



Compositions of Dust Fall around Semi-Urban Areas in Malaysia

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ABSTRACT

This study aimed to determine the amount and composition of dust fall in semi-urban areas in Selangor, Malaysia from mid-July to mid-October 2010. The dust fall samples (a combination of dry and wet deposition) through rainwater were collected using polyethylene bottles on a monthly basis so as to determine the concentration of total solids, soluble and insoluble solids from eight monitoring stations. The concentration of cations (Ca^{2+} , K^{+} and Mg^{2+}) and heavy metals (Pb, Fe, Zn, Al, Cr and Cd) were analyzed using an inductively coupled plasma-mass spectrometer (ICP-MS) while the concentration of anions (F^{-} , Cl^{-} , NO_3^{-} and SO_4^{2-}) in the rainwater was determined through ion chromatography (IC). Results showed that the average concentration of total solids in the dust fall ($131.50 \pm 71.95 \text{ mg/m}^2/\text{day}$) below the value of $133 \text{ mg/m}^2/\text{day}$ as recommended by the Malaysian Department of the Environment. The dominant anion in rainwater was found to be SO_4^{2-} followed by NO_3^{-} , Cl^{-} and F^{-} , and the dominant cation was Ca^{2+} , followed by K^{+} and Mg^{2+} . Zn was recorded as the dominant heavy metal in rainwater followed by Fe, Al, Pb, Cd and Cr. This study suggests that sea salts, earth crust and anthropogenic activities such as motor vehicles were the main sources of dust fall in the study areas.

Keyword: Dust fall; Particulate matter; Semi-urban area; Anion; Cation; Heavy metals.

INTRODUCTION

Urbanization is a process which involves economic and industrial development and consequently population growth. This in turn leads to higher energy production and consumption, resulting in problems related to air pollution. According to Fenger (1999) and Cao *et al.* (2011), air pollution is a major problem in urban areas due to emissions from transportation and the interaction of air pollutants originating from a variety of sources and may have a significant effect on the environment. Emissions from the urban atmosphere which originate from anthropogenic activities, such as the use of motor vehicles, open burning, coal burning and industrial emissions (Ashbaugh *et al.*, 2003; Abdul-Wahab 2006; Xia *et al.*, 2007; Cao *et al.*, 2011) are the major sources of air pollution in Malaysia (Afroz *et al.*, 2003). Chemical reactions between pollutants and the atmospheric components generally result in the deposition of the pollutants and their derivatives upon the earth's surface when brought down by rainfall (Afroz *et al.*, 2003) or dry

precipitation (Jaradat *et al.*, 2004; Satyanarayana *et al.*, 2011).

Air pollutants can be emitted in the form of particulate matter or gases which are discharged directly or through secondary processes (Duh *et al.*, 2008). Particulate matter with a size of greater than $10 \mu\text{m}$ and the ability to settle from the air is the main component of dust fall (Ahmad, 1975; Cadle, 1975; Espinosa *et al.*, 2001; Sami *et al.*, 2006). Dust fall makes a significant contribution when ascertaining pollution in urban areas (Harrison *et al.*, 1997; Xia *et al.*, 2007; Balakrishna *et al.*, 2010), industrial activities and at construction sites (Fuglsang, 2002; Pandey *et al.*, 2008) as well as from vehicle exhaust emissions and soil dust (Arsene *et al.*, 2007). In order to understand regional dust pollution, the dust fall rate and its chemical constituents must first be determined (Harrison, 1986; Pandey *et al.*, 2008). Several studies, for example, by Sharma *et al.* (2008) and Chabas *et al.* (2010) stated that dust fall makes an important contribution to soil, water and the destruction of forests and buildings. Moreover, the deposition and settling of heavy metals which accumulate in soil can in turn enter the food chain (Sharma *et al.*, 2008) and consequently be found in the human circulatory system (Gulson *et al.*, 1995). It is important therefore to identify the composition of dust fall in order to avoid other effects, for instance, acid rain, which often occurs in both industrial and urban areas. Acidic rainwater itself predominantly affects the environment

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(Brimblecombe, 2001; Arsene *et al.*, 2007) through causing damage to buildings, producing harmful chemicals and corroding piping, which may subsequently also affect drinking water supplies (Balasubramanian *et al.*, 1999; Rodhe *et al.*, 2002; Hu *et al.*, 2003; Das *et al.*, 2005; Nagase and Silva, 2007).

The objective of this study was to determine the level of dust fall as well as its composition in terms of heavy metals, anions and cations, at eight sampling stations located in semi-urban areas in Selangor, Malaysia. This, in effect, would indicate how influential urbanization was on the concentration and composition of dust fall.

MATERIALS AND METHODS

Study Area

Sampling was undertaken at eight sampling stations around semi-urban areas in Kajang and Bandar Baru Bangi, Selangor, Malaysia. (Fig. 1) (Table 1).

Kajang

Kajang is a town in the eastern part of Selangor state, Malaysia and the district capital of Hulu Langat. It is located 21 kilometers from Malaysia's capital, Kuala Lumpur and has a rapidly growing population (Latif *et al.*, 2006). The area in which it is located is predominantly characterized by intensive commercial activity, which, with a rapidly increasing population, has led to severe traffic congestion during the morning and afternoon rush hours.

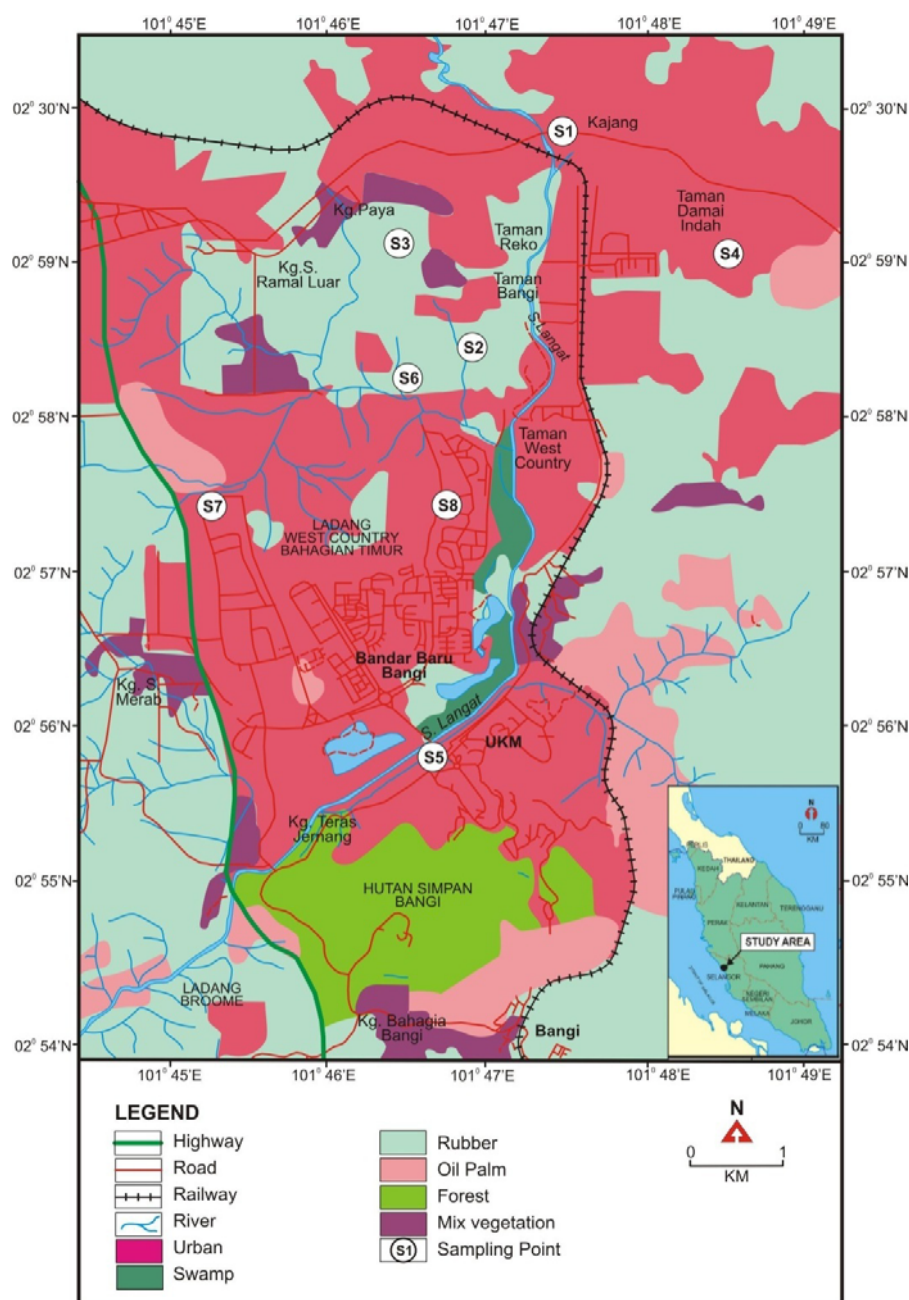


Fig. 1. Location of sampling stations in the study area.

Table 1. Sampling locations.

Station	Study area	Description	Latitude	Longitude
S1	Kajang	Roadside, fast developing area with high volume of traffic	2°59'50"N	101°47'18"E
S2	Kajang	Residential area	2°58'25"N	101°46'56"E
S3	Kajang	Residential area near a busy town	2°59'7"N	101°46'24"E
S4	Kajang	Residential area	2°59'9"N	101°48'35"E
S5	Bangi	Roadside, main road	2°55'47"N	101°46'39"E
S6	Bangi	Residential area	2°58'20"N	101°46'28"E
S7	Bangi	Fast developing area with high volume of traffic	2°57'31"N	101°45'10"E
S8	Bangi	Residential area	2°57'29"N	101°46'46"E

Bangi

Bangi is a semi-urban area situated in the south of Kajang. This area is developing fast, particularly Bandar Baru Bangi (the new town of Bangi) and contains a large population and residential area with a high level of vehicles usage. Before Bangi was developed into a residential area spreading out from Kajang Town, it had a great number of plantations, predominantly for rubber and palm oil.

Sampling Method

Dust fall consisting of wet and dry deposition was collected once a month, from July to October 2010, using polyethylene bottles equipped with a funnel (diameter size 5 cm). The containers were put on the roof of building at a height above 2 m so as to avoid any interference from soil or surface dust. In order to prevent lichen forming during the sampling period, 10 mL of 0.02 N copper sulphate ($\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$) solution was added to each container (BSI, 1969). The amount of dissolved SO_4^{2-} from $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ has been considered for SO_4^{2-} determination in rainwater. After samples were collected, they were filtered using Whatman glass microfiber filters GF/C (47 mm diameter with a 0.45 μm pore size) to determine the composition of insoluble matter and then stored (4°C) before other analyses were carried out.

Determination of Insoluble Matter

The filter paper after filtration of the rainwater was dried in an oven at 105°C for 2 hrs, then kept in a desiccator before being weighed a further three times until the weight of filter paper and insoluble materials became constant. The concentration of insoluble materials was calculated following the method stated by Norela *et al.* (2009) based on the different weight of filter papers (before and after the sample was filtered), number of days (in one month) and the area of the separating funnel (which was calculated based on the diameter of the separating funnel) as Eq. (1):

$$\text{Concentration of insoluble solids (mg/m}^2\text{/day)} = \frac{(M_2 - M_1)}{AT} \quad (1)$$

where,

M_1 = Weight of the filter without sample (mg)
 M_2 = Weight of the filter with dry insoluble solids after filtration (mg)
 A = Funnel surface area (m^2)
 T = Period of sampling days (days)

Determination of Total Soluble Matter

The filtrated rainwater (100 mL) was concentrated to around 50 mL in a flask on a hot plate. The sample was then placed on the pre-weighed evaporating dish until dried. The sample in the evaporating dish was subsequently dried in the oven at 105°C for 2 hrs and then cooled in a desiccator before being weighed. The concentration of total soluble solids was calculated following the method stated by Latif and Rozali (1999) based on the different weights of the evaporating dish (before and after the drying of the sample), the volume of rainwater collected and the water used for analysis (100 mL), the number of days (in one month) and the surface area of the separating funnel (which was calculated based on the diameter of the separating funnel) as Eq. (2):

$$\text{Concentration of soluble solids (mg/m}^2\text{/day)} = \frac{[(M_4 - M_3)(V_1 / V_2) - 18.5]}{AT} \quad (2)$$

where,

M_3 = Weight of evaporating dish without the soluble matter (mg)
 M_4 = Weight of soluble matter with the weight of the evaporating dish (mg)
 V_1 = Volume of all the rainwater collected (mL)
 V_2 = Volume of rainwater which evaporated (100 mL)
 A = Funnel surface area (m^2)
 T = Period of sampling (days)
 18.5 = Factor for dissolved solids from 10 mL 0.02 N $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ (BSI, 1969)

Determination of Anions, Cations and Heavy Metal in Rainwater

Anions (SO_4^{2-} , NO_3^- , Cl^- and F^-) were determined using ion chromatography (Metrohm 850) with method detection limits of 2.3 $\mu\text{g/L}$ for SO_4^{2-} , 2.5 $\mu\text{g/L}$ for NO_3^- , 1.2 $\mu\text{g/L}$ for Cl^- , 0.7 $\mu\text{g/L}$ for F^- , while cations (K^+ , Mg^{2+} , Ca^{2+}) and heavy metals (Pb, Cd, Fe, Zn, Al and Cr) were determined using an inductively coupled plasma-mass spectrometer (ICP-MS, PerkinElmer ELAN 9000) with method detection limits of 0.0005 $\mu\text{g/L}$ for Pb, 0.00009 $\mu\text{g/L}$ for Cd, 0.0003 $\mu\text{g/L}$ for Fe, 0.0003 $\mu\text{g/L}$ for Zn, 0.005 $\mu\text{g/L}$ for Al, 0.0002 $\mu\text{g/L}$ for Cr, 0.006 $\mu\text{g/L}$ for K^+ , 0.005 $\mu\text{g/L}$ for Mg^{2+} and 0.004 $\mu\text{g/L}$ for Ca^{2+} . The standard solutions of the heavy metals were prepared by the dilution of each standard solution (1000 mg/L) with ultra pure water in volumetric flasks. The standard solution for anions were prepared by

dilution of 1000 mg/L stock solution of SO_4^{2-} , NO_3^- , Cl^- and F^- with ultra pure water.

Statistical Analysis

In order to compare the concentration air pollutants between stations, several statistical analyses were conducted to determine the distribution of air pollutants' concentrations using the Normal P–P Plot, the Normal Q–Q Plot and the One-Sample Kolmogorov–Smirnov test. Correlation coefficient and correlation significance were analyzed using the one-way ANOVA test and the *t*-test using the Statistical Package for Social Sciences (SPSS Version 12) so as to estimate any notable differences between the mean concentration of total dust fall, insoluble materials, soluble materials, heavy metals, cations and anions in rainwater. Meanwhile, cluster analysis was done to group the data based on the same characteristic (Lu *et al.*, 2011) and the possible sources of anion, cation and heavy metals concentration was analyzed using Principal Component Analysis (PCA) which can help in reducing the variables to smaller factors that can give the most information from the various data (Khotai *et al.*, 2008). The XLSTAT 2012 software was used to analyze cluster analysis and PCA.

Trajectory Analysis

A 72 h backward kinematics trajectory was computed from 0600 UTC located at 100 m A.G.L. above the centroid location of the study area (2.98°N; 101.78°E). The selected height level of the particles release ensured that the trajectories started in the atmospheric boundary layer (Eva and Lambin, 1998). The back trajectories were calculated using version 4.9 of the Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT) developed by the National Oceanic and Atmospheric Administration (NOAA)'s Air Resource Laboratory (Draxler and Rolph, 2003; Rolph, 2003). The meteorological drivers used to compute the trajectories were obtained from the NCEP Final Analysis (FNL) archive which is maintained by ARL.

Quality Control

In order to ensure that the results of dust fall measurements were not affected by any interference, all polyethylene bottles and glassware utilised for dust fall, heavy metal, anion and cation analysis were treated using a 20% nitric acid bath before being rinsed several times

using ultra-pure water with a 18.2 MΩcm resistivity. The calculation of anionic balance was done by summing up the equivalent concentrations of cations and anions of the samples. In our case the difference between the sum of anions and cations were not equal because we only measured selected anion and cations.

RESULTS AND DISCUSSIONS

Deposited Particulates

Results in Table 2 show the concentration of insoluble solids, soluble solids and total dust fall at the eight sampling locations. S1 had the highest concentration of total dust fall with 202.84 ± 65.64 mg/m²/day while S8 had the lowest concentration at 79.68 ± 70.64 mg/m²/day. The one-way ANOVA showed there were no significant differences ($p > 0.05$) between the concentration of insoluble solids, soluble solids and total at different sampling stations. Overall, the mean of total dust fall concentration in this study was found to be 131.50 ± 71.95 mg/m²/day, which is lower but close to the level recommended by the Malaysian Department of Environment (DOE, 2005); 133 mg/m²/day. The high concentration of total dust fall at S1 is most likely due to it being located near to a roadside which a high level of deposited particulates are emitted from moving vehicles. Conversely, the low concentration of total dust fall at S6 may be the result of a lower population density within a small residential area with a far lower volume of traffic on the nearby road. According to Kleeman and Cass (1998), Pandey *et al.* (1998), Pio *et al.* (1998) and Sami *et al.* (2006) high automobile exhaust emissions in urban areas are a principle contributor to high particulate pollution. Comparisons with other studies (Table 3), show that the concentration of total dust fall in this study is lower than that found by Hindy and Farag (1983), Kozak *et al.* (1993), Latif and Rozali (1999) for both Teluk Kalung and Air Keruh Area, Momani *et al.* (2000), Norela *et al.* (2004), and Norela *et al.* (2009) but higher than Latif and Rozali (2000) during haze episodes. Lower dust fall values shown, in this study can be mainly due to the fact that the other studies were undertaken near the industrial area and power station, which in turn is likely to have produced more particulate matter from the combustion of fossil fuels.

For soluble solids, S5 had the highest concentration compared to S6 which had the lowest concentration with a

Table 2. Mean concentration for insoluble, soluble and total solids (mg/m²/day) (n = 3).

Sampling station	Insoluble solids	Soluble solids	Total solids
S1	95.15 ± 52.81	107.69 ± 12.84	202.84 ± 65.64
S2	54.95 ± 43.53	50.47 ± 42.76	105.42 ± 86.29
S3	81.08 ± 37.24	104.82 ± 16.39	185.90 ± 53.63
S4	60.89 ± 35.69	73.62 ± 55.97	134.51 ± 91.66
S5	44.05 ± 47.03	134.21 ± 56.33	178.26 ± 103.36
S6	37.50 ± 18.96	33.05 ± 21.15	70.55 ± 40.11
S7	26.93 ± 12.82	67.92 ± 51.46	94.85 ± 64.28
S8	24.14 ± 29.04	55.54 ± 41.60	79.68 ± 70.64
Mean	53.08 ± 34.64	78.41 ± 37.31	131.50 ± 71.95

Note: Malaysian guidelines (133 mg/m²/day) for a period of 30 days.

Table 3. Summary of selected dust fall research results compared with current study (mg/m²/day).

References	Area	Insoluble solids	Soluble solids	Total solids
This study	Semi-urban area (Kajang and Bangi, Malaysia)	53.08 ± 34.64	78.41 ± 37.31	131.50 ± 71.95
Norela <i>et al.</i> , 2009	Residential Area (Nilai, Malaysia)	37.83 ± 13.53	216.11 ± 35.45	253.950 ± 7.54
Norela <i>et al.</i> , 2004	Power Station (Serdang, Malaysia)	213.86 ± 114.81	226.69 ± 171.50	440.54 ± 9.07
Latif and Rozali, 2000	Dust fall during haze Malaysia,	58.38 ± 34.04	59.07 ± 34.05	117.45 ± 54.79
Momani <i>et al.</i> , 2000	Urban Area (Amman, Jordan)	nd	nd	163.60
Latif and Rozali, 1999	Industrial Area (Air Keroh, Malaysia)	63.85 ± 42.02	74.45 ± 62.76	139.39 ± 87.88
Latif and Rozali, 1999	Industrial Area (Teluk Kalung, Malaysia)	69.84 ± 48.53	80.12 ± 64.63	149.96 ± 90.50
Kozak <i>et al.</i> , 1993	Rural and Recreational Area (Poland)	nd	nd	198.60
Hindy and Farag, 1983	Urban Area (Cairo, Egypt)	nd	nd	475.00

nd: no data

range between 33.10–134.21 mg/m²/day. The high concentration at S5 is most likely the result of road dust and emissions from moving vehicles. Moreover, S5 is located near to roadsides where a high level of dust is present and where vehicular emissions as well as open and biomass burning in and around the study area influence air quality. According to Arimoto *et al.* (2006), biomass burning, particularly in South East Asia, produces substances which contribute to the dust present in the atmosphere. The lower concentration of soluble solids at S6 was most probably a result of the low ground turbulence of wind speed which deposited a small volume of soluble solid into the dust fall collectors. Strong ground turbulence of wind speed can also contribute to the source of dust and fine particulates originating from exposed ground surface (Stout, 1998; Sami *et al.*, 2006; Pandey *et al.*, 2008). A study by Latif and Rozali (1999) in Teluk Kalung, Norela *et al.* (2004) and Norela *et al.* (2009) in Nilai and Serdang respectively recorded a higher concentration of soluble solids, while a study by Latif and Rozali (1999) in another sub-location in the Air Keroh industrial area and Latif and Rozali (2000) detected a lower concentration of soluble solids. Most of the studies used for the purposes of comparison were conducted at industrial and residential areas, such that it may be concluded that these areas contributed to the higher concentration of soluble solids in the samples. Meanwhile, S1 gave the highest concentration of insoluble solids with 95.15 ± 52.81 mg/m²/day and S8 gave the lowest concentration (24.14 ± 29.04 mg/m²/day). The reason for the high concentration of insoluble solids in S1 is predominantly due to its being located in a rapidly developing area with a high population and level of vehicle combustion compared to S8 (which has a low population in a residential area where there is a low level of traffic congestion). The concentration of insoluble solids in this study is significantly less than that reported by Norela *et al.* (2004) which was four times higher; it is also lower than the findings of Latif and Rozali (1999) and Latif and Rozali (2000) but higher than Norela *et al.* (2009). This might well be because the other studies were conducted in industrial areas where more dust was emitted into the atmosphere. Based on the results obtained, possible sources of dust fall in this study are: soil dust, industrial emissions, small particles from solid waste, plant residue along with the major source, which is most likely originates from vehicular fuel combustion; strongly correlating to the high traffic load. Moreover, soluble solids showed a higher mean concentration compared to insoluble solids, with a ratio of 1.48. It can be suggested therefore that wet deposition was the main process which brought down particles from the air. As particulates are inhalable they may as a consequence have significantly adverse effects on humans through entering the human respiratory system (Roorda-Knappe *et al.*, 1999; Polizzi *et al.*, 2007).

Anions and Cations in Rainwater

Volume-weighted mean values with volume-weighted standard deviation for anions and cations in rainwater are summarized in Table 4. The SO₄²⁻ showed the highest mean concentration values for anions, followed by NO₃⁻,

Table 4. Monthly volume-weighted mean (VWM) concentrations and volume-weighted mean standard deviation (VWSD) ($n = 3$) of anions and cations in rainwater ($\mu\text{eq/L}$).

Station	F^-	Cl^-	NO_3^-	SO_4^{2-}	K^+	Mg^{2+}	Ca^{2+}
S1	14.74 ± 17.37	30.74 ± 10.15	31.44 ± 7.42	319.38 ± 4.79	2.05 ± 0.26	5.76 ± 5.76	12.97 ± 5.99
S2	27.90 ± 0.00	25.39 ± 8.46	25.97 ± 6.77	92.86 ± 52.88	2.30 ± 1.02	4.94 ± 1.65	10.96 ± 13.47
S3	27.90 ± 0.00	20.60 ± 1.69	32.10 ± 19.52	146.37 ± 87.03	2.30 ± 1.02	4.93 ± 2.47	26.45 ± 9.99
S4	13.69 ± 19.48	32.44 ± 3.98	48.06 ± 13.22	135.33 ± 37.06	1.79 ± 0.26	8.23 ± 4.94	46.40 ± 34.93
S5	27.37 ± 0.00	23.13 ± 4.51	23.39 ± 10.00	229.44 ± 26.86	3.07 ± 2.81	4.11 ± 2.47	14.97 ± 5.49
S6	13.69 ± 19.48	19.46 ± 7.05	31.44 ± 8.71	70.79 ± 11.24	1.80 ± 0.00	5.76 ± 3.29	12.48 ± 6.99
S7	30.00 ± 3.16	51.05 ± 38.64	33.38 ± 4.84	109.72 ± 41.02	1.53 ± 0.51	4.94 ± 2.47	15.97 ± 5.00
S8	14.21 ± 18.95	20.87 ± 9.87	17.74 ± 19.35	64.96 ± 4.16	1.28 ± 0.26	3.29 ± 1.65	10.48 ± 3.50
Mean	25.27 ± 9.47	27.92 ± 24.2	30.32 ± 11.13	145.53 ± 33.10	2.05 ± 0.77	4.94 ± 3.29	19.97 ± 10.48

Cl^- and F^- . The highest SO_4^{2-} value was recorded at S1 with a concentration of $319.38 \pm 4.79 \mu\text{eq/L}$ and the lowest at S8 with a concentration of $64.96 \pm 4.16 \mu\text{eq/L}$. The highest NO_3^- concentration was recorded at S4 with a concentration of $48.06 \pm 13.22 \mu\text{eq/L}$ with the lowest recorded at S8. Moreover, there was no sign of any significant difference between F^- and Cl^- between stations. Statistical analysis was performed to see whether there were any substantial variations between stations in terms of the concentration of F^- , Cl^- , NO_3^- and SO_4^{2-} . The one-way ANOVA showed there were no significant differences ($p > 0.05$) between the ions. However, there were noticeable differences for SO_4^{2-} ($p < 0.05$) among the various sampling stations. This was most probably due to dissimilarities between locations, such that S1 and S5 were affected by vehicle emissions. In general, S1 showed the highest mean concentration of SO_4^{2-} which was most probably a direct result of the large number of heavy vehicles passing through the area as well as smoke from biomass and open burning other than the emission of industrial gases particularly sulfur dioxide. S8 gave the lowest concentration of SO_4^{2-} and a very low concentration of other anions, which was most likely because it is located in a less-developed area and is far from any industrial activity. Moreover, NO_3^- was found to have the second highest concentration of anions. This may be caused by biomass burning and vehicle exhaust emissions, especially at S4. According to Coelho *et al.* (2011), a high concentration of ions are associated with biomass burning, fertilizer use in agriculture and fossil fuel combustion. In general, SO_4^{2-} and NO_x gases from S and N compounds are predominantly released into the atmosphere as a consequence of fossil fuel combustion (Parashar *et al.*, 2001). Raciti *et al.* (2011) stated that NO_3^- was attributed to anthropogenic sources and urban development. Normally, the phenomenon of acid rain is due to the presence of high concentrations of SO_2 and NO_2 in the atmosphere (Al-Momani *et al.*, 1995; Wai *et al.*, 2005; Norela *et al.*, 2009; Song and Gao, 2009) which indicates that the occurrence of fossil fuel combustion was dominant over mobile sources (Satyanarayana *et al.*, 2011).

On the other hand, Ca^{2+} was observed as being the highest cation, followed by Mg^{2+} and K^+ respectively. These cations did not show any significant difference in concentration between the stations, ranging from 10.48 – $46.40 \mu\text{eq/L}$ for Ca^{2+} , 3.29 – $8.23 \mu\text{eq/L}$ for Mg^{2+} and 1.28 – $3.07 \mu\text{eq/L}$ for K^+ . Statistical analysis was performed to identify any significant variations but the one-way ANOVA results showed none ($p > 0.05$) for the values of Ca^{2+} , K^+ and Mg^{2+} among any of the sampling stations selected.

A high Ca^{2+} concentration was most likely be attributed to an increase in the quantity of windblown sand particles entering the atmosphere as occurring naturally or through human activity, whilst the concentration of Mg^{2+} was attributed to natural sources. According to Yusuf and Rashid (1995) and Banerjee (2008) a high Ca^{2+} concentration originates from carbonates generated from alkaline sources such as soil dust and earth crust while the Mg^{2+} element can be the result of sea spray particles which have been washed down by precipitation. Moreover, a low cation concentration (K^+ , Mg^{2+} and Ca^{2+}) compared to anion concentration may

be affected by heavy rainfall during the sampling period due to the effect of dilution and low sources of contaminants. Rainfall composition was found to be connected to atmosphere containing contaminants from local industries, traffic and agricultural sources (Tsai and Kuo, 2005; Hsieh *et al.*, 2009; Tsai *et al.*, 2011).

Calcium concentrations in this current study were lower when compared to the majority of those from previous studies, for example, Al-Momani (2000), Jain *et al.* (2000), Kulshrestha *et al.* (2003), Alkhashman (2005), Ozsoy *et al.* (2007), Banerjee (2008), Cao *et al.* (2009), Calvo *et al.* (2010), and Satyanarayana *et al.* (2011) which are listed in Table 5. The highest concentration of SO_4^{2-} was obtained by Parashar *et al.* (2001) during sampling which was performed in an urban area. On the other hand, the study undertaken by Al-Momani *et al.* (1995) and Santos *et al.* (2011) which was performed in industrial and urban areas, obtained the highest concentration of Cl^- as these areas were influenced by marine and crustal sources such as soil particles which were exposed to the atmosphere. According to the data collected and analyzed by Coelho *et al.* (2011) from an agriculture area, the highest concentration of ionic substances were NO_3^- followed by SO_4^{2-} and Ca^{2+} . Such results were obtained as a consequence of biomass burning, the use of fertilizer, along with animal waste and fossil fuel combustion in the sampling area, indicating that the NO_3^- concentration in this study can also be attributed to biomass burning activity. According to Kulshrestha *et al.* (1996) and Aherne and Farrell (2002), high temperature environment affected the ammonium concentration and conversion to NO_3^- but it cannot be proved in this study due to the lack of ammonium data. Fluoride ions are not usually determined in rainwater, most likely because of the low concentration and the extremely limited contribution of this ion to the atmosphere.

Trace Elements in Rainwater

The presence of heavy metals (Pb, Cd, Fe, Zn, Al and Cr) in the rainwater is shown in Table 6. Based on the data, the total concentration of heavy metals in order is $\text{Zn} > \text{Fe} > \text{Al} > \text{Pb} > \text{Cd}$ and Cr. Zinc showed the highest concentration compared to the other heavy metals with a mean concentration of $47.93 \pm 26.10 \mu\text{g/L}$ during the sampling period, followed by Fe, Al, Pb and Cd. Cr had the lowest concentration at $0.52 \pm 0.3 \mu\text{g/L}$. Zinc also showed the highest concentration in S3. In fact, it was 1.7 times higher than the mean concentration of all the sampling locations with the range $32.11\text{--}82.65 \mu\text{g/L}$. Moreover, S1 showed the highest concentration of Al with a concentration of $30.52 \pm 6.79 \mu\text{g/L}$, while S2 gave the highest concentration of Fe with a concentration of $44.18 \pm 7.67 \mu\text{g/L}$. Statistical analysis was performed to see whether there were any significant variations between stations regarding the concentration of Zn, Fe, Al, Pb, Cd and Cr. The results from the one-way ANOVA test showed there were no discernible dissimilarities ($p > 0.05$) in the values of Zn, Al, Cd and Cr. However, there was a significant difference ($p < 0.05$) between Pb and Fe values among the sampling stations, which was most likely due to differences in location and the differing exposure each one had to heavy metals.

Table 5. Summary of selected ionic research results compared with current study ($\mu\text{eq/L}$).

References	Site	F^-	Cl^-	NO_3^-	SO_4^{2-}	K^+	Mg^{2+}	Ca^{2+}
This study	Urban	25.27 ± 9.47	27.92 ± 24.21	30.32 ± 11.13	145.53 ± 33.10	2.05 ± 0.77	4.94 ± 3.29	19.96 ± 10.48
Coelho <i>et al.</i> , 2011	Agricultural	nd	2.80 ± 0.30	12.80 ± 1.40	3.50 ± 0.30	3.50 ± 0.40	1.80 ± 0.10	5.10 ± 0.40
Santos <i>et al.</i> , 2011	Urban	nd	145.00 ± 26.20	28.00 ± 3.40	23.50 ± 2.90	nd	nd	nd
Satyanarayana <i>et al.</i> , 2011	Rural	2.00 ± 5.00	52.00 ± 44.00	7.00 ± 6.00	20.00 ± 11.00	11.00 ± 8.00	13.00 ± 10.00	53.00 ± 52.00
Calvo <i>et al.</i> , 2010	Rural	nd	35.30 ± 33.90	25.20 ± 19.40	37.70 ± 22.70	3.90 ± 1.80	15.10 ± 11.10	42.10 ± 41.40
Cao <i>et al.</i> , 2009	Industrial	12.70 ± 1.60	86.80 ± 21.2	53.40 ± 8.10	163.30 ± 21.80	32.90 ± 14.50	17.00 ± 5.20	103.60 ± 14.30
Banerjee, 2008	Industrial	nd	63.07 ± 25.04	16.12 ± 3.71	44.22 ± 22.54	4.18 ± 1.71	36.72 ± 5.11	106.86 ± 25.26
Ozsoy <i>et al.</i> , 2007	Industrial	nd	65.50 ± 13.50	28.50 ± 2.10	75.90 ± 6.00	4.70 ± 0.40	30.90 ± 6.80	192.80 ± 18.20
Alkhashman, 2005	Industrial	nd	121.50 ± 29.94	63.70 ± 12.74	121.50 ± 29.66	51.10 ± 15.48	133.60 ± 17.71	192.10 ± 40.42
Kulshrestha <i>et al.</i> , 2003	Urban	nd	46.80 ± 30.80	10.30 ± 9.70	32.40 ± 20.90	35.90 ± 36.80	20.40 ± 3.70	119.20 ± 110.30
Parashar <i>et al.</i> , 2001	Urban	nd	2.20	5.90	27.70	10.20	0.80	7.50
Al-Momani <i>et al.</i> , 2000	Rural	nd	94.00	25.90	100.20	9.50	39.80	131.30
Jain <i>et al.</i> , 2000	Industrial	nd	18.00 ± 4.70	8.20 ± 1.80	21.80 ± 7.40	10.50 ± 3.50	15.20 ± 3.20	37.90 ± 7.10
Al-Momani <i>et al.</i> , 1995	Industrial	nd	117.00 ± 123.00	23.00 ± 32.00	66.00 ± 69.00	17.00 ± 23.00	101.00 ± 105.00	81.00 ± 86.00

nd: no data

Table 6. Mean concentrations of heavy metals in rainwater ($\mu\text{g/L}$) ($n = 3$).

Stations	Pb	Cd	Fe	Zn	Al	Cr
S1	5.05 ± 0.28	0.87 ± 0.54	18.07 ± 2.48	50.69 ± 38.47	30.52 ± 6.79	0.47 ± 0.26
S2	5.78 ± 1.49	1.62 ± 1.17	44.18 ± 7.67	50.74 ± 34.09	11.84 ± 6.15	0.56 ± 0.26
S3	6.08 ± 2.21	0.24 ± 0.12	9.80 ± 4.05	82.65 ± 6.88	15.54 ± 10.85	0.50 ± 0.41
S4	7.03 ± 2.31	1.26 ± 1.08	14.29 ± 5.88	46.01 ± 34.39	27.97 ± 26.47	0.45 ± 0.36
S5	2.04 ± 1.73	0.07 ± 0.06	15.30 ± 11.75	37.36 ± 25.71	23.48 ± 32.40	0.75 ± 0.14
S6	2.80 ± 0.75	1.44 ± 1.21	14.99 ± 10.08	51.24 ± 26.42	9.91 ± 8.62	0.54 ± 0.33
S7	1.03 ± 0.07	0.22 ± 0.19	17.73 ± 12.67	32.11 ± 27.13	3.91 ± 0.97	0.39 ± 0.33
S8	2.98 ± 0.77	0.41 ± 0.44	10.56 ± 10.26	32.69 ± 15.75	14.21 ± 18.64	0.56 ± 0.45
Mean	4.10 ± 1.20	0.76 ± 0.60	18.11 ± 8.10	47.93 ± 26.10	17.17 ± 13.86	0.52 ± 0.31

The highest concentration of Zn was found at S3, which is most likely because this station is located in an urban city area associated with vehicle combustion. According to Sternbeck *et al.* (2002) and Norela *et al.* (2005), Zn in the atmosphere were attributed to the use of car brakes and petrol fuel as used in vehicles such as motorcycles, cars and lorries. Aluminium showed the highest concentration in S1. This is most likely a direct consequence of the area itself which is rich in this mineral, along with the weathering process of rocks. Cd, Cr and Pb showed a lower concentration compared to other metals as fewer metal manufacturing industries were located near the sampling locations.

Heavy metals such as Cd and Pb are highly toxic and can remain in the environment longer periods of time (Joshi and Balasubramanian, 2010). Low concentrations of Pb in this study may be a consequence of the number of vehicles passing through the area near sampling stations. High levels of atmospheric lead are also frequently observed in industrial, residential and commercial areas such as for example, S4. These metals can be accumulated in soils from atmospheric deposition by sedimentation. The accumulation of heavy metals, including Pb, Zn, and Cu, on urban surfaces arises from vehicle exhaust emissions, industrial discharge, oil lubricants, automobile parts, the corrosion of building materials and atmospheric deposition (Lai *et al.*, 2007). According to Pouyat *et al.* (2007) and Raciti *et al.* (2011), urban areas can absorb a high concentration of heavy metals through dust fall. The concentration of Zn was lower compared when compared to study results from Vukmirovic *et al.* (2003), Ozsoy and Ornektekin (2009), Cheng and You (2010), Joshi and Balasubramanian (2010) and Cheng *et al.* (2011) but higher when compared to Alkhashman (2005) and Al-Momani (2003) as given in Table 7. Yet, when comparing the results obtained in the current study, the concentration of Pb, Cd and Cr are noticeably lower compared to Latif *et al.* (2001). This may be due to the fact that the location of this study was not affected by factories that can contribute certain metal concentrations in rainwater. Moreover, another study by Ozsoy and Ornektekin (2009) had higher concentrations of terrigenous particles (Fe, Al) and marine sources compared to Alkhashman (2005), who found a higher concentration of Pb to be caused by the metal's solubility and a high concentration of metals in rainwater. Furthermore, a study undertaken by Cheng and You (2010) found a much higher concentration of Pb,

about 420 times greater than that found in this study, which was affected by the Mindulle and Aere typhoon events in 2010 and 2011 respectively.

The statistical correlations matrix test indicated the suitability of rainwater quality data for factor analysis and showed that the correlation matrix was an identity matrix which would indicate the relationship between the ions present in rainwater (Sillapapiromsuk and Chantara, 2010). The results of the Pearson Correlation test for heavy metals, cations and anions were calculated and are listed in Table 8. The correlation between Mg^{2+} and Al ($R^2 = 0.544$, $p < 0.01$) and NO_3^- and Mg^{2+} ($R^2 = 0.953$, $p < 0.01$) was strongly correlated, suggesting that the sources of these elements originated from crustal elements. While there were also other correlations between NO_3^- and Ca^{2+} ($R^2 = 0.829$, $p < 0.05$), SO_4^{2-} and Al ($R^2 = 0.753$, $p < 0.05$), Cd and Pb ($R^2 = 0.472$, $p < 0.05$), Fe with Cd ($R^2 = 0.44$, $p < 0.05$) and Ca^{2+} with Mg^{2+} ($R^2 = 0.453$, $p < 0.05$). Correlation between Ca^{2+} and Mg^{2+} suggests the main source as being natural elements such as soil particulates. An abundance of heavy metals including Pb, Cd, and Fe in the atmosphere usually occurs as a result of vehicle exhaust emissions, industrial discharge, oil lubricants, automobile parts along with the corrosion of building materials (Lai *et al.*, 2007). Thus, it can be deduced that these metals most likely originate from the same sources, namely; industrial activity and vehicular emissions. Based on the correlation matrix of elements in rainwater from the study area, it can be shown that the area was influenced by anthropogenic sources such as vehicles, land clearing and factory emissions from the local area.

Source Apportionment of Anion, Cation and Heavy Metals in Rainwater

Fig. 2 showed the result of cluster analysis of anion, cation and heavy metals in rainwater that was group in 5 classes (C1–C5) consist of Class 1 (NO_3^- , F^- , K^+ , Mg^{2+} , Ca^{2+} , Pb, Cd and Cr), Class 2 (Cl^-), Class 3 (Fe), Class 4 (Zn) and Class 5 (Al). The result shows that the Cluster 1 elements contain the similarity of concentration which is in term of low concentrations that indicate least pollution from these element compared to other elements in other cluster that have significant concentration. Cluster 3 and 5 have strong similarities because they are from the same source which is from industries while the elements in cluster 1 also have related to this source. Other cluster shows less similarity that suggested wide sources contribution.

Table 7. Summary of selected trace element research results compared with current study ($\mu\text{g/L}$).

References	Site	Pb	Cd	Fe	Zn	Al	Cr
This study	Urban	4.10 \pm 1.20	0.76 \pm 0.60	18.11 \pm 8.10	47.93 \pm 26.10	17.17 \pm 13.86	0.52 \pm 0.31
Cheng <i>et al.</i> , 2011	Rural	9.60	0.02	15.30	65.20	34.70	0.04
Cheng and You, 2010	Urban	1710	nd	nd	64.2	13.8	0.4
Joshi and Balasubramanian, 2010	Industrial	90.25	4.57	3146	1127	769.30	213.80
Ozsoy and Ornekten, 2009	Urban	11.36 \pm 0.81	0.81 \pm 0.09	743.2 \pm 115	50.2 \pm 6.06	484.5 \pm 49.5	5.72 \pm 0.43
Alkhashman, 2005	Desert	51 \pm 36.40	42 \pm 22.86	21.50 \pm 32.84	32 \pm 32.61	nd	nd
Al-Momani, 2003	Rural	2.57 \pm 2.33	0.42 \pm 0.63	92 \pm 104	6.52 \pm 7.84	382 \pm 323	0.77 \pm 0.84
Vukmirovic <i>et al.</i> , 2003	Recreational	120.25	\leq 0.1	nd	75.9	nd	nd
Latif <i>et al.</i> , 2001	Industrial	6.17 \pm 4.47	4.49 \pm 4.57	nd	nd	nd	3.02 \pm 3.83

nd: no data

Table 8. Correlation matrix of different parameters of trace metals, cations and anion concentration in rainwater.

Parameters	Pb	Cd	Fe	Zn	Al	Cr	K ⁺	Mg ²⁺	Ca ²⁺	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
Pb	1												
Cd	.472*	1											
Fe	.134	.44*	1										
Zn	.317	.069	.165	1									
Al	.326	.172	.231	.298	1								
Cr	-.069	.010	-.171	-.170	-.096	1							
K ⁺	-.076	.241	-.124	-.042	-.124	.111	1						
Mg ²⁺	.306	.077	.113	.055	.544**	-.066	-.033	1					
Ca ²⁺	.146	.182	-.007	-.012	.058	.179	.053	.453*	1				
F ⁻	-.222	-.423	.348	.145	-.436	.144	.487	-.383	-.095	1			
Cl ⁻	-.284	-.184	.077	-.420	-.210	-.632	-.316	.241	.134	.301	1		
NO ₃ ²⁻	.494	.331	-.127	.237	.297	-.596	-.129	.953**	.829*	-.198	.405	1	
SO ₄ ²⁻	.130	-.248	-.109	.105	.753*	.138	.515	.124	-.052	-.014	.085	.089	1

*, Correlation is significant at the 0.05 level.

**, Correlation is significant at the 0.01 level.

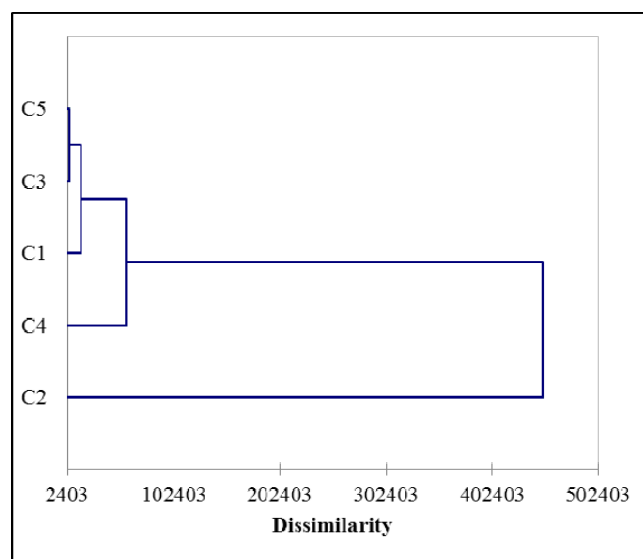


Fig. 2. Dendograms of cluster analysis for anion, cation and heavy metals in rainwater.

Principle Component Analysis and Multiple Linear Regression were done to identify the possible source of anion, cation and heavy metals in the rainwater (Fig. 3). This analysis separated four major sources that have high factor loading obtained from varimax rotated factor analysis (Kothai *et al.*, 2008). The obtained results revealed that sea spray was the main source of pollutant in the rainwater with contribution of 82.1% while soil contributes 15.1% followed by motor vehicle with 1.89% and earth crust with 0.94%. Sea spray was considered as the major source of pollutant due to the high value of factor loading of Cl^- and K^+ . Similar result was obtained by Santos *et al.* (2011) that also have major source of sea element in the rainwater. High factor loading of Cl^- is the indicator of sea salt influence on precipitation (Raman and Ramachandran, 2011) and the K^+ can be come from soil and open burning that can also mix with other element to produce high factor loading in PCA analysis. Sea spray might be the highest contributor because of the location of this study that affected by air mass transport and the wind trajectory that shows the influence

of wind direction from sea during southwest and inter monsoon between July to October (Fig. 4). While soil is the second highest sources with high factor loading for Mg^{2+} and Ca^{2+} which strongly indicate the crustal element as the source. High factor loading of F^- , NO_3^- , SO_4^{2-} , Zn and Cr were suggested the source of motor vehicle instead Pb, Cd and Fe that indicate the element of earth.

CONCLUSION

This study has shown that the concentrations of total solids in the deposited particulates were found to be under the permissible limit of $133 \text{ mg/m}^2/\text{day}$ as recommended by the Malaysian Department of Environment. In addition, based on the statistical analysis undertaken using the one-way ANOVA test, no significant correlations ($p > 0.05$) in the values of deposited particulates were found at the selected sampling stations. The dominant anion was found to be SO_4^{2-} followed by NO_3^- while the dominant cation was Ca^{2+} , followed by K^+ and Mg^{2+} ions. The dominant

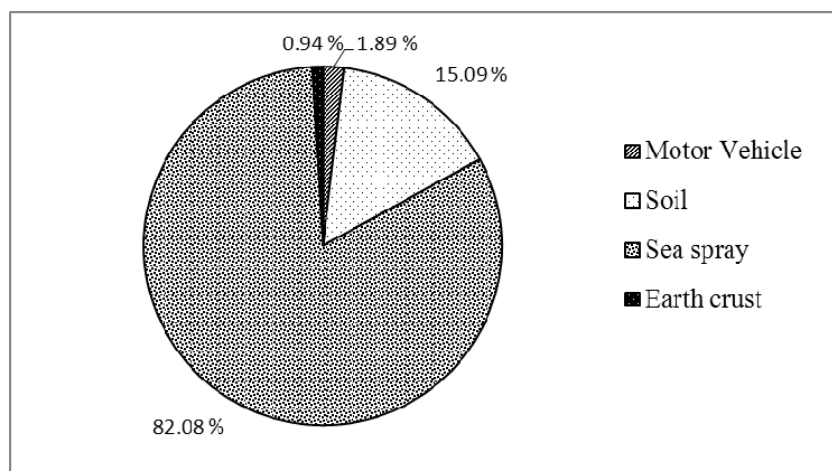


Fig. 3. Source apportionment of anions, cations and heavy metals in the rainwater.

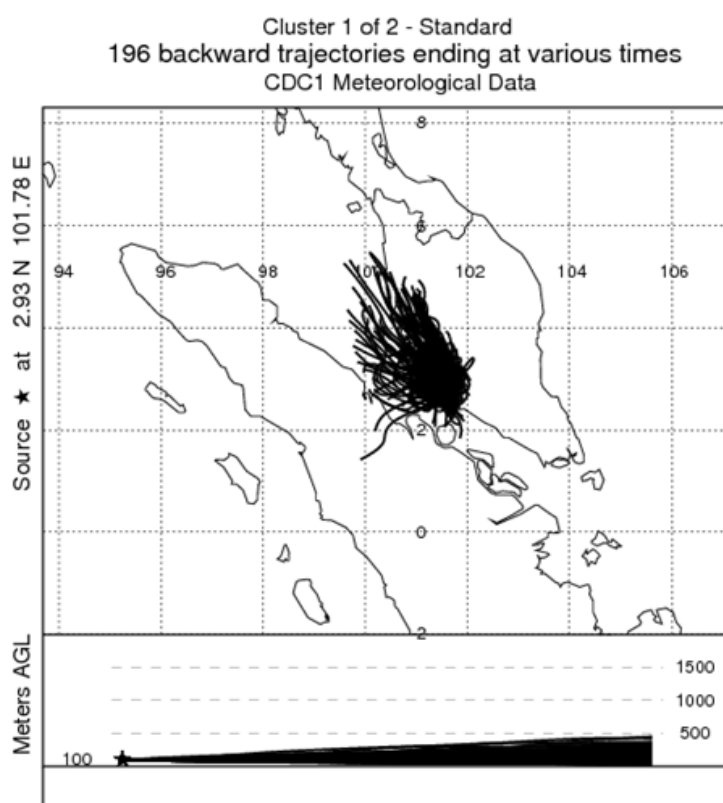


Fig. 4. Cluster of backward wind trajectories to the centre of the study area the during sampling time.

trace element was Zn followed by Fe, Al, Pb, Cd and Cr. From the results it can be concluded that the concentration of deposited particulates and contaminations in the study area most likely originated from sea salt, surfaces and crustal particle, as well as anthropogenic sources, particularly from motor vehicle emissions. Several precautionary procedures therefore need to be implemented by local authorities so as to maintain the concentration of dust fall around the areas studied. The quantity of vehicles, industrial activity along with bare land and biomass burning are shown to significantly increase the concentration of dust fall and its composition.

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