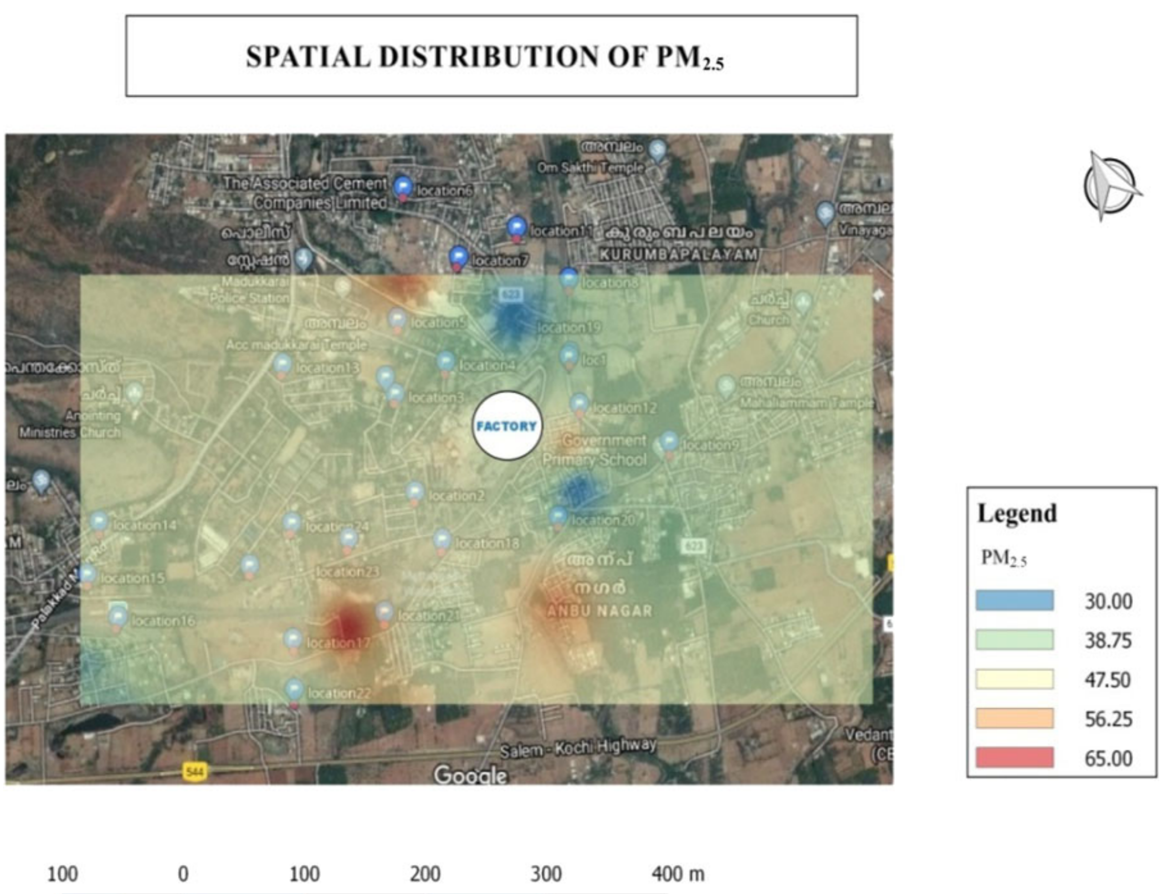

Figure 11. Spatial distribution of $PM_{2.5}$.

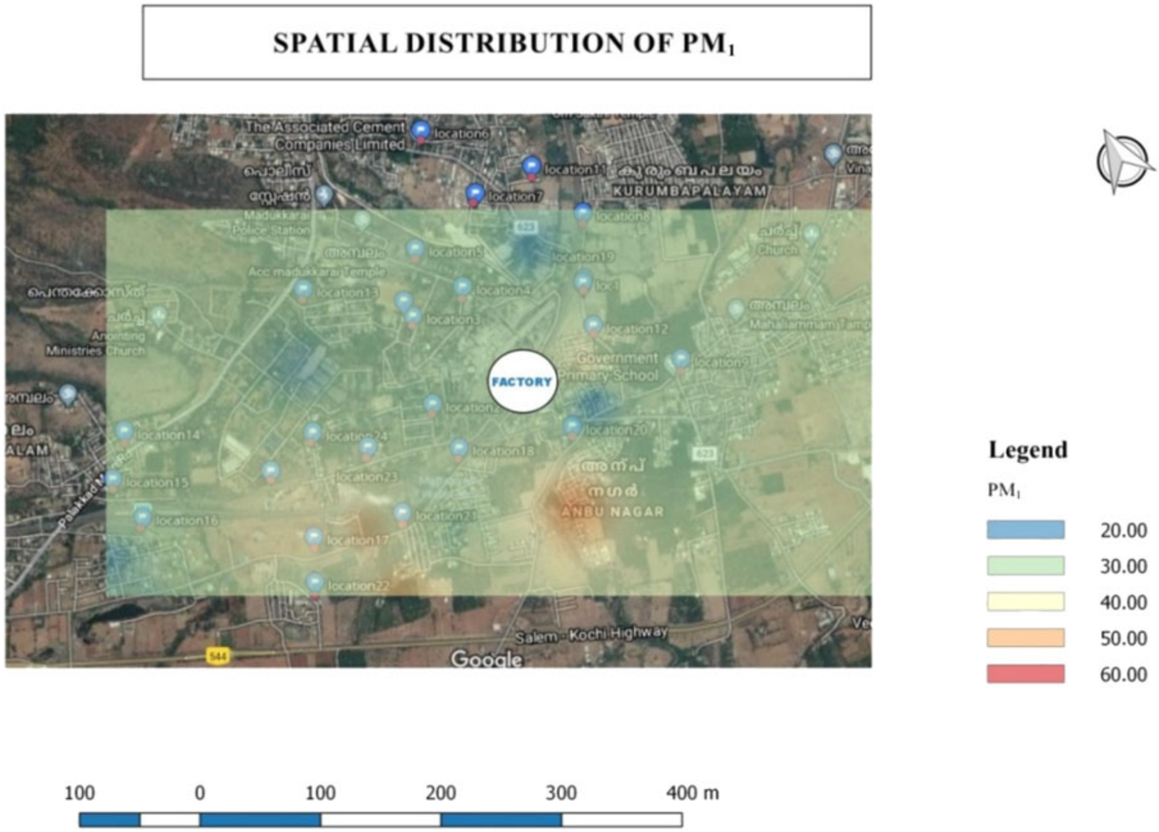

Figure 12. Spatial distribution of PM_1.

The PM_{10} concentration in the study area was considerably higher than the specifications of air quality standards. It was also significantly higher than the background concentrations taken from a site, far off the cement factory. But, PM_1 and $PM_{2.5}$ were found to be distributed nearly uniformly all throughout. As the size of particulates emitted from cement factory mostly range from 5 to 50 µm, and the PM_{10} values at places near the cement industry falling in the direction of wind are very high, it can be concluded that the particulates of sizes above 5 µm in PM_{10} are contributed by the cement industry. This conclusion is further confirmed by the similarity between the SEM images of particles collected in the analyser, and deposited in tree leaves and cement particles. A pictorial representation of the PM concentration distribution using GIS made the conclusions regarding the source easier to demonstrate even to a layman.

There can be many incidents of pollution similar in nature to the one investigated. The methodology adopted for investigation here can be used as a guideline in such incidents of pollution.

Acknowledgement

The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for providing access to the archives of their HYSPLIT transport and dispersion model repository (http://www.ready.noaa.gov).

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