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Source apportionment and health risk assessment of PM_{10} in a naturally ventilated school in a tropical environment

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ABSTRACT

This study aimed to investigate the chemical composition and potential sources of PM_{10} as well as assess the potential health hazards it posed to school children. PM_{10} samples were taken from classrooms at a school in Kuala Lumpur's city centre (S1) and one in the suburban city of Putrajaya (S2) over a period of eight hours using a low volume sampler (LVS). The composition of the major ions and trace metals in PM_{10} were then analysed using ion chromatography (IC) and inductively coupled plasma-mass spectrometry (ICP-MS), respectively. The results showed that the average PM_{10} concentration inside the classroom at the city centre school (82 g/m^3) was higher than that from the suburban school (77 g/m^3). Principal component analysis-absolute principal component scores (PCA-APCS) revealed that road dust was the major source of indoor PM_{10} at both school in the city centre (36%) and the suburban location (55%). The total hazard quotient (HQ) calculated, based on the formula suggested by the United States Environmental Protection Agency (USEPA), was found to be slightly higher than the acceptable level of 1, indicating that inhalation exposure to particle-bound non-carcinogenic metals of PM_{10}, particularly Cr exposure by children and adults occupying the school environment, was far from negligible.

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1. Introduction

Children are far more susceptible to the effects of air pollutants than adults as they breathe in more air per unit of body weight at a given level of exertion and are also less capable of dealing with the effects of toxic chemicals. Studies have shown that there is a direct correlation between exposure to air pollutants and the concentration performance (CP) of students in the school environment (Mohai et al., 2011; Twardella et al., 2012). In addition, indirect indicators, such as absenteeism from school, provide evidence of the impact of air pollutants on school children's health (Mendell and Heath, 2005; Peter et al., 1997). As a lot of children these days spend much of their time in an indoor environment, indoor air quality (IAQ) has consequently been recognised as one of the key influences on their health. The classroom is one of the main places where children spend their time when not in the home environment (Chatzidiakou et al., 2012; Raysoni et al., 2013). Poor air quality inside classrooms has been found to have connections to several common childhood diseases, such as asthma, rhinitis and rhinoconjunctivitis (Cui et al., 2013; Fsadni and Montefort, 2013).

The pollutants within a classroom have been found to predominantly originate from an infiltration of outdoor sources, such as: soil dust, traffic and industrial emissions (Diapouli et al., 2008; Krugly et al., 2014; Reche et al., 2015). Many studies have demonstrated that the level of air pollutants, such as particulate matter (PM), carbon dioxide and volatile organic compounds in classrooms is directly influenced by increasing levels of air pollutants outside school buildings (Canha et al., 2014; Daisey et al., 2003; De Gennaro et al., 2013; Hassanvand et al., 2014; Razali et al., 2015). This is in addition to the pollutants which originate from the classroom environment itself, such as those from furniture, paint, chalk, cleaning agents and the re-suspension of soil dust (Dorizas et al., 2015; Mejía et al., 2011). Other factors, such as the activities students are involved in, the level of occupancy, ventilation systems and temperature have also been found to affect a classroom's air quality (Raysoni et al., 2013). As for organic matter, skin flakes and cotton fibres from clothes are noted as two of the most important sources found in the indoor environment, in this case, school classrooms (Braniš and Šafránek, 2011; Viana et al., 2011). The composition of particulate matter and dust in the

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indoor environment is crucial when it comes to determining their sources and toxicity.

Receptor models, such as chemical mass balance (CMB) (Watson et al., 2002), positive matrix factorisation (PMF) (Paatero, 1997; Pekey et al., 2013) and principal component analysis (PCA) (Thurston and Spengler, 1985) have previously been applied in order to identify and quantify aerosol sources, including those found in the indoor environment. However, for PCA, the results are only applicable for source identification and cannot be used directly for source apportionment. As a result, for the sake of this study it was deemed necessary to use a combined source apportionment technique, principal component analysis-absolute principal component scores (PCA-APCS) (Habil et al., 2013; Khan et al., 2010; Song et al., 2006; Thurston et al., 2011). This method involved obtaining and analysing PM samples for major components and source markers. The main advantage of using PCA-APCS is that it is capable of identifying different sources and their composition features without any prior knowledge of the sources. The results of PCA-APCS analysis from a previous study concluded that the burning of fossil fuels, geological sources and biomass combustion are among the main contributors to particulate matter pertaining to the school environment (John et al., 2007).

With respect to health, metal compounds associated with PM have been found to affect the respiratory system, which is the principal route of entry for air pollutants (Huang et al., 2014). Although present in a small portion of PM mass, toxic trace metals have been found to have a significant impact on the respiratory health of children and other susceptible individuals, such as those with low lung function, asthma and bronchitis (WHO, 2003). In order to examine the potential risks resulting from human exposure to toxic contaminants, a health risk assessment has previously been undertaken worldwide (USEPA, 2006). In addition, numerous studies have shown that particulate matter contaminated with heavy metals and other pollutants, such as PAHs,

can directly enter the body through inhalation, dermal contact and oral ingestion exposure pathways (Khairy and Lohmann, 2013; Sun et al., 2015; Wei et al., 2015). The study by Lu et al. (2014) in nursery schools demonstrated that both non-cancer and cancer risks of the studied metals were within the safe range, HQ < 1 and within the range of 10^{-6}–10^{-4}, respectively, as recommended by the US Environmental Protection Agency (USEPA, 2000).

The aim of this study was to assess the concentration of PM_{10} in two classrooms at two Malaysian primary schools which were in different settings: one, which was located in Kuala Lumpur's city centre and the other in the suburb of Putrajaya. The concentration of PM_{10} and its composition in the two classrooms was then compared with levels outdoors. To determine the origins of the indoor PM_{10} composition in the schools, the enrichment factor (EF) and principal component analysis-absolute principal component scores (PCA-APCS) were applied to the datasets of the PM_{10} samples collected. The estimated health risk posed by the selected metals was also measured using the USEPA health risk assessment system to assess the impact on the children using those classrooms.

2. Materials and methods

2.1. Description of sampling sites

The locations of the sampling sites are shown in Fig. 1. The two public primary schools were selected according to their locations: one in the city centre (Kuala Lumpur) and the other in a new suburban area (Putrajaya). These two sites were chosen so as to obtain a general view of particulate matter concentrations, its composition as well as the effect of its exposure on school children attending schools in different settings. School 1, Jalan Raja Muda Primary School (S1), located in Kuala Lumpur's city centre, sits at
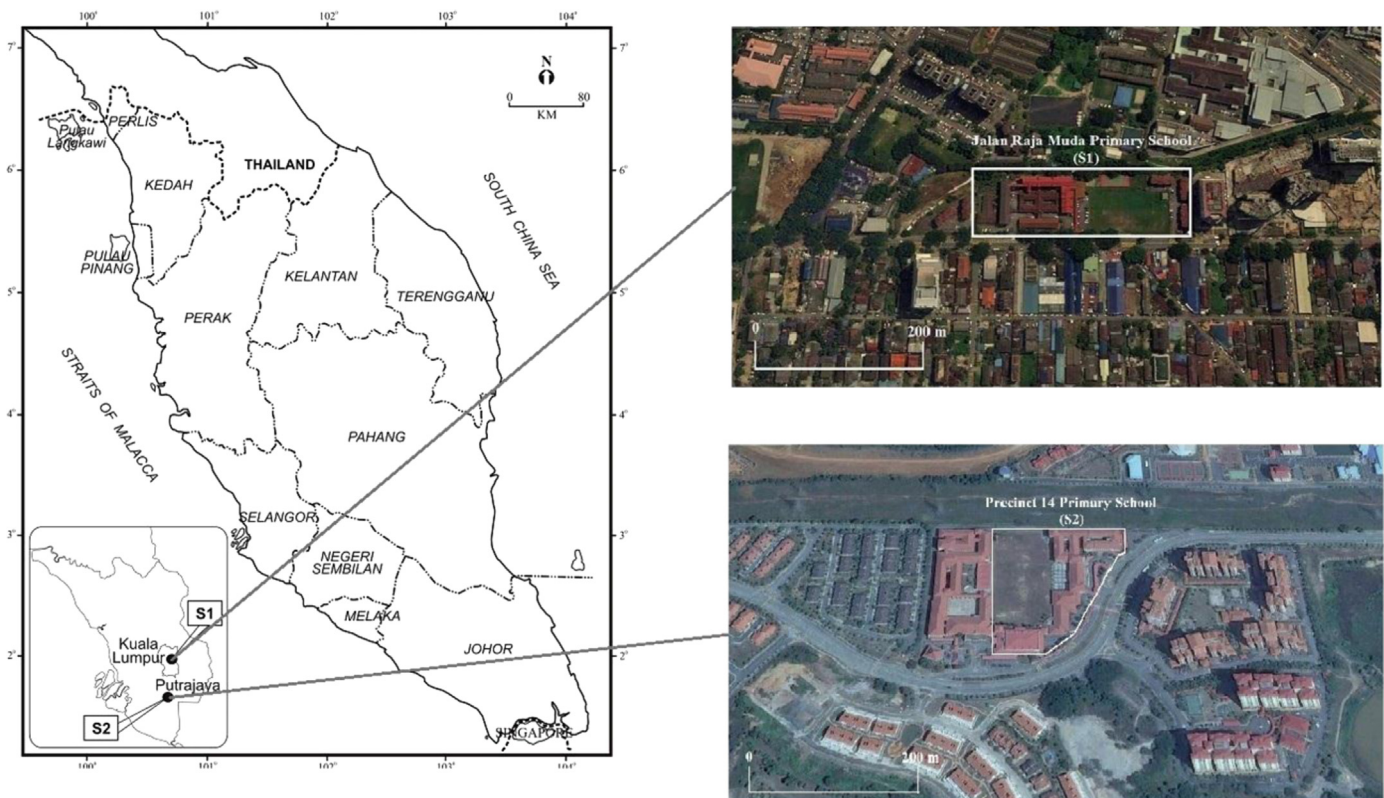

Fig. 1. Location of the two primary schools.

the side of one of the busiest roads in the area so is impacted by a high traffic density. It is also in a densely-populated area which is surrounded by tall commercial buildings and some small-scale industries, including paint and automotive. Furthermore, there is some ongoing construction occurring nearby. The classroom selected for the study was on the ground floor and used by a class of 30 eight-year-old students. It was equipped with standard school tables and chairs, with a blackboard and a small whiteboard at the front of the room and wooden bookshelves at the back of the classroom. The floor of the classroom was made of cement and the windows were timber louver windows.

School 2, Putrajaya Precinct 14 Primary School (S2) is located in a residential area in Putrajaya and is adjacent to a major road. There has been a substantial growth in the number of houses in this city, particularly in recent years and at that the time of sampling there was also some ongoing construction projects taking place near the sampling site. The classroom selected from this school was on the first floor and was used by 33 seven-year-old students. It was equipped with standard school tables and chairs, with a whiteboard at the front of the room, an overhead LCD projector and metal bookshelves at the side of the classroom. The floor in the classroom was tiled and the windows were single-glazed with aluminium window frames. The building materials generally consisted of bricks and concrete. Both of the classrooms used for the study were equipped with a fan and were often ventilated by keeping doors and windows open. They were both occupied from Monday to Friday, except during school or public holidays. Details of each of the schools' characteristics are presented in Table S1.

2.2. Sampling protocol

A total of 88 indoor and outdoor PM_{10} samples were collected using a low volume sampler (LVS) (Airmetrics MiniVol, USA). The LVS operated at a flow rate of 5 L/min on an 8 h cycle (from 7:30 a. m. to 3:30 p.m.). Averaged 8 h sampling was conducted on three consecutive weekdays at each sampling sites between January and November 2012. PM_{10} measurements for each school were attained separately due to the limited availability of sampling equipment. For indoor sampling, the LVS was positioned at the back of the classroom (at least one metre from the wall) out of concerns for child safety. The location of sampling sites is shown in Fig. S1. The inlet of the indoor sampler was positioned about one metre above floor level so as to simulate the location of the breathing zone of the students. Meanwhile, for the outdoor sampling at S1, the sampler was positioned approximately three metres away from the classroom but within five metres of the major road which was in close proximity. For the outdoor sampling at S2, the sampler was placed in the playground beside the school field (about three metres from the classroom) and within 50 m from the major roadway situated nearby. Once again, the position of sampler inlet was at least one metre above the ground.

PM_{10} samples were collected on 47 mm (in diameter) Teflon coated glass fibre membrane filters (0.47 m pore size, PallFlex, USA) which were used to determine the total mass, water soluble ionic species (WSIS) and trace metals in the samples. PM_{10} mass concentrations were obtained through the gravimetric method using a 5-digit electronic microbalance (GR202, AND, Japan) with $\pm$ 0.001 mg uncertainty. Prior to weighing, the filters were equilibrated for 48 h inside desiccator in order to condition them. They were then transported to and from the field in clean resealable plastic bags. After sampling, the samples were conditioned, weighed and stored in a desiccator until they underwent chemical analysis.

2.3. Chemical analysis

2.3.1. Water-soluble ionic species analysis

PM_{10} ionic analysis was performed using ion chromatography (IC; 881 Compact IC Pro, Metrohm, Switzerland) for sulfate (SO_4^{2-}) and nitrates (NO_3^-). The concentrations of ammonium (NH_4^+), sodium (Na^+), potassium (K^+), calcium (Ca^{2+}) and magnesium (Mg^{2+}) were determined using IC (850 Professional IC, Metrohm, Switzerland). The ionic species were extracted from half of each filter paper in 20 ml ultra pure water (UPW; 18.2 MΩ cm^{-1} resistivity, Easypure II, Thermo Scientific, Canada) using an ultrasonic bath (Elmasonic S150, UK) for 1 h at 30 °C and then centrifuged (Kubota 5100, Japan) at 1500 rpm for 1 h to leach the chemical composition into the UPW. After which the samples were filtered using a Luer lock plastic syringe attached to a syringe filter (22 mm in diameter, 0.2 m pore size, Pall, USA) to remove any non-soluble species. Calibration curves for all ions were obtained with five points (0.5, 1, 2, 5 and 10 ppm) using certified standard solutions (CertiPUR® Merck, Germany).

2.3.2. Trace metals analysis

One fourth of each filter paper was used to determine which trace metals were present and underwent a closed-vessel microwave-assisted acid digestion procedure. These filter papers were digested in high-pressure Teflon bombs containing a mixture of 8 ml of 65% nitric acid (HNO$_3$; Friendemann Schmidt, Australia) and 2 ml of 30% hydrogen peroxide (H$_2$O$_2$; Calbiochem, Germany) and then placed in the microwave digestion system (Milestone, MLS 1200 Mega, Italy). The microwave acid digestion process consisted of four steps. The first step involved the use of an operating power of 250 W for 1 min which was then held for 1 min; for the second step, the power kept at 250 W and held it for 8 min; in the third, it was raise to 400 W in 5 min, and the final step, it was increased to 650 W and then held it for 5 min. After a cooling process of approximately 30 min, the digested solutions were filtered using a 20 ml Terumo syringe attached to a syringe filter (22 mm in diameter, 0.2 m pore size, Pall, USA) and diluted with UPW to the final volume of 50 ml. Then, the solutions were transferred to a high density polyethylene (HDPE) bottle and stored in a refrigerator at 4 °C while awaiting further analysis. The trace metals found (Al, Fe, Zn, Pb, As, Cd, Co, Cu, Cr, V and Ni) were analysed using inductively coupled plasma-mass spectrometry (ICP-MS; ELAN 9000, Perkin Elmer, USA). The calibration was performed using multi-element calibration standard 3 (CertiPUR® PerkinElmer, USA) at concentrations of 10, 20, 30 and 50 ppb.

2.4. Quality Assurance/Quality Control (QA/QC)

Quality control was practised throughout the analysis in order to limit the likelihood of error and avoid any interference. Detailed descriptions of the QA/QC adhered to can be found in our previous studies (Han et al., 2014; Zhong et al., 2014). The limit of detection (LOD) was estimated as being three times the standard deviation of the blank samples. This estimate was made in order to determine the lowest concentration level that could be detected by the filter. The recovery efficiencies for WSIS were calculated by spiking samples with a known amount of ion standard. The recoveries obtained were noted as being in the range of 98–118%, and 84–118%, respectively. In order to ensure the accuracy and validity of the results obtained by the methods used in the trace metal analysis, two standard reference materials, SRM 2584 (indoor dust) and SRM 1648 (urban particulate matter) were used. The percentage of recovery of the trace metals for SRM 2584 and SRM 1648 from this procedure ranged between 67% and 121%, and between 54% and 92%, respectively.

2.5. Receptor modelling

2.5.1. Enrichment factor analysis

The enrichment factor (EF) was calculated so as to determine whether indoor PM_{10} originated from crustal or non-crustal sources. The concept of EF analysis is simple and the calculation is detailed in our previous studies (Han et al., 2014; Latif et al., 2014). In this calculation, the sample metal concentrations were normalised using upper continental crust (UCC) data (Taylor and McLennan, 1985). The metals most commonly used as reference elements were Al, Fe, and Mn (Duan et al., 2012; Yongming et al., 2006). In the present study, Fe was used as the reference element, based on the assumption that it has a relatively high natural concentration.

2.5.2. Principal component analysis

Principal component analysis (PCA), a multivariate analysis technique, was selected as the means of identifying the possible sources of 18 elements in indoor PM_{10} through the use of the IBM Statistical Package for the Social Sciences (SPSS Version 21.0, USA). PCA was performed to reduce the data and extract a small number of factors (principal components, PCs) which would explain the variance observed in a larger number of variables in a more effective and efficient way (Theodosi et al., 2010; Yongming et al., 2006). Prior to PCA, quality control (QC) measures were applied, whereby data below the detection limit was replaced by half of the detection limit (Fromme et al., 2008). After which the data was normally distributed.

PCA can be explained mathematically using Thurston and Spengler's formula (Thurston and Spengler, 1985):

$$Z_{ij} = \sum_{k=1}^{p} g_{ik} h_{kj} \tag{1}$$

where Z_{ij} is the elemental concentration of jth in the ith sample at the receptor site, $k = 1, 2, …, p$ sources, and g_{ik} and h_{jk} are the factor loadings and factor scores, respectively. This technique was performed on the datasets, which were composed of a selection of 18 major ions and trace elements concentrations from the classroom of each school. A varimax rotation of the normalised factor loadings was performed in order to maximise the divergence of the variables of the squared normalised factor loadings. After extraction, only factors with eigenvalues greater than one were considered to be the principal component based on Kaiser's criterion (Kaiser, 1960). This step was applied to gain a better understanding and interpretation of the data. The communality values for all the elements were found to exceed 0.70, indicating that the component solutions were quite satisfactory. The identification of each factor as a source, was based on the interpretation of the correlations, such that absolute loading values greater than 0.75 were categorised as "strong loading", between 0.50 and 0.75 as "moderate loading", and between 0.30 and 0.50 as "weak loading".

The absolute principal component scores (APCS) method was applied in order to quantify the contributions of each source to specific measured pollutants (Guo et al., 2004; Khan et al., 2010). APCS have been found to be useful in providing meaningful physical factors for interpretation (Chan and Mozurkewich, 2007) and were calculated through the factor score matrix, which can be explained mathematically following Thurston and Spengler's formula (Thurston and Spengler, 1985):

$$C_i = (b_0)_i + \sum APCS_p \times b_{pi} \tag{2}$$

where C_i is the mass concentration measurement in sample i, $(b_0)_i$ is the constant term of multiple regression for pollutants i, b_{pi} is the coefficient of the source p for pollutant i, and the $APCS_p$ is the scaled value of the rotated factor p for the samples. The APCS were estimated by subtracting the factor scores for the artificial sample from those of each true sample. Subsequently, the multiple linear regression (MLR) procedure was applied to estimate the source contribution, using PM_{10} mass concentrations as dependent variables and absolute score factors as independent variables. The main advantage of PCA-APCS is that it is capable of identifying different sources and their composition features, without any prior knowledge about the sources.

2.6. Human health risk assessment

The model of human health risk assessment for inhalation exposure was applied as per the USEPA (1989). In this study, Pb, Cd, Co, Ni, Cu and Cr were regarded as non-cancer effect elements based on the classification group orders defined by the IARC (International Agency for Research on Cancer) (IARC, 2011). For this, the mean concentrations of these trace metals were used as input data in order to assess how exposure to PM_{10} impacted children's health. Exposure assessment was undertaken to estimate the degree of risk children were at from trace metals (non-carcinogenic and carcinogenic) being absorbed through the inhalation pathway-the primary route of PM_{10} (Hieu and Lee, 2010; Massey et al., 2012). The average daily doses (ADD) for non-carcinogenic elements were calculated using the following equation:

$$ADD = \left(\frac{C \times IR_{inh} \times EF \times ED \times ET}{BW \times AT} \right) \times CF \tag{3}$$

where, C represents the concentration of each individual metal in indoor PM_{10} (ng/m^3); IR_{inh} is the inhalation rate per day (m^3/d); EF is the exposure frequency (d/y); ED is the exposure duration to air (y); ET is the exposure time (h/d), BW is the body weight of the subject (kg), AT is the average time for non-carcinogenic health effects to develop, (d) and CF is the conversion factor. Two population groups were included in the health risk assessment: children and adults. The values for the parameters are listed in Table S2.

Non-carcinogenic health risks were assessed by the calculation of the hazard quotient (HQ) and hazard index (HI). The ADD of each element was divided by the specific reference doses (RfD) to yield a HQ. The sum of the hazard quotients (HQ) for all trace metals produced a hazard index (HI). Whereby when the HI is less than one it is believed that there is no increased risk of non-cancer health effects at a site. In addition, it can be assumed that non-cancer effects would not be expected to occur as the result of the presence of any individual chemical or their combination with others. However, when HI exceeds one, there is a possibility that some non-cancer effects may well occur. Generally speaking, larger HQs are cause for greater levels of concern. The HQ and HI are expressed as follows:

$$HQ = \frac{ADD}{RfD} \tag{4}$$

$$HI = \sum HQ \tag{5}$$

3. Results and discussion

3.1. PM_{10} concentrations

The descriptive statistics of 8 h indoor and outdoor PM_{10} concentrations measured at two sampling schools, S1 (urban) and S2 (suburban) are presented in Table 1. The PM_{10} concentrations recorded in the classroom at the school in Kuala Lumpur's city centre (S1) ranged between 25 and 125 g/m³, with an average of 82 g/m³, while the concentrations of PM_{10} at the school in Putrajaya (S2),

Table 1
Descriptive statistics of 8 h mean indoor and outdoor PM_{10} and its components.

	Unit	S1 (City centre)						S2 (Suburban)					
		Indoor ($n=22$)			Outdoor ($n=22$)			Indoor ($n=22$)			Outdoor ($n=22$)		
		Avg ± SD	Min	Max	Avg ± SD	Min	Max	Avg ± SD	Min	Max	Avg ± SD	Min	Max
PM_{10}	g/m³	82 ± 27	25	125	80 ± 26	33	129	77 ± 26	17	117	75 ± 24	38	121
Ionic													
Na^+	g/m³	5.11 ± 3.51	Nd	11.5	5.21 ± 3.54	1.20	10.4	5.16 ± 1.86	Nd	9.13	5.21 ± 2.19	1.30	9.87
NH_4^+	g/m³	0.591 ± 0.260	Nd	1.20	1.02 ± 0.607	0.267	2.63	1.40 ± 0.678	0.275	2.80	1.55 ± 0.884	0.27	3.40
K^+	g/m³	1.08 ± 0.169	Nd	1.57	1.53 ± 0.733	Nd	2.97	1.46 ± 0.221	Nd	2.03	1.46 ± 0.308	Nd	2.20
Ca^{2+}	g/m³	10.1 ± 1.26	Nd	12.6	8.95 ± 1.54	Nd	10.9	4.49 ± 3.20	1.27	11.3	3.98 ± 3.02	1.03	11.2
Mg^{2+}	g/m³	0.863 ± 0.054	Nd	0.933	0.821 ± 0.070	0.771	1.07	0.635 ± 0.007	Nd	0.643	0.713 ± 0.010	Nd	0.733
NO_3^-	g/m³	4.61 ± 1.81	Nd	7.80	4.85 ± 1.41	Nd	7.63	5.47 ± 2.24	Nd	10.5	4.98 ± 2.38	Nd	10.3
SO_4^{2-}	g/m³	4.50 ± 0.644	Nd	5.83	4.28 ± 1.30	Nd	6.80	5.12 ± 2.75	Nd	9.60	5.49 ± 2.84	1.47	10.0
Trace metals													
Al	ng/m³	3700 ± 1830	942	7740	3480 ± 1460	1170	5970	4360 ± 1500	1760	7180	3840 ± 1380	1070	6780
Fe	ng/m³	1370 ± 707	129	2810	1050 ± 553	33.9	1880	1030 ± 613	10.4	2090	901 ± 426	114	1800
Zn	ng/m³	3610 ± 1120	1650	5450	3980 ± 1420	1700	7360	4420 ± 1190	1820	6340	4400 ± 797	2700	6120
Pb	ng/m³	17.6 ± 7.22	2.97	35.8	12.5 ± 5.22	0.551	20.9	27.5 ± 15.2	0.801	53.9	10.7 ± 4.33	5.47	21.9
As	ng/m³	1.59 ± 0.698	0.164	3.00	1.23 ± 0.688	0.081	2.41	2.57 ± 0.952	0.581	4.58	2.36 ± 1.45	0.414	5.75
Cd	ng/m³	0.266 ± 0.145	Nd	0.610	0.259 ± 0.082	Nd	0.443	0.180 ± 0.112	Nd	0.360	0.273 ± 0.104	Nd	0.443
Cr	ng/m³	433 ± 215	137	906	454 ± 298	65.5	1126	392 ± 178	102	635	395 ± 197	87.4	772
Co	ng/m³	5.36 ± 1.73	2.32	8.74	5.65 ± 2.52	0.407	9.24	5.36 ± 2.89	1.41	11.4	4.67 ± 1.98	1.24	7.57
Cu	ng/m³	23.8 ± 10.3	10.6	46.3	28.7 ± 15.8	5.26	70.3	28.5 ± 23.4	2.43	93.9	24.7 ± 16.8	3.76	60.0
Ni	ng/m³	85.9 ± 40.3	32.6	182	145 ± 72.0	42.3	251	126 ± 70.4	46.2	321	107 ± 37.9	33.9	159
V	ng/m³	12.5 ± 3.09	6.80	17.7	11.4 ± 4.44	4.64	19.7	11.1 ± 4.34	4.72	21.3	9.83 ± 4.62	3.22	19.9

Nd: Not detected.

ranged between 17 and 117 g/m³. The mean concentration of PM_{10} recorded in the classroom at S2 was found to be slightly lower than that at S1 with the value 77 g/m³. No significant difference ($p > 0.05$) was observed regarding indoor PM_{10} concentrations between the two different classrooms.

The average 8 h concentration of indoor PM_{10} in this study was considered to be low. No exceedances were observed when this concentration was compared to the PM_{10} concentration suggested in Hong Kong's air quality standards for public buildings, which is 180 g/m³ (good class) for an 8 h averaging time (IAQMG, 2003). Nevertheless, the average concentration recorded was far greater (74–76% exceeded) than the target value suggested by the Finnish Society of Indoor Air Quality and Climate (2001), which is 20 g/m³ within the same averaging time (Herring, 2006). When compared

with other similar studies, it was found that the average concentration of indoor PM_{10} recorded in this study was close to the value determined in previous studies, (70.6 g/m³), as reported in a study undertaken in a Serbian primary school by Jovanović et al. (2014), (66.7–81.3 g/m³) at childcare centres in Korea (Oh et al., 2014), (108 g/m³) at a school in Wroclaw, Poland (Zwoździak et al., 2013) and (30–146 g/m³) at three primary schools in Lisbon, Portugal (Almeida et al., 2011). However, the indoor PM_{10} concentration in this study was found to be three times higher when compared to the value (24.3 g/m³) obtained in a study carried out at 39 schools in Barcelona, Spain (Viana et al., 2013).

Outdoor PM_{10} concentrations measured at S1 (80 g/m³) and S2 (75 g/m³) were found to be slightly lower when compared to indoor air. The average outdoor concentrations of PM_{10} recorded at S1 and
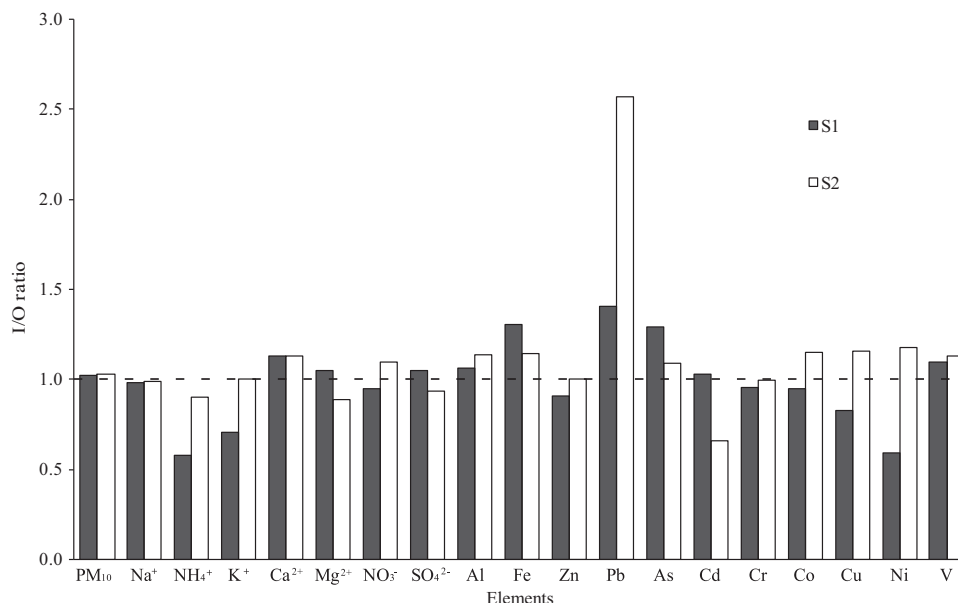

Fig. 2. Indoor-to-outdoor (I/O) ratios of PM_{10} and its elements.

S2 ranged from 33 to 129 g/m^3 and from 38 and 121 g/m^3, respectively. The average concentrations of indoor PM$_{10}$ compared to outdoor are given as the indoor-to-outdoor (I/O) ratio, for which the value of I/O ratio for each school was 1.03 (Fig. 2), thus indicating equal indoor and outdoor levels. This result concurs with those obtained from previous studies carried out at schools where I/O ratios were also found to exceed one (Braniš et al., 2005; Habil et al., 2013; Janssen et al., 1997; Pegas et al., 2012).

3.2. Chemical composition

3.2.1. Indoor concentrations of water-soluble ionic species

The 8 h mean concentrations of indoor water-soluble ionic species (WSIS) and trace metals in PM$_{10}$ are presented in Table 1. For indoor S1, the ionic concentrations in PM$_{10}$ can be arranged in descending order, namely as: Ca^{2+} > Na^+ > NO_3^- > SO_4^{2-} > K^+ > Mg^{2+} > NH_4^+. Ca^{2+} in PM$_{10}$ along with Na^+, NO_3^- and SO_4^{2-} had higher contributions to the total ionic mass. Ca^{2+} and Na^+ together accounted for approximately 50% of the total ionic mass and 19% of the PM$_{10}$ mass. Meanwhile, NO_3^- and SO_4^{2-} accounted for around 34% of the total ionic mass (11% of the PM$_{10}$ mass). The rest of the species, such as K^+, Mg^{2+} and NH_4^+, represented 4%, 3% and 2% of total ionic mass, respectively. A comparison of ionic concentrations between indoor S1 and S2 revealed that the concentration of Ca^{2+} (10.1 g/m^3) at S1 was twice as high as that of Ca^{2+} (4.49 g/m^3) at S2, indicating that the source could well be from the use of chalk on the blackboard.

The concentrations of WSIS measured in the PM$_{10}$ collected indoors at S2 can be arranged in the following descending order: NO_3^- > Na^+ > SO_4^{2-} > Ca^{2+} > K^+ > NH_4^+ > Mg^{2+}. Of all the analysed ions, NO_3^- was found to be the largest component from those collected indoors (S2) and its contribution to total ions was approximately 23%, followed by SO_4^{2-}, Na^+, Ca^{2+}, K^+, NH_4^+ and Mg^{2+} with 22%, 22%, 19%, 6%, 6% and 3% of total ionic mass, respectively. On average, the concentrations of indoor Na^+, NH_4^+, K^+, NO_3^- and SO_4^{2-} were found to be higher (5.16, 1.40, 1.46, 5.47 and 5.12 g/m^3, respectively) at S2 compared to the concentrations (5.11, 0.591, 1.08, 4.61 and 4.50 g/m^3, respectively) at S1. The high NH_4^+ concentration indoors at S2 (1.40 g/m^3) was twice as high as at S1 (0.591 g/m^3), which suggests that the contribution of this pollutant is possibly from sources such as: agricultural waste, particularly from agricultural activities occurring at palm oil plantations in the area surrounding the sampling school. Among the ionic concentrations studied, Ca^{2+}, Mg^{2+}, SO_4^{2-} and NH_4^+ showed significant differences ($p < 0.05$) between indoor S1 and S2.

Comparative data showing the concentrations of elements in indoor PM$_{10}$ from this study as well as from those undertaken in other countries is presented in Table S3. Concentrations of WSIS in indoor PM$_{10}$ samples from this study were all found to concur with results from studies conducted at schools in India (Chithra and Nagendra, 2013), with the exception of SO_4^{2-} concentrations, which were recorded as being almost three times lower. In contrast, concentrations of SO_4^{2-} in this study were noted as being greater than those from schools in other countries including Spain, Portugal and Germany. Whilst comparisons made with the same studies showed higher mean concentrations of Na^+, K^+, Ca^{2+}, Mg^{2+} and NO_3^- (Fromme et al., 2008; Pegas et al., 2012; Viana et al., 2013).

The relationships between the indoor concentrations of major ions studied in city centre (S1) and suburban (S2) schools were determined using Pearson correlation analysis; the results of which are shown in Table S4. From this table, it can be seen that the concentrations of the some of the major ions analysed in this study were related to each other, indicating that they most likely originated from the same source. At S1, the concentration of SO_4^{2-} was significantly related to that of K^+ ($p < 0.01$, $r = 0.697$) and the concentrations of Na^+ and Ca^{2+} were strongly correlated ($p < 0.01$, $r = 0.867$). In addition, the concentration of Na^+ exhibited a significant correlation with that of NH_4^+ ($p < 0.01$, $r = 0.572$) at S1. Similar results showed there was a significant correlation between Na^+ and NH_4^+ which was also noted at S2. Likewise, the correlation of K^+ with Na^+ and NH_4^+ were 0.489 and 0.520, respectively.

3.2.2. Indoor concentrations of trace metals

The average total of trace metals concentrations was 9265 ng/m^3, which accounted for 11% of the total indoor PM$_{10}$ mass concentration. As per Table 1, Al was found to be the most abundant metal in the PM$_{10}$ collected from the classroom at S1, followed by Zn, Fe, Cr, Ni, Cu, Pb, V, Co, As and Cd. Among the trace metals analysed, Al, Zn and Fe were the predominant metals with 8680 ng/m^3, contributing to about 10% of the total PM$_{10}$ mass. At S2, the average mass of total trace metals was 10 398 ng/m^3, which accounted for 13% of the total indoor PM$_{10}$ mass concentration. Zn had the highest concentration of the remaining trace metals 6% of the total PM$_{10}$ mass concentration, followed by Al (6%), Fe (1%) and Cr (1%). Other trace metals, namely Ni, Cu, Pb, V, Co, As and Cd accounted for less than 1% of the total PM$_{10}$ mass concentration. Among the trace metals analysed, Pb, Zn, As, and Ni were present in significantly higher concentrations in the PM$_{10}$ samples taken from inside the classroom at S2 compared with those taken from S1's classroom ($p < 0.05$).

The concentrations of certain metals (Al, Fe, Zn, Cr and Cu) as measured in this study, were noted as being rather high when compared to the concentrations recorded in school environments in several urban areas in Europe and elsewhere. For example, average concentrations of Al, Fe and Zn in our study were two to six times higher than those reported in a study conducted by Avigo Jr et al. (2008), which were all given as being below 700 ng/m^3. Lower indoor mean concentrations of several elements such as: Zn, Fe, As, Cr, Co, Pb and Cu were also noted in a study by Viana et al. (2013). However, higher concentrations of Pb and Cu, which were almost 30–80% greater than those from the present study, were observed at several schools in Wroclaw, Poland (Zwoździak et al., 2013) and elsewhere in Malaysia (Latif et al., 2011).

The correlation coefficient between the trace metals concentrations for both sampling sites is presented in Table S5. A significant correlation was found between Al–Zn ($r = 0.694$), V–Co ($r = 0.618$) and V–As ($r = 0.543$), which were all significant at 0.01 level at S1. Cd–Cu ($r = 0.511$), Cd–V ($r = 0.488$), Cd–Pb ($r = 0.447$), Zn–Fe ($r = 0.434$), Zn–Ni ($r = 0.435$), As–Cr ($r = 0.443$) and As–Co ($r = 0.445$) were significantly correlated with each other at 0.05 level. Meanwhile at S2, a positive significant correlation ($p < 0.01$) was found between Al–Zn ($r = 0.883$), Zn–Co ($r = 0.554$), Cd–V ($r = 0.608$), Cr–Co ($r = 0.556$) and Co–V ($r = 0.778$), while a negative correlation was found between Pb–Cr ($r = -0.475$). In addition, a significant correlation ($p < 0.05$) was observed for Ni with Cr ($r = 0.518$), Co ($r = 0.440$) and V ($r = 0.495$). A positive correlation at 0.05 level could also be observed for Fe with Cu ($r = 0.525$), V ($r = 0.457$) and Cr ($r = 0.434$). Pb exhibited a negative correlation at 0.05 level with Cr ($r = -0.475$) and a negative correlation could also be observed between Zn and V ($r = -0.431$).

3.3. Relationships of indoor and outdoor concentrations of chemical composition

The average chemical composition concentrations of indoor to that of outdoor are shown as the indoor-to-outdoor (I/O) ratio. The I/O ratio has been used in order to describe the relationship between indoor and outdoor chemical composition concentrations (Fig. 2). Based on the I/O ratio, the elements were divided into two categories: those having a ratio above 1 and those having one below 1. Average I/O ratios in this study ranged from 0.58 to 2.56.

The lowest value determined was for NH_4^+ (S1) in line with the absence of indoor sources. The highest value, however, was for Pb (S2), indicating that lead-based paint on the walls or other painted equipments (desk and chairs) most likely contributed to elevated Pb levels along with the art orientated activities which the seven-year-old children who occupied the classroom at S2 were involved in, such as: drawing, painting and colouring. Recent research by Rebelo et al. (2015) also determined the Pb content in artists' paints, predominantly in gouache, acrylics, watercolours and finger paints.

When comparing outdoor concentrations of the elements studied with those measured inside the classroom at S1, outdoor concentrations of Na^+, NH_4^+, K^+, NO_3^-, Zn, Cr, Co, Cu, and Ni were found to be significantly higher than those indoors with an I/O ratio of less than 1 (ranging between 0.58 and 0.98). Meanwhile, at S2, elements such as SO_4^{2-}, Na^+, NH_4^+, Mg^{2+}, Cd and Cr were found to be higher outdoors than indoors with an I/O ratio ranging between 0.66 and 0.99, thus indicating fewer indoor sources and the contribution of outdoor sources associated with these particular elements. This phenomenon could be explained by the indoor-outdoor air exchange as windows and doors were generally left open so as to ventilate the classroom during class sessions (at least eight hours per day). Additionally, the outdoor elements could have been transported into the classroom via students' shoes, hair, skin and clothes. Ca^{2+}, Al, Fe, Pb and As were all found to have ratios close to 1 at both schools. This finding must relate to the infiltration of the elements from outdoor sources as no obvious indoor sources for these elements were identified, apart from for Ca^{2+} which links to the use of chalk by teachers at S1. A high I/O ratio for these elements was also found in a study by Goyal and Khare (2009) and clearly showed that the building envelope permitted entry of pollutants from outside. The outdoor infiltration process was noted to occur as a result of natural ventilation, namely through doors and windows being left open (Latif et al., 2014; Razali et al., 2015), such that it could well be the case that the elements were influenced by local pollution sources from outside. S1 itself was surrounded by a commercial area and near a busy road which experienced a high density of traffic whilst S2 was surrounded by a combination of residential and agriculture areas. Despite Putrajaya (S2) having been transformed into Malaysia's administrative capital, there are still some agricultural activities, particularly palm oil plantations and residential building projects which are ongoing in this area.

3.4. Enrichment factor analysis

The enrichment factor (EF) of trace metals in PM_{10} relative to the Earth's upper crust can indicate crustal and non-crustal origins. According to the EFs analysis results, the selected metals in PM_{10} were divided into three groups. Group 1 included those metals with an EF value below 10, indicating a low enrichment factor. Group 2 included those metals showing an EF in the range from 10 to 100 which were considered to be moderately-enriched. Group 3 represented metals with an EF value greater than 100, which were therefore considered to be highly-enriched. The dominance of trace metals, such as: Al, Zn, Fe, Cr and Ni at both schools indicates the existence of indoor anthropogenic sources for these elements (Almeida et al., 2011; Avigo Jr et al., 2008). This concurs with the enrichment factor result shown in Fig. S2, which demonstrate high enrichment values for Zn, Cr and Ni (> 100), as well as moderately-enriched values for Cd, Cu, Pb, As and Co, thus indicating the influence of anthropogenic sources. The high EF of these metals could be explained by motor vehicle exhaust emissions, as well as industrial and construction activities occurring near to the sampling schools.

In this case, although Cr showed low concentration levels among all the metals studied, the EF values were high thus suggesting anthropogenic sources contributed to this metal. In contrast, the enrichment factor for Al was found to be < 10 slightly even though it had a higher concentration among the trace metals analysed, suggesting that it predominantly originated from the natural particles of mineral dust-re-suspension. The intense traffic and construction activities that were being undertaken in close proximity to the schools were expected to enhance the re-suspension of dust which originated from outside of the school buildings. When the results of the EF obtained at S1 and S2 were compared, the estimated EFs for each metal were found to be very similar for both sampling classrooms. For example, the sequences of the EF values for the metals in the indoor PM_{10} at both S1 and S2 were almost identical. Despite the characteristics of the schools and their surroundings being quite different, the grouping of each metal was still in accordance with the typical anthropogenic and natural sources. The closely-related EF distribution patterns for each metal suggest a common source for these metals and indicate that the location of both schools, namely in reference to the city centre and suburban areas, contribute to the major input of metals at both sampling sites.

3.5. Chemical mass closure

The chemical mass closure of identified chemical classes for indoor PM_{10} at the two monitoring schools is shown in Figure S3. The results may be considered as quantitative estimations for all chemical classes and their contributions reflecting differences in emission sources. The overall PM_{10} mass was divided into five main components consisting of: mineral aerosols (MIN), sea salt (SS), secondary inorganic aerosols (SIA), trace metals (TM) and unidentified mass (UM). The MIN originated from soil and estimates were based on the quantities of crustal elements such as: Al, Ca, Fe and K assumed to be present through the existence of metal oxides in the soil (Al_2O_3, CaO, Fe_2O_3, K_2O) (Sillanpaa et al., 2006). Thus, the mass of MIN type aerosols was computed by multiplying each element by the appropriate factor for converting them to their common oxides (Cheung et al., 2011; Hueglin et al., 2005): $[MIN] = 1.89[Al] + 1.40[Ca] + 1.21[K] + 1.43[Fe]$. SIA was given as the sum of non-sea salt (nss)-SO_4^{2-}, NO_3^- and NH_4^+, where nss-SO_4^{2-} was determined based on: $[SO_4^{2-}] - 0.252[Na^+]$ (Cheung et al., 2011). Sea salt was calculated by using Na^+ concentrations multiplied by 1.4486, where 1.4486 is the ratio of the concentrations of all elements except Cl^- in sea water to the Na^+ concentration (Viana et al., 2007). The quantity of TM was calculated from the concentrations of trace metals (excluding mineral elements). In the present study, the unidentified mass group was used to refer to undetermined chemical constituents.

The percentage fractions of each constituent were found to be quite similar for both schools. On average, the indoor percentages of the UM group for both schools dominated the total mass of PM_{10}, representing 41–45% of the total mass. It was observed that the percentages for the MIN group at indoor S1 (31% of total mass) were higher than S2 (27% of total mass). Conversely, the SIA were found to be higher inside S2 (at 14% of total mass) than S1 (at 9% of total mass). SS was found inside the classroom of each school at percentages lower than 10% of total mass. Recent studies also observed a large unaccounted for mass (20–50%) in PM_{10} samples (Cheng et al., 2015; Cheung et al., 2011). The unaccounted mass included other unmeasured mineral components such as: carbonate and silica, carbonaceous particles (e.g., biogenic, soot and organic particles) and fibres. Viana et al. (2014) reported that organic matter was the most dominant for coarse particles ($PM_{2.5–10}$) in Barcelona schools, representing about 45–77% of the mass.

Table 2
Varimax rotated PCA factor loadings. Loadings > 0.75 are marked in bold; loadings between 0.50 and 0.75 are in italics.

Location	Variable	Al	Fe	Pb	Zn	As	Cd	Cr	Co	Cu	Ni	V	NO_3^-	SO_4^{2-}	Na^+	NH_4^+	K^+	Ca^{2+}	Mg^{2+}	Eigenvector	Cumulative (%)
S1	F1	0.09	−0.16	0.62	0.08	0.24	**0.76**	0.03	0.66	0.70	0.36	0.56	0.08	−0.04	0.03	−0.21	−0.20	0.05	0.55	4.38	24.31
	F2	**−0.87**	0.02	0.30	0.61	0.43	−0.24	−0.11	0.19	−0.21	0.01	0.04	−0.20	0.09	0.27	0.60	−0.04	**0.78**	−0.14	3.34	42.87
	F3	0.25	**0.75**	−0.34	0.56	0.17	−0.22	0.07	0.12	0.21	0.49	−0.11	**0.78**	0.14	−0.27	−0.37	−0.31	0.33	0.32	1.88	53.32
	F4	−0.12	0.08	−0.17	−0.31	0.64	0.28	0.72	0.27	−0.11	−0.36	0.41	0.18	−0.10	**0.77**	0.39	−0.15	−0.10	−0.28	1.85	63.61
	F5	0.07	−0.18	−0.26	−0.01	−0.27	0.08	−0.31	−0.30	−0.01	−0.11	−0.51	0.35	**0.82**	0.03	−0.10	**0.77**	0.21	0.48	1.37	71.19
S2	F1	**−0.91**	−0.20	−0.39	**−0.89**	−0.08	−0.13	0.24	0.49	−0.02	0.09	0.26	0.25	−0.12	0.65	0.35	0.64	0.41	−0.37	5.58	31.00
	F2	0.05	0.09	−0.04	−0.15	0.18	**0.79**	0.04	0.49	0.00	0.31	**0.76**	−0.31	0.20	0.50	**0.78**	0.41	0.56	0.05	3.07	48.06
	F3	−0.03	**0.81**	−0.16	−0.05	0.27	0.08	0.65	0.51	**0.78**	0.58	0.50	−0.16	0.15	0.14	−0.14	−0.26	0.46	0.16	1.80	58.03
	F4	−0.09	−0.08	−0.05	0.08	**0.80**	0.13	0.21	−0.00	−0.02	0.62	−0.12	−0.23	0.32	−0.24	−0.11	−0.17	−0.17	0.70	1.61	67.00
	F5	0.02	−0.10	0.63	0.13	0.20	0.07	−0.33	−0.17	0.15	0.09	−0.08	0.69	**0.75**	−0.20	−0.02	−0.07	−0.21	0.24	1.31	74.27

3.6. Source apportionment

According to the PCA results, five factors with eigenvalues > 1 were extracted from indoor S1 and S2. However, only factor loading > 0.75 (strong) and between 0.50 and 0.75 (moderate), which demonstrate significance within a factor, were selected to interpret the sources for each factor. Some of the elements appeared in diverse combinations for the different factors at each school, suggesting that these elements originated from multiple sources. The large variation in terms of sources might well be attributed to the different land uses occurring near the sampling schools, including busy roadways along with commercial and residential sites, and industrial and agricultural areas. Table 2 shows PCA results for indoors S1 and S2.

3.6.1. Indoor S1

The total percentage variance was approximately 71% for indoor S1 and is explained by five factors. Factor 1 had a strong loading for Cd, and moderate loadings of Cu, Co, Pb, V and Mg^{2+}, these were attributed to the re-suspension of road dust. Whilst Cd had been associated with smelters and road dust in previous studies (Khan et al., 2015; Sternbeck et al., 2002; Thurston et al., 2011), they may well have derived from the combustion of accumulators and vehicle carburettors in the current study due to the school being located close to a major road. In other studies, it was concluded that Cu was derived from brake linings and was a major contributor to the element's presence in coarse particles (Godoy et al., 2009; Harrison et al., 2003). Whilst V is widely distributed in the earth's crust, its distribution in the environment has been linked to the emission of fossil fuels, particularly the combustion of heavy fuel oil (Amato et al., 2011; Balakrishna et al., 2011; Gugamsetty et al., 2012; Khan et al., 2015). In addition, V in the form of vanadium oxide, is a component of steel which is used in the production of automobile parts, spring and ball bearings (Baez et al., 2007; De Miguel et al., 2007). As such, this possible element might have entered the classroom via the doors and windows, which were frequently left open for ventilation during the school session.

Factor 2 revealed a strong loading for Al and Ca^{2+}, and a moderate loading for Zn and NH_4^+. The sources were identified as non-combustion motor vehicles coupled with indoor activities in which elements were emitted in the classroom through the re-suspension of dust. Activities such as moving in and out of the classroom by the occupants most likely played a role in the re-suspension of surface dust. Indoor cleaning activities that took place during after school hours may also have had a significant effect on the indoor levels of these metals. Moreover, Al and Zn in this classroom might well be have been derived from corrosion occurring on windows and door frames. These elements were also noted as being a component in several products which were applied in order to protect steel, walls, wooden surfaces, doors and windows (John et al., 2007). At S1, teachers used chalks for teaching purposes. Ca^{2+} is often associated with the chalk used on blackboards and is also a common constituent of gypsum ($CaSO_4.2H_2O$) as well as common building materials such as plaster (Pey et al., 2009; Srivastava and Jain, 2007). The re-suspended of dust from the broken cement floor, as a result of cleaning activities, was also found to contribute to Ca^{2+}. Basha et al. (2010) have, in a previous study, demonstrated that a major source of Zn in atmospheric particles is the abrasion of rubber tyres on roads. A study in Xi'an, China also found that Zn was widely used as a component in building materials, in the coal and oil industries, as well as in light industry (Cao et al., 2011).

Factor 3 had strong loadings of NO_3^- and Fe, both of which are frequently used as indicators of soil or crustal sources and commonly found together in soil-derived particles. It is possible that

soil dust from outside was brought into the classroom on students' clothing and shoes. In fact, windblown dust emanating from paved or unpaved roads and construction sites located near the school would no doubt have been brought into the classroom through dust. Factor 4 showed a strong positive loading for Na^+ which may be attributed to the effects of sea spray penetrating from outside. Sea breezes in the daytime were conducive to transporting marine aerosols from coastal areas (about 40 km from the school) and spreading them inland. In addition, a moderate loading both for Cr and As in this factor suggested industrial activity as a source, due to the presence of automobile repair shops in close proximity to the sampling site. High temperatures during industrial processes released these metals into the atmosphere as fly ash, explaining why dispersion into the surrounding area was greater (Subhani et al., 2015). Factor 5 showed a strong positive loading for SO_4^{2-} and K^+, which mostly originated from the local emission sources, such as vehicle exhausts and domestic waste burning. In a previous study, vehicular emissions and dust from paved roads were also listed as sources of SO_4^{2-} (Chakraborty and Gupta, 2010). According to Pateraki et al. (2012), the great photochemical activity favoured the formation of SO_4^{2-} measured over an urban Mediterranean area.

3.6.2. Indoor S2

More than 74% of the variance at indoor S2 was extracted in the form of five factors. Factor 1 showed a strong loading of two metals, Al and Zn, and a moderate loading of Na^+ and K^+, indicating that the source was possibly a mixture of natural and anthropogenic origins. Zn and Al were found in the indoor environment as components in building materials, with similar sources (indoor activities) as discussed above for Factor 2 at S1. The presence of Na^+ was quite likely to be the result of sea breezes, since S2 was located about 38 km from the Malacca Strait. K^+ was predominantly the result of biomass burning which occurred near the school area as a result of agricultural activities. These activities included forest-clearing and the burning of agricultural waste, particularly from palm oil plantations.

A strong loading for Cd, NH_4^+ and V, along with a moderate loading for Ca^{2+} and Na^+, were identified in Factor 2. These elements are known to be common constituents of motor vehicle emission dust and were coupled with construction and building materials. Cd and V were associated with Factor 1 at S1. These two trace metals have been widely cited in literature as components of road dust and are released from motor vehicle-related sources (Amato et al., 2011; Gugamsetty et al., 2012; Khan et al., 2015; Sternbeck et al., 2002; Thurston et al., 2011). Since ongoing construction was taking place as part of residential development near to the sampling periods, it was expected that metallic compounds such as stainless steel would also be present (Almeida et al., 2005; Goix et al., 2011; Song et al., 2006) along with cement and sand (Duan et al., 2012). Factor 3 was identified as road dust through the presence of a strong loading of Fe and Cu, and a moderate loading of Cr, Ni, Co and V. These elements have previously been determined as markers of either exhaust emissions or non-engine combustion sources, such as brake and clutch wear (Al-Khasman, 2007; Weber et al., 2000). The presence of these elements indoors resulted from the corrosion of the metallic parts of vehicles along with residue oil being deposited on nearby roads (Balakrishna et al., 2011; Thurston et al., 2011) and subsequently re-suspended in crustal or mineral dust (Fe).

Factor 4 was found to originate from an industrial source due to the presence of a strong loading of As and a moderate loading of Ni and Mg^{2+}, showing these metals to be potential markers for the small-scale industries located close to the sampling site. Additionally, these compounds may well have originated from paint sources which were predominantly generated from the surfaces of

buildings (Eštoková et al., 2010). Factor 5 was identified as originating from crustal sources, such as: soil dust, based on the composition of elements such as a strong loading of SO_4^{2-} and a moderate loading of NO_3^- and Pb. The soil dust was most likely mixed with Pb which could be traced primarily to the Pb found in paint pigments. Paint which had peeled from interior and exterior wall, as well as from the equipment in the school playground, was found to have contributed to the level of lead contamination in the classrooms. Studies previously undertaken at several schools and preschools in Malaysia and at homes in India, reported that Pb is found at high levels in the paint used on walls (Kulshrestha, 2014; Latif et al., 2011). Furthermore, suspended sand and dust were found to be effectively transported indoors through open doors and windows on drier days.

In order to apportion the percentage mass contribution of each identified source, the absolute factor scores were regressed against the 8 h PM_{10} mass concentration using MLR analysis as per Thurston and Spengler (1985). The contribution of the possible sources of different elements in PM_{10} is presented in Fig. 3. Inside the classroom at S1, re-suspension of road dust dominated with a 36% contribution to PM_{10} on a mass basis, followed by sea spray and industrial activity (31%), biomass burning (19%), non-combustion motor vehicles and indoor activities (9%) and crustal sources (5%). In contrast, the contribution of road dust accounted for 55% of PM_{10} inside the classroom at S2. The other sources were indoor activities, sea spray and biomass burning (21%), motor vehicle emissions, construction and building materials (12%), crustal sources (11%) and industrial activity (1%). Our comprehensive analysis of the data shows that the possible sources contributing to the range of elements in PM_{10} at the two different sites arose locally.

3.7. Human health risk assessment

The results of the hazard quotient (HQ) and hazard index (HI) were derived from the average daily doses (ADD) values calculated with relative toxicity values. Both HQ and HI were calculated to evaluate the non-carcinogenic health risks from individual metal and combined metals (Pb, Cd, Cr, Co, Ni and Cu). Due to being an important pathway for the exposure to trace metals with respect to PM_{10} in the classrooms, only the inhalation pathway was focused on in this study. The HQ and HI results are presented in Table 3.

Based on the exposure assessment, it was determined that the ADD values for both children and adults at each school were lower than the reference dose (RfD), thus indicating there were unlikely to be any adverse health effects apart from those deriving from Cr (Wei et al., 2015). The Cr ADD values for both populations at the two sampling schools demonstrate that the inhalation of PM_{10} can potentially cause adverse effects on human health. The HQs of the trace metals for both groups at indoor S1 were ranked in descending order Cr > Co > Ni > Pb > Cu > Cd, whilst at indoor S2, the HQs decreased in the order of Cr > Co > Pb > Ni > Cu > Cd. Cr exhibited the highest values at each school (in the case of children) and were almost twice as high as the safe level of HQ=1, as determined by USEPA (1989), with a value of 1.86×10^0 (S1) and 1.69×10^0 (S2). A similar trend was also found for adults at S1 and S2, with values of 1.15×10^0 and 1.04×10^0, respectively. Other trace metals were found to have values below the safe level (HQ=1). Pb, Cd, Co, Cu and Ni were recorded in range between 3.27×10^{-5} (Cd) and 1.15×10^{-1} (Co), and between 3.40×10^{-5} (Cd) and 1.15×10^{-1} (Co) for children at S1 and S2, respectively, while for adults, the range of these metals at S1 and S2 varied between 2.02×10^{-5} (Cd) and 7.13×10^{-2} (Co), and 2.11×10^{-5} (Cd) and 7.13×10^{-2} (Co).

The HI (combined for all metals) for both groups at S1 and S2 was above the safe level of 1, indicating a higher potential of non-carcinogenic risk to children and adults, particularly in relation to
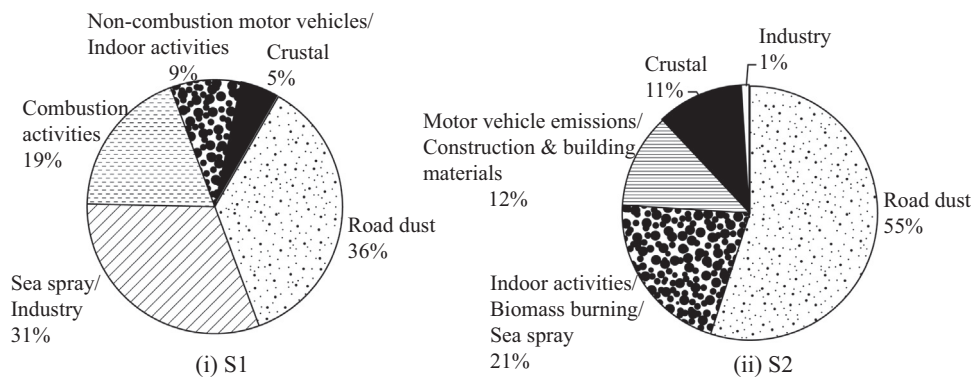
Fig. 3. Contribution of possible sources of elements in PM_{10} at the city centre school (S1) and suburban school (S2).

Table 3
Hazard quotients (HQ) and hazard index (HI) for non-carcinogenic metals via inhalation exposure of PM_{10} (Values in bold = the potential health risks were much greater than the acceptable levels, HQ or HI > 1).

| | RfD (mg/kg/d) | S1 | | | | S2 | | | |
| | | ADD (mg/kg/d) | | HQ | | ADD (mg/kg/d) | | HQ | |
		Children	Adults	Children	Adult	Children	Adults	Children	Adult
Pb	3.52×10^{-3}	2.28×10^{-6}	1.41×10^{-6}	6.48×10^{-4}	4.01×10^{-4}	3.38×10^{-6}	2.09×10^{-6}	9.60×10^{-4}	5.94×10^{-4}
Cd	1.00×10^{-3}	3.27×10^{-8}	2.02×10^{-8}	3.27×10^{-5}	2.02×10^{-5}	3.40×10^{-8}	2.11×10^{-8}	3.40×10^{-5}	2.11×10^{-5}
Cr	2.86×10^{-5}	5.32×10^{-5}	3.29×10^{-5}	$\mathbf{1.86 \times 10^{0}}$	$\mathbf{1.15 \times 10^{0}}$	4.82×10^{-5}	2.98×10^{-5}	$\mathbf{1.69 \times 10^{0}}$	$\mathbf{1.04 \times 10^{0}}$
Co	5.71×10^{-6}	6.58×10^{-7}	4.07×10^{-7}	1.15×10^{-1}	7.13×10^{-2}	6.59×10^{-7}	4.07×10^{-7}	1.15×10^{-1}	7.13×10^{-2}
Ni	2.06×10^{-2}	1.37×10^{-5}	8.45×10^{-6}	6.63×10^{-4}	4.10×10^{-4}	1.54×10^{-5}	9.55×10^{-6}	7.49×10^{-4}	4.63×10^{-4}
Cu	4.02×10^{-2}	2.92×10^{-6}	1.81×10^{-6}	7.27×10^{-5}	4.49×10^{-5}	3.51×10^{-6}	2.17×10^{-6}	8.72×10^{-5}	5.39×10^{-5}
				$\Sigma HI = \mathbf{1.98 \times 10^{0}}$	$\Sigma HI = \mathbf{1.22 \times 10^{0}}$			$\Sigma HI = \mathbf{1.80 \times 10^{0}}$	$\Sigma HI = \mathbf{1.12 \times 10^{0}}$

Cr exposure. Cr exposure to both groups, via inhalation exposure, was found to contribute about 94% to the total HI which strongly suggests that the presence of Cr raises concerns with regard to potential adverse health effects. The possible reason for the higher Cr result was that the metal was present in a greater concentration in PM_{10} compared with the other metals. In the present study, Cr predominantly originated from anthropogenic sources, such as: road dust, industrial and vehicle emissions and is considered to pose health risks to both the children and teachers occupying the classrooms. Additionally, results of the HI showed that children had a higher degree of exposure than adults because of their low tolerance to toxins and are also subject to a greater potential non-carcinogenic risk than adults. Similar findings by Hu et al. (2012) and Lu et al. (2014) determined that Cr presented the greatest potential risk to children's health from inhalation exposure in microenvironments, such as: campus buildings and nursery schools. The results also demonstrated that a degree of exposure to metals in PM_{10} could well increase the risk of individuals developing cardiovascular and respiratory diseases, as well as lung cancer, particularly among school children exposed to PM_{10} over the long-term. Exposure to Cr has been found to trigger various symptoms, including coughing, sneezing and nasal irritation in humans (RAIS, 2013).

4. Conclusions

The present study revealed that the indoor concentrations of PM_{10} at the school in Kuala Lumpur's city centre (S1) (82 g/m³) were higher when compared to the suburban area of Putrajaya (S2) (77 g/m³). Ca^{2+}, Na^+, SO_4^{2-} and NO_3^- dominated the composition of water-soluble ionic species in the PM_{10} samples

collected from both schools. Regarding trace metal composition, Al, Fe and Zn were found to be abundant in the PM_{10} samples and further analysis, using PCA-APCS for element source classification and contribution, showed that a major source of the elements in indoor PM_{10} for S1 and S2 was road dust, representing 36% and 55%, respectively. Other key sources of the elements in indoor PM_{10} samples from S1 were sea spray and industrial activity (31%), biomass burning (19%), non-combustion motor vehicles and indoor activities (9%) and crustal sources (5%). Other major sources at indoor S2 included mixed sources from indoor activities, biomass burning and sea spray (21%), motor vehicle emissions, construction and building materials (12%), crustal sources (11%) and industrial activity (1%). Zn and Cr were found to have the highest enrichment factor (EF), thus indicating the potential transport of outdoor pollutants from vehicle emissions and industrial sources into the school environment. The health risks associated with the inhalation of indoor PM_{10} emissions were estimated for both children and adults, and the results of HQ for Cr were found to be greater than the acceptable level of 1, as proposed by USEPA (1989). This indicates that the children in the school classrooms face a potentially high health risk and that the presence of certain trace elements (such as Cr) in high concentrations is cause for great concern. The results from this study provide a valuable evaluation of the health risks associated with exposure to trace elements in both of the classrooms selected for the study. Consequently, certain mitigation procedures should be conducted in order to reduce the distribution of toxic heavy metals in the school classroom, both in urban and suburban settings. Whilst building materials and classroom facilities were found to contribute to the presence of heavy metals, the influence of outside sources, such as motor vehicles, also necessitates regular monitoring. Additionally, further analysis of fine and ultrafine PM with a larger number of

samples and sampling points with additional information on meteorological factors such as ventilation, temperature and relative humidity needs to be undertaken by way of a comprehensive investigation associated with source apportionment approaches and the investigation of health risks, particularly in the school environment.

Acknowledgements

Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.ecoenv.2015.11.002.

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