ANALYSIS OF PM_{10} IN KUALA TERENGGANU BY INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS

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Key words: airborne particulate matter, trace elements, PM_{10}, INAA, Kuala Terengganu

Abstract

Instrumental neutron activation analysis was used for the determination of trace elements in airborne particulate matter (PM_{10}) for air pollution monitoring. For the collection of air samples, the PM_{10} high volume sampler unit and Whatman 41 cellulose filter papers were employed. Samples were collected at 13 selected sampling sites covering areas in the city center, inner and outer city of Kuala Terengganu during the month of March 2005. The average PM10 was 69.64 μgm^{-3}, 83.58 μgm^{-3} and 72.22 μgm^{-3} for sampling stations located in the city center, inner and outer city of Kuala Terengganu, respectively. It was found that the mass of air particles in the study area was higher compared to Bangi and Kuala Lumpur. Chemical analysis of selected elements (Al, Fe, Cu, Pb, V, Mn, Zn, Cr, As Cd), ionic species (Na^+, SO_4^{2-}, Cl^-, NH_4^+,Mg^{2+}, K^+ , Ca^{2+}) and some rare earth elements (REE) were included in this study. In general, most of the average concentration of trace elements in the city center sampling stations was generally higher than the inner and outer city sampling stations. The concentrations of trace elements in sampling stations follow the general trend of Al>Fe>Zn>Cu>Mn>Pb>V>Cr>As>Ni>Cd. The elements concentration ranged from 680-2119 ngm^{-3}, 170-1132 ngm^{-3}, 8.13-122.4 ngm^{-3}, 8.48-77.3 ngm^{-3}, 7.68-14.4 ngm^{-3}, 1-90.4 ngm^{-3}, 1.47-3.25 ngm^{-3}, 1.43-5.03 ngm^{-3}, 1.15-4.45 ngm^{-3}, 0.24-3.75 ngm^{-3} and 0.28-1.36 ngm^{-3}, respectively.

Abstrak

Kajian kualiti udara telah dijalankan melalui penentuan paras kepekatan beberapa logam dalam habuk halus (PM10) dengan kaedah pengaktifan neutron (NAA). Pensampel isipadu tinggi PM10 yang dilengkapi kertas turas selulosa jenis Whatman 41 telah digunakan untuk menentukan kandungan zarahan terampai udara. Sebanyak 13 lokasi pensampelan telah di pilih merangkumi pusat bandar, luar pusat bandar dan pinggir bandar Kuala Terengganu. Purata kepekatan habuk halus (PM10) yang diperolehi adalah 69.64 $\mu g\ m^{-3}$, 83.58$\mu g\ m^{-3}$ dan 72.22 $\mu g\ m^{-3}$ pada ketiga-tiga lokasi pensampelan tersebut. Kepekatan habuk halus (PM10) yang dicatat di ketiga lokasi ini adalah lebih tinggi berbanding dengan Bandar Bangi dan Kuala Lumpur. Kandungan kepekatan logam (Al, Fe, Cu, Pb, V Mn, Zn, Cr, As Cd dan REE), cation dan anion (Na^+, SO_4^{2-}, Cl^-, NH_4^+,Mg^{2+}, K^+ , Ca^{2+}) dalam habuk halus (PM10) telah dikaji. Secara amnya, turutan logam dalam habuk halus (PM10) adalah seperti berikut Al>Fe> Zn>Cu>Mn >Pb>V>Cr>As>Ni>Cd dan julat kepekatan masing-masing adalah seperti berikut 680-2119 ng/m^3, 170-1132 ngm^{-3}, 8.13-122.4 ngm^{-3}, 8.48-77.3 ngm^{-3}, 7.68-14.4 ngm^{-3}, 1-90.4 ngm^{-3}, 1.47-3.25 ngm^{-3}, 1.43-5.03 ngm^{-3}, 1.15-4.45 ngm^{-3}, 0.24-3.75 ngm^{-3} dan 0.28-1.36 ngm^{-3}.

Introduction

The effects of atmospheric particulate matters on environment and human health have been of great global concern. Many epidemiological studies [1,2,3] have established an association between the particle concentration in the atmospheric and adverse effects on health; the PM10 fraction (diameter < 10 µm), and particularly the PM2.5 fraction (diameter < 2.5 µm) are known to be the primary cause of COPD (Chronic Obstructive Pulmonary Disease), asthma exacerbation, respiratory symptoms, morbidity and mortality, decrement in lung function and possible risk of lung cancer.

Atmospheric aerosol found in urban areas represent a mixture of primary particles emitted from various sources (e.g. vehicles exhausts, coal-fired power plants, oil refineries, forest fires, industrial emission, sea spear, volcano eruption etc) and secondary particles from aerosols formed by chemical reactions [4,5]. The morphology and composition of these particles may change through several processes, including vapor condensation, evaporation and coagulation. The final 'products' usually vary according to origin, chemical composition and physical properties, leading to particular deposition patterns in the human respiratory system. For this reason, intensive efforts to control pollution sources and to examine contamination levels through the analysis of various air pollution parameters are being followed up all around the region.

Mass concentrations of PM10 in the ambient air are usually less than 0.1 mgm^{-3} as and may contain elements at low concentration range (ngg^{-1}). In order to get good results, the methods of analyzing these elements need to be sensitive and precise, and also be able to identify multielements simultaneously because of the diversity of elements in the samples. Generally, these requirements could not be satisfied by conventional techniques (ICP-AES, and AAS) in a single chemical analysis. However, instrumental neutron activation analysis (INAA) using thermal neutrons in a nuclear reactor as an irradiation source, and high-resolution semiconductor detectors as measuring equipment, is one of the most suitable methods for satisfying the above mentioned requirements. INAA for airborne particulate matter can analyze up to μgg^{-1} ~ ngg^{-1} level of concentrations for 30~40 trace elements simultaneously [6,7,8].

Previous study conducted by Poh [9] on TSP levels in Kuala Terengganu has shown moderate particulate matter concentrations (17.2-148μgm^{-13}) in different areas. However, to the best of the authors' knowledge, there have been very little studies on the chemical characterization in PM10 fraction in Terengganu. The main objective of this study was to determine the distribution characteristics of trace elements in a rapidly urbanized area around Kuala Terengganu. Using appropriate statistical analysis such as correlation and enrichment factor analysis, attempt will be made to identify and apportion source(s) of these particulate matters in an effort to provide some baseline data on air quality for Kuala Terengganu for future references.

Experimental

Study Sites and Location

Kuala Terengganu is the capital of state Terengganu and located in the eastern part of Penisular Malaysia. The area of the capital is 605.28 km^2 with a population of 360,708 people, which accounts for about 35% of the total state population. Samples were taken during March 2005, aerosol samples were collected in 13 sampling stations representing town center (4 station), inner (5 station) and outer city (4 station). Figure 1 illustrated the map off sampling location. Particles with an aerodynamic diameter smaller than 10 µm (PM10) were collected on 8'x10' cellulose membrane filters exposed for 24 hours using PM10 high-volume samplers (Environmental Tisch, USA) at the average flow rate of 1.13 m^3min^{-1}. Filters were pre-weighed and then dried in a desiccator for at least 24 hours after being exposed to air.

PM10 and Elemental Concentration

The mass of particulate matter on filters were determined by gravimetric method using a microbalance to an accuracy of ±0.0001g. The chemical analysis was done for soluble ionic species such as Cl^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+} and metallic elements such as Al, Fe, Zn, Mn, As, Cr, Cd, Pb, Co, Sb, Th, Cs, Sm, Sc and Eu. For ionic species analysis, portion of filters (5cm in diameter) were placed in 25ml pp centrifuge tube and sonicate for 60 minutes in the ultrasonic bath. The sonic bath temperatures were maintained within 27°C. After removing from the sonic bath, the tubes were continuously shaken for 12 hours. The water extracted ionic species were determined using ion chromatography (DX120, Dionex). Most of the elemental concentrations in particles were determined by INAA. The filter samples were cut into a portion of 5cm in diameter then folded into clean polyethylene vials. Together with the standards, vessel and filter blanks were irradiated in MINT Triga reactor. Both short and long irradiation programs were used for the analysis. In short irradiation program, samples were irradiated individually using a pneumatic transfer system (Rabbit). Samples were counted for 5 minutes and subsequently for 30 minutes on PC based gamma spectrometer systems. For measurement of elements that produced long-lived radionuclides (6 hour irradiation) after 2 weeks of cooling period, the samples were also counted for 1 hour on gamma spectrometer systems. The elemental concentrations were calculated based on comparative method. Standard references material NIST SRM1648 Urban Particulate Matter and IAEA CRM SL-1 Lake Sediment were analysed flowing thesame procedures in every batch of irradiation for quality assurance and quality control purposes. In this work, Pb and Cd determination were unpractical using INAA. Since Cd and Pb are key element for sources apportionment, acid digestion couple with ICP-AES method suggested by Poh [9] was used .

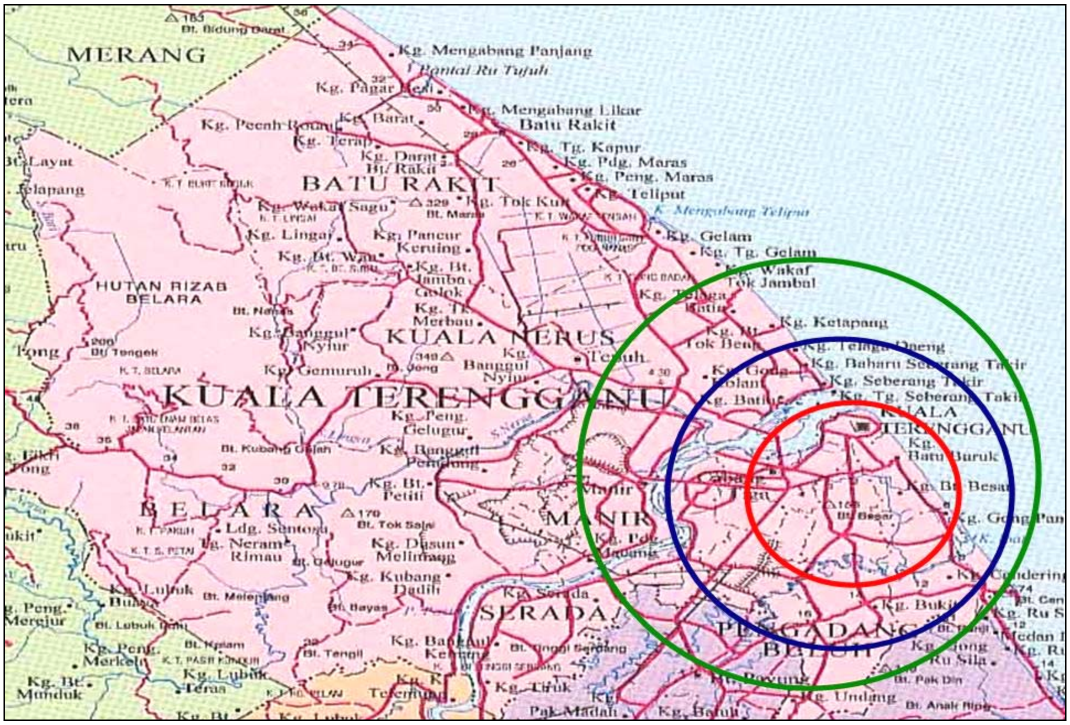

Figure 1. Map of Kuala Terengganu and sampling zones; (red-town center, blue-inner city, green-outer city).

Results and Discussion

Recoveries of the elements in SRM and CRM

Table 1 shows the results of the recovery study using NIST 1648 (Urban Particulate Matter) and IAEA CRM (SL-1 Lake Sediment). Briefly, most of the elements recoveries were within 80-120%. In the INAA analysis of PM10 samples, some elements like Fe, Zn, Mn, As, Cr, Co, La, Sb, Th, Cs, Sm, Sc and Eu were mostly detected in samples. However it was not easy to determine Na, K, Ti, V, Mg, Cl and Al in most of the samples as the sensitivities for these elements were not good. Especially the essential elements (Ca, Na, K, Mg, Cl), the accuracy of recoveries test were far to further from the recommended errors, 15%. For Al, Ti and V samples always lost count in gamma detector due to very short half-life radionuclides. Besides, some of the key elements like Pb and Cd were not determined because of unpractical determination using INAA or most of the recoveries results were too bad in many samples. In order to improve accuracies and uncertainties, cations (NH_4^+, K^+, Na^+, Mg^{2+} and Ca^{2+}), and anions (Cl^- and SO_4^{2-}) were determined by Ion Chromatography (DX 120) and INAA unpractical elements (Pb and Cd) and other poor recoveries elements was determined by more sensitive instruments, ICP-OES (Pb, Cd and others elements).

PM10 and Elementals concentration

Table 2 and Figure 2 illustrated average mass concentrations of PM10 in this study. Highest mean concentration was observed at inner city with value of 83.58 µgm^{-3} followed by outer city with mean value of 72.22 µgm^{-3}. Surprisingly town center poses the lowest concentration (69.64µgm^{-3}); the town center zone was associated with relatively high population and traffic density. It was found that the mass of air particles in the study area was higher compared to Bangi (63.6±26.4 µgm^{-3}) and Kuala Lumpur (55.4±30.3 µgm^{-3})[14].The high mass concentrations of inner city are caused by high value obtained from the fifth monitoring stations called *Losong.* During the sampling period, we managed to observe major soil excavation and building demolishing activities at about 3 km radius from *Losong* monitoring station. This contributed significantly to the increase in the mass concentration. The range of mass concentration in outer city (suburban residential areas) were relatively small compared to others. The free flow of air and totally no tall obstruction (buildings, trees and vehicles) around the monitoring stations make it an ideal criteria to collect air samples. All the PM10 values obtained were still well below the recommended Malaysia guideline for PM10 (mean of 24-hour measurement = 150 µgm^{-3}).

Table 3 presents the concentration of the measured elements and ionic species in PM10 in the three zones during to entire sampling period. The concentration levels between each zone are given in term of average mean and range of the value. In general, most of the average concentration of trace elements in the city center sampling stations was generally higher than the inner and outer city sampling stations. The concentrations of trace elements in sampling stations follow the general trend of Al>Fe> Zn>Cu>Mn>Pb>V>Cr>As>Ni>Cd. The elements concentration ranged from 680-2119 ngm^{-3}, 170-1132 ng m^{-3}, 8.13-122.4 ngm^{-3}, 8.48-77.3 ngm^{-3}, 7.68-14.4 ngm^{-3}, 1-90.4 ngm^{-3}, 1.47-3.25 ngm^{-3}, 1.43-5.03 ngm^{-3}, 1.15-4.45 ngm^{-3}, 0.24-3.75 ngm^{-3} and 0.28-1.36 ngm^{-3}, respectively. For outer city, the concentration of REE showed the trend of La>Sb>Th>Cs>Sm>Sc>Eu, whereas in inner city and town center followed the trends of Sb>La>Th>Cs>Sc>Sm>Eu.

In general, the average of the concentration of ionic species follows the trend of Na^+>SO_4^{2-}>Cl^-,>NH_4^+>Mg^{2+}> K^+ >Ca^{2+}. High loading of Na^+,Mg^{2+} and Cl^- species in samples were found in inner city zones. Whereas, higher concentration of NH_4^+, K^+ and SO_4^{2-} species were found in outer city aerosol samples. The average and range of species ionic concentration are given in Table 3. The high loadings of Na^+, Mg^{2+} and Cl^- in inner city were most probably generated from sea breeze. Correlation analysis indicated there was no clear correlation between these species in data combined from the three zones (Table 4) due to large variation in the analysis results for some ionic species in three different zones. However, the plot of Na against Mg^{2+} and Cl^- (Fig. 3) within inner city samples only show some trends of better correlation (Cl^-, r = 0.995; Mg^{2+}, r = 0.999) which might explain the role of sea breeze as major sources for Na^+, Mg^{2+} and Cl^- in inner city.

Table 1. Recoveries results of certified/standard references material (INAA method)

Element	SRM 1648			CRM SL 1		
	Certified Value	Obtained Value	Recovery (%)	Certified Value	Obtained Value	Recovery (%)
Al	34200	30882	90.3	89000	77990	87.63
Mn	786	694	88.4	3460	3699	106.93
Fe	39100	36722	93.92	67400	58253	86.43
As	115	110	95.76	27.5	28.7	104.47
Cr	403	349	86.68	104	120	115.56
Co	609	579	95.10	19.8	20.8	105.15
Zn	4760	3839	80.66	223	276.5	123.97
Eu	0.8	0.73	92	1.6	1.72	108
Sm	4.4	3.73	85.5	9.25	7.80	84.42
Th	7.4	6.75	91.31	14	15.02	107.32
Sc	7	6.22	88.91	17.3	19.03	110.21
La	42	39.44	93.92	52.6	60.67	115.36
Ce	55	53.43	97.16	117	120.40	102.91
Sb	45	40.62	90.28	1.31	1.45	110.76

Table 2**.** PM10 range and average within 3 different sections

Site	Average (range)
Town center	69.64 (58.37-83.06)
Inner city	83.58 (52.91-134)
Outer city	72.22 (70.02-73.25)

Element	Outer City (ng/m^3)	Inter City (ng/m^3)	Town Center (ng/m^3)
Na^+	18366 (5708-41262)	35094 (1063-64518)	15526 (2175-30417)
NH_4^+	5184 (1571-9393)	4461 (3863-5099)	3660 (2935-4378)
K^+	1881 (1418-2211)	1484 (592-2594)	1758 (944-3243)
Mg^+	2574 (727-4507)	2243 (216-7176)	1784(335-3461)
Ca^{2+}	1527 (903-2069)	1160 (441-2671)	1764 (731-2760)
Cl^-	5393 (2898-8678)	7717 (475-21453)	6298 (2213-10409)
SO_4^{2-}	10960 (5115-17065)	9477 (3055-15282)	9872 (6982-13555)
Al	780 (680-2119)	630 (510-1520)	1020 (950-1380)
Fe	430 (170-650)	660 (360-1040)	690 (570-1132)
As	1.09 (1.32-4.45)	1.53 (1.15-2.35)	1.70 (1.57-2.93)
Pb	3.973 (1.00-7.35)	25.34 (3.61-51.3)	55.45 (53.6-90.4)
Cd	0.329 (0.28-0.39)	0.563 (0.24-1.07)	0.843 (0.25-1.36)
Ni	1.003 (0.24-2.05)	0.959 (0.48-1.39)	1.715 (0.49-3.75)
V	2.362 (1.47-2.89)	2.391 (1.83-2.73)	2.376 (1.91-3.25)
Cu	30.67 (8.48-41.0)	39.72 (20.2-77.3)	43.85 (22.4-66.5)
Zn	16.14 (8.13-42.8)	59.34 (14.6-88.2)	72.36 (7.34-122.4)
Cr	1.30 (1.43-2.20)	1.32 (1.73-2.97)	2.65 (1.67-5.03)
Mn	8.29 (7.68-8.89)	9.89 (7.78-11.5)	11.2 (8.45-14.4)
Cs	0.29 (0.28-0.30)	0.23 (0.11-0.28)	0.19 (0.13-0.44)
Sb	1.41 (0.45-2.58)	1.79 (1.21-3.06)	1.96 (0.89-3.51)
Sc	0.14 (0.04-0.20)	0.18 (0.09-0.31)	0.19 (0.06-0.32)
La	2.04 (1.72-4.04)	1.67 (0.89-2.27)	1.56 (1.12-2.29)
Sm	0.21 (0.18-0.23)	0.14 (0.09-0.18)	0.14 (0.10-0.12)
Th	0.47 (0.37-0.57)	0.51 (0.20-0.77)	0.45 (0.23-0.85)
Eu	0.063 (0.055-0.086)	0.095 (0.071-0.147)	0.087 (0.089-0.153)

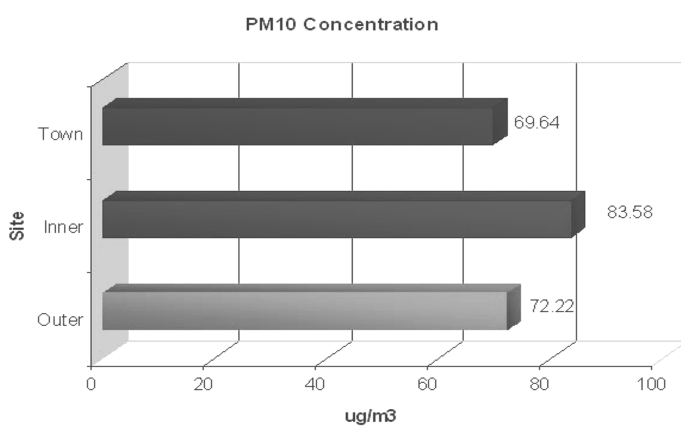

Figure 2. PM10 comparisons within 3 sections

Table 3. The range of concentration and mean value of elements and ionic species in PM10

Table 4. Correlation analysis of ionic species and element data combined from 3 sampling zones (n=13)

* Correlation is significant at the 0.05 level (2-tailed).

	Na	Cl	NH4	K	SO4	Al	Cd	Cu	Ni	Pb	Ti	V	Zn	As	Fe	Cr
Na	1	-0.227	-0.553	-0.217	-0.124	-0.064	0.076	0.229	-0.133	-0.064	0.076	0.229	-0.133	**0.685(*)**	0.173	-0.334
Cl	-0.227	1	0.130	0.276	0.325	0.074	0.061	0.478	0.134	0.074	0.061	0.478	0.134	0.264	-0.057	0.185
NH4	-0.553	0.130	1	-0.081	0.070	-0.311	-0.009	0.141	0.122	-0.311	-0.009	0.141	0.122	-0.084	0.080	0.191
K	-0.217	0.276	-0.081	1	**0.619(*)**	**0.855(**)**	0.441	0.379	**0.737(**)**	**0.855(**)**	0.441	0.379	**0.737(**)**	0.127	-0.191	-0.498
SO4	-0.124	0.325	0.070	**0.619(*)**	1	**0.696(*)**	0.565	**0.631(*)**	**0.568(*)**	**0.696(*)**	0.565	**0.631(*)**	**0.568(*)**	0.300	-0.351	-0.328
Al	-0.064	0.074	-0.311	**0.855(**)**	**0.696(*)**	1	0.327	0.424	**0.786(**)**	**0.795(**)**	0.327	0.424	**0.786(**)**	0.597	-0.030	-0.752
Cd	0.076	0.061	-0.009	0.441	0.565	0.327	1	0.393	0.540	0.327	**0.789(**)**	0.393	0.540	0.190	-0.428	-0.347
Cu	0.229	0.478	0.141	0.379	**0.631(*)**	0.424	0.393	1	0.194	0.424	0.393	**0.847(**)**	0.194	**0.758(*)**	-0.260	-0.507
Ni	-0.133	0.134	0.122	**0.737(**)**	**0.568(*)**	**0.786(**)**	0.540	0.194	1	**0.786(**)**	0.540	0.194	**0.687(**)**	0.044	-0.032	-0.336
Pb	-0.064	0.074	-0.311	**0.855(**)**	**0.696(*)**	**0.795(**)**	0.327	0.424	**0.786(**)**	1	0.327	0.424	**0.786(**)**	0.597	-0.030	-0.752
Ti	0.076	0.061	-0.009	0.441	0.565	0.327	**0.789(**)**	0.393	0.540	0.327	1	0.393	0.540	0.190	-0.428	-0.347
V	0.229	0.478	0.141	0.379	**0.631(*)**	0.424	0.393	**0.847(**)**	0.194	0.424	0.393	1	0.194	**0.758(*)**	-0.260	-0.507
Zn	-0.133	0.134	0.122	**0.737(**)**	**0.568(*)**	**0.786(**)**	0.540	0.194	**0.687(**)**	**0.786(**)**	0.540	0.194	1	0.044	-0.032	-0.336
As	**0.685(*)**	0.264	-0.084	0.127	0.300	0.597	0.190	**0.758(*)**	0.044	0.597	0.190	0.758(*)	0.044	1	0.418	-0.227
Fe	0.173	-0.057	0.080	-0.191	-0.351	-0.030	-0.428	-0.260	-0.032	-0.030	-0.428	-0.260	-0.032	0.418	1	0.587
Cr	-0.334	0.185	0.191	-0.498	-0.328	-0.752	-0.347	-0.507	-0.336	-0.752	-0.347	-0.507	-0.336	-0.227	0.587	1

** Correlation is significant at the 0.01 level (2-tailed).

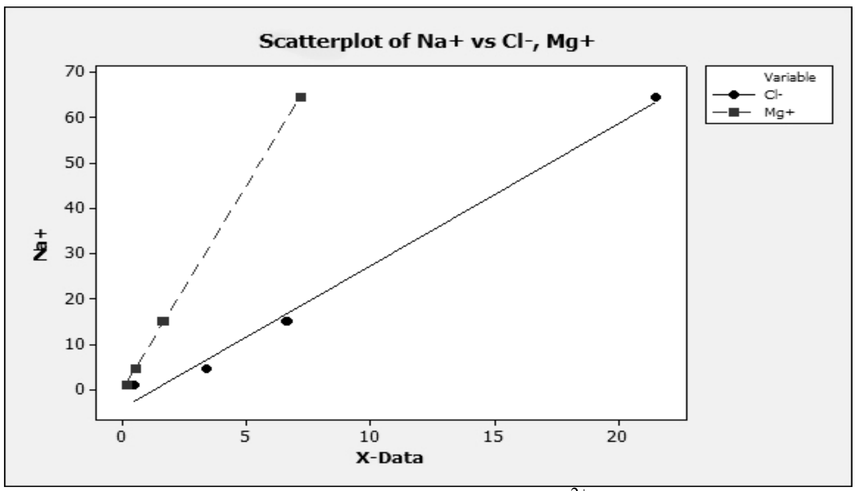

Figure 3. The plot of Na against Mg^{2+} and Cl^-

Enrichment Factor

The enrichment factor method has commonly been used as a tool to evaluate the strength of enrichment or depletion, relative to specific sources. The enrichment factor for any element X relative to control/unpolluted references material is defined by

$$EF_X = \frac{(X/Y)_{air}}{(X/Y)_{control}}$$

Where $EF_{,X}$ is the enrichment factor of X, Y is a reference element for crustal material and $(X/Y)_{air}$ is the concentration ratio of X to Y in the samples and $(X/Y)_{control}$ is the same ratio in control/unpolluted references material. If EF_X approached unity, crustal soils are likely the predominant sources for element X[10]. In this study, the $(X/Y)_{control}$, ratio calculation for each elements was based on samples collected from a rural area call *Manir* (one of the sampling station in outer city). Al is normally use as references element since it can be accurately measured by a number of analytical method and abundance in the earth crusts [11].

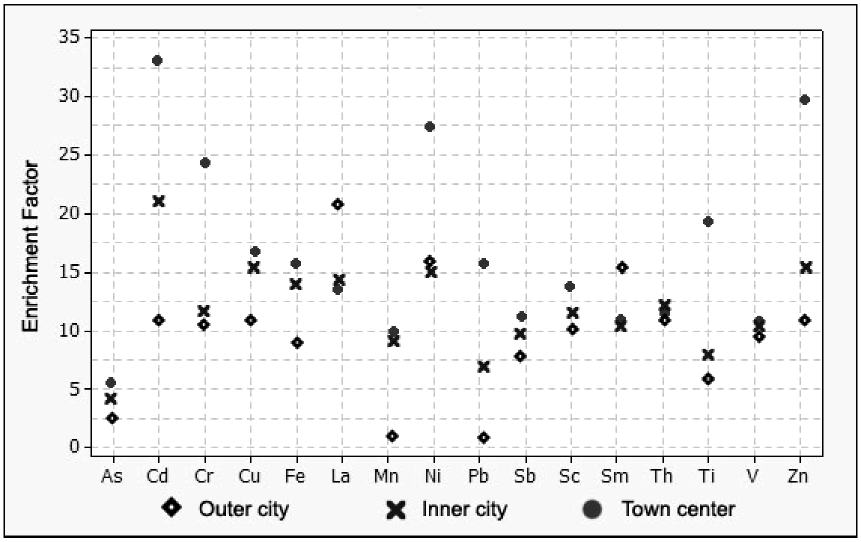

Figure 4. Enrichment factors of selected trace elements relatively to Al for the PM10 samples at three locations.

Figure 4 indicates that the enrichment factors of all elements except La and Sm in town center are higher than those in inner and outer city. The rapid urbanization activities in Kuala Terengganu explains for the high elements loading to ambient atmospheric. The enrichment factor for Cd, Cr, Ni, Ti, Pb and Zn in town center exhibited at least 10 times higher than other places. The strong correlation (Table 4) between Pb, Zn, Ni and SO_4^{2-} ($r \geq 0.696$) explained these elements may originate from traffic emission sources. Cadmium, Pb, Cr, Ti and Zn probably emitted on atmospheric as part of brake dust, road paint, diesel exhaust particles, road construction materials, or car catalyst materials [12]. Whereas Ni, which are frequently associated with fuel burning, can be easily attributed to emissions from the petrochemical combustion such as vehicle gasoline. According to Chiarenzelli *et al.*[13] rare earth elements may be useful as reference elements in environmental studies due to its transport in the particulate phase, lack of significant anthropogenic sources, coherent group geochemistry, generally robust concentrations, and upper crustal signatures. For REE except La, most of the enrichment factors were below 15 and close to each study zones. This indicates the REE may appear to be of soils origin.

Conclusion

Highest mean PM10 concentration was observed in inner city with value of 83.58 μgm^{-3} followed by outer city with mean value of 72.22 μgm^{-3} and town center, 69.64μgm^{-3}. The Al, Fe, Cu, Zn, Mn, Pb, V, Cr, As, Ni, Cd, Cs, Sb, Sc, La, Sm, Th and Eu concentration ranged from 680-2119 ngm^{-3}, 170-1132 ngm^{-3}, 8.13-122.4 ngm^{-3}, 7.68-14.4 ngm^{-3}, 1-90.4 ngm^{-3}, 1.47-3.25 ngm^{-3}, 1.43-5.03 ngm^{-3}, 1.15-4.45 ngm^{-3}, 0.24-3.75 ngm^{-3} and 0.28-1.36 ngm^{-3}, 0.11-0.44 ngm^{-3}, 0.0.45-3.51 ngm^{-3}, 0.04-0.32 ngm^{-3}, 0.89-4.40 ngm^{-3}, 0.09-0.23 ngm^{-3}, 0.2-0.85 ngm^{-3} and 0.05-0.153 ngm^{-3}, respectively. For ionic species, all the three locations follow a similar trends of $Na^+ > SO_4^{2-} > Cl^-, > NH_4^+ > Mg^{2+} > K^+ > Ca^{2+}$. The high loadings of Na^+, Mg^{2+} and Cl^- in inner city were most probably generated from sea breeze. Finally, enrichment factors for Cd, Cr, Ni, Ti, Pb and Zn indicate that these elements may origin from traffic emission sources and REE may appear to be of soils origin.

Acknowledgment

The authors wish to acknowledge the National Nuclear Agency for the financial support to conduct this research.

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