Development of Land Use Regression Models for Particulate Matter and Associated Components in Low Air Pollutant Concentration Airshed

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Abstract

Perth, Western Australia represents an area where pollutant concentrations are considered low compared with international locations. Land Use Regression (LUR) models for PM_{10}, $PM_{2.5}$ and $PM_{2.5}$ Absorbance ($PM_{2.5}$Abs) along with their elemental components: Fe, K, Mn, V, S, Zn and Si were developed for the Perth Metropolitan area in order to estimate air pollutant concentrations across Perth. The most important predictor for PM_{10} was green spaces. Heavy vehicle traffic load was found to be the strongest predictor for $PM_{2.5}$Abs. Traffic variables were observed to be the important contributors for PM_{10} and $PM_{2.5}$ elements in Perth, except for $PM_{2.5}$ V which had distance to coast as the predominant predictors. Open green spaces explained more of the variability in the PM_{10} elements than for $PM_{2.5}$ elements, and population density was more important for $PM_{2.5}$ elements than for PM_{10} elements. The $PM_{2.5}$ and $PM_{2.5}$Abs LUR models explained 67% and 82% of the variance, respectively, but the PM_{10} model only explained 35% of the variance. The $PM_{2.5}$ models for Mn, V, and Zn explained between 70% and 90% of the variability in concentrations. PM_{10} V, Si, K, S and Fe models explained between 53% and 71% of the variability in respective concentrations. Testing the models using leave one-out cross validation, hold out validation and cross-hold out validation suggested the validity of LUR models for PM_{10}, $PM_{2.5}$ and $PM_{2.5}$Abs and their corresponding elements in Metropolitan Perth despite the relatively low concentrations.

Keywords: Land use regression (LUR) model, air pollution, particulate matter, PM elements

1. Introduction

Particulate matter (PM) is a heterogeneous mixture of suspended particles and varies in chemical composition and size (Liang, 2013). Long term exposures to PM mass with an aerodynamic diameter smaller than 10µm (PM_{10}) and 2.5µm ($PM_{2.5}$) respectively, have been associated with mortality, cardiovascular disease, lung cancer, and both chronic and acute respiratory diseases, including asthma, even at concentrations below ambient air quality standards (Andersen et al., 2012; Beelen et al., 2014; Cesaroni et al., 2014; Hoek et al., 2013; Raaschou-Nielsen et al., 2013).

The composition of PM may be important in determining the contribution of particular sources. A source apportionment study conducted in four Australian cities identified species-related sources of PM, including aluminium (Al) and silicon (Si) from crustal/soil dust; iron (Fe), copper (Cu), zinc (Zn) and Manganese (Mn) from motor vehicle emissions; potassium (K) from biomass burning emissions; and heavy metals (Fe, Cu, Zn) from industrial emissions (Chan et al., 2008; Larson et al., 2004).

There is some evidence that the composition of PM may also be important in determining health effects (Eeftens et al., 2014; Stanek et al., 2011). PM_{10}-associated zinc (Zn) has been associated with the risk of pneumonia among children in seven birth cohort studies in Europe (Fuertes et al., 2014). $PM_{2.5}$-associated nickel (Ni) and vanadium (V) have also been associated with daily mortality in time-series studies in two northern American cities (Zhou et al., 2010). Small decreases in lung function among young children have also been related with exposure to other PM-associated elements such as Cu, Fe, K, Si, sulfur (S) and Zn (Eeftens, et al., 2014).

There are relatively little data available on the intra-urban speciation of PM_{10} and $PM_{2.5}$ for estimating exposures in large population-based health studies in Australia. Land Use Regression (LUR) models have been used to predict small-scale spatial variations in exposure to air pollutants including species of PM, within cities in other location (Gulliver et al., 2011; Hoek et al., 2008; Zou et al., 2009). The recent European Study of Cohorts for Air Pollution Effects (ESCAPE) developed PM speciated LUR models for eight elements in 15 countries. Most of those LUR models explained a large fraction of the spatial variation within the study area with R^2 ranging from 50% and 79% (de Hoogh et al., 2013) and were used to investigate the associations with a range of health outcomes (Eeftens, et al., 2014).

This paper describes the development of LUR models for PM_{10}, $PM_{2.5}$, $PM_{2.5Absorbance}$ ($PM_{2.5}$Abs), and PM-associated elements (Cu, Fe, K, V, S, Si, Zn, Ni, and Mn) for the Perth metropolitan area, Western Australia, an area with lower air pollutant concentrations compared with most European and North American cities. The models were then used to assign exposures to PM and PM elements for a cohort of older men in Perth, the Health in Men Study (HIMS) (Norman et al., 2009).

2. Methods

2.1. Area of study

The study was conducted in the Perth Metropolitan Area which is the capital of Western Australia. It is located on the Indian Ocean with the Darling Ranges to the East, and with two main waterways, the Swan and Canning rivers. Perth metropolitan area has an area of 6,418 km² and the population was around 1.97 million in 2012. Its topography is mainly flat with an altitude of approximately 31.5 m above sea level and a Mediterranean climate (Yimin et al., 2003).

2.2. Sampling sites selection

The monitoring site selection has been described elsewhere (Dirgawati et al., 2015)**.** Twenty sites were selected to measure $PM_{2.5}$ and PM_{10}, comprising two regional background sites; eight urban background sites; and ten street sites based on the criteria describe in the protocol for the ESCAPE study (http://www.escapeproject.eu/manuals/). One of the regional background sites was co-located at a monitoring station operated by the Western Australian Department of Environment Regulation for comparison with standard government monitoring. A reference site was also operated continuously throughout the sampling period to adjust for any temporal variability of particle concentrations between seasons. Figure 1 shows a map of the sampling sites.

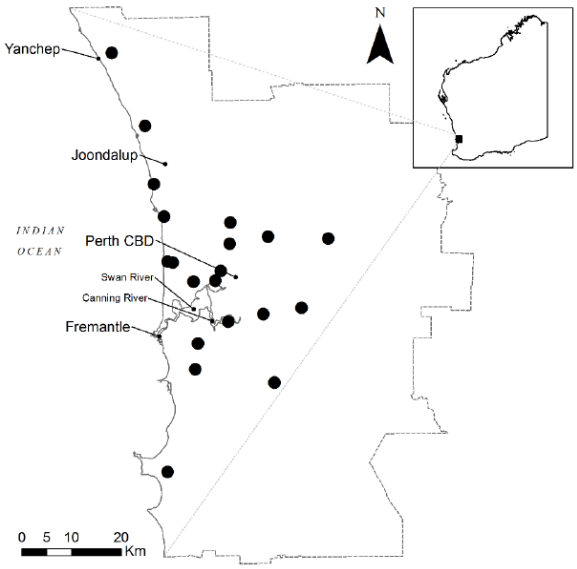

Figure 1 Air sampling sites for PM_{10}, and $PM_{2.5}$ across Perth Metropolitan Area, Western Australia, 2012 (Dotted line indicates boundaries of the Perth Metropolitan Area).

92 *2.3. Sampling and analysis method*

93 Following the ESCAPE protocol, sampling occurred over three seasons, summer, autumn and
94 winter from January 31 to September 5, 2012, at 20 sites, shown in Figure 1. Samples were collected
95 from each site for a two-week period in each season, with a maximum of six sites being monitored
96 concurrently. This resulted in four sampling periods per season. The reference site was operated
97 continuously throughout 2012.

98 Harvard Impactors (MS&T, Air Diagnostics and Engineering Inc. Harrison, ME) were used to
99 collect $PM_{2.5}$ and PM_{10} samples onto Teflon filters (37mm 2um pore size PALL Life Sciences PTFE
100 Membrane). Sampler flow rates were 10 litres per minute ± 5% and recorded before and after the
101 collection. Samples were collected for 15 minutes every 2 hours to prevent overloading so that a 42-hr
102 sample was collected over two weeks. The sampling time and the mean flow rate values were then used
103 to calculate the sample volume. All individual samples at all sites were checked to decide whether the
104 sample were valid to precede subsequent analyses. Valid samples of PM mass are those with total
105 sampling time of at least 67% of 42-hr over fourteen days were included in subsequent analyses. Those
106 considered as invalid samples (n=9) were removed from subsequent analyses.

107 Valid filters were weighed after being placed in the weighing room at a temperature of 23±1ºC
108 and relative humidity of 37±2% for 48 hours. The pre and post weighing was undertaken on a
109 microbalance to obtain mass values as described in the ESCAPE protocol
110 (http://www.escapeproject.eu/manuals/). The mass was then divided by the sample volume to determine
111 the corresponding concentrations of PM_{10} and $PM_{2.5}$. The PM_{coarse} concentrations were obtained by
112 subtracting $PM_{2.5}$ from PM_{10} concentrations.

113 Reflectance was measured on all $PM_{2.5}$ post-weighed samples using a Smokestain reflectometer
114 (EEL43M Smokestein Reflectometer (Diffusio System Ltd)). Reflectance was transformed into
115 absorbance according to the International Standardization Organization method (1993) (Eeftens,
116 Beelen, et al., 2012). The measurements were limited to $PM_{2.5}$ filters as most of elemental carbon has
117 been found in $PM_{2.5}$ fraction (Ref: EEftens). All PM mass and reflectance samples were prepared and
118 analysed at the School of Natural Sciences laboratory, Edith Cowan University.

Filters were then analysed for 48 elements by energy dispersive X-ray fluorescence (ED–XRF). These analyses were undertaken by Cooper Environmental Services, Portland, Oregon, USA. Elemental concentrations were calculated by multiplying the reported mass per area of an element ($\mu g/cm^2$) with the exposed filter area (7.8 cm^2), subtracting the mean field blank and dividing by the individual filter's sample volume (de Hoogh, et al., 2013).

The elements were selected for LUR model development based upon environmental emission characteristics, high percentage of detectability (more than 75% of detected samples), and evidence of impact on human health (de Hoogh, et al., 2013) and attribution of particular sources (Bukowiecki et al., 2010; de Hoogh, et al., 2013; Zhang et al., 2015). Using these criteria, nine elements for Perth were identified, included Cu, Fe, and Zn for anthropogenic and traffic sources; K for biomass burning; S, and V for fossil fuel combustion; Si for soil and dust; Mn and Ni for industrial activities.

2.4. Quality assurance

Blanks and duplicate samples were collected for PM_{10} at the reference site only. Field blanks were collected at 19 time points throughout the annual sampling period. The duplicate samples were used to test the precision of the measurements and were assessed by calculating the absolute value of the difference between one instrument reading and the mean of the two, divided by that mean.

The limit of detection (LOD) was generated as three times the standard deviation of the field blanks. The LOD for PM mass was 0.56 $\mu g/m^3$. The LODs provided by the ED–XRF for V, Si, K, S, Fe, Mn, Zn, and Cu were 2.6×10^{-5} $\mu g/m^3$, 8.4×10^{-4} $\mu g/m^3$, 2.1×10^{-4} $\mu g/m^3$, 1.8×10^{-4} $\mu g/m^3$, 2.9×10^{-4} $\mu g/m^3$, 4.8×10^{-5} $\mu g/m^3$, and 7.5×10^{-5} $\mu g/m^3$, and 8.6×10^{-5} $\mu g/m^3$ respectively.

The measured concentrations of individual samples of PM_{10}, $PM_{2.5}$, PM_{coarse}, and $PM_{2.5}$Abs that were below the LOD were assigned by a random value between 0 and the LOD. Elements were excluded from further evaluation if 25% or more of the samples were below the LOD. Concentration of individual samples below the LOD were not replaced with any other value, resulting in some negative values after the blank corrections (subtract the mean field blank from the sample values) were applied.

146 *2.5. Calculation of annual average concentration*

147 There were six Harvard Impactors available and hence the maximum number of sampling periods
148 per season was four to enable the rotation of instruments to the varying locations. While the
149 measurements were conducted to capture spatial variability, differences in the measured concentrations
150 among the sites may exist because of temporal variations. Therefore, the concentrations at all sites
151 across the three sampling seasons were adjusted using data from the reference site operated every two-
152 week for the whole year 2012. A temporal correction factor for each two-week period within each
153 season was calculated as the difference between the two week-specific-concentrations at the reference
154 site and the annual average concentrations at the reference site. The correction factor was then
155 subtracted from each measurement for the same two-week period to obtain a site specific adjusted
156 concentration for each season. The adjusted seasonal concentrations were averaged to provide adjusted
157 annual average concentrations for each site. These procedures were also applied to all elements.
158 Annual averages for specific elements (Cu, Fe, K, V, S, Si, Zn, Ni, and Mn) were calculated when two
159 or more seasons of data were available per site.

160 A scatterplot between unadjusted average concentrations and the adjusted concentrations
161 including the R^2 and the linear regression equation were generated to evaluate the impact of temporal
162 adjustment (data are not presented here). PM mass and the elements that were in poor agreement
163 between the measured and corrected concentrations data were excluded from subsequent analyses.

164 Further, a correlation matrix of the measured concentrations of PM mass fractions and the
165 elements was generated to investigate the relationships among these pollutants and differences in the
166 source profiles.

167

168 *2.6. Environmental predictor variables*

169 Environmental potential predictors for developing the models were generated to cover a range of
170 air pollutant sources that fit the local characteristics of Perth Metropolitan area and categorised as: land
171 use, population/household density and traffic variables.

172 The land use variables included buildings, industries, presence of water, proximity to water
173 bodies, and open green spaces. The variables of buildings and industries were developed based on the

174 Planning Land Use Classes (PLUCs) from the Valuer General's Office (VGO) of Western Australia
175 Ministry for Planning for the year 2009, while data on water bodies were sourced from the 2009 Western
176 Australia Land Information Authority (Landgate).

177 Industries were grouped into: (1) industrial facilities such as manufacturing/ processing/
178 fabrication, storage and distribution, and service industry; (2) commercial activities such as shops,
179 retails, offices/ business, entertainment/ recreational & cultural; (3) primary and rural activities such as
180 farms & conservation areas; and (4) utilities such electricity, gas, water and waste services, transport,
181 postal, and warehousing. For LUR model development, three definitions of industry variable were used:
182 Industry-1 comprised: (1) industrial, (2) commercial, (3) primary/rural, and (4) utilities groups;
183 Industry-2 comprised: (1) industrial, (3) primary/rural, and (4) utilities; and Industry-3 comprised: (1)
184 industrial, and (2) commercial activities only.

185 The building variable represented all surrounding building types including residential buildings.
186 The water body was characterized by sea, lakes and rivers. The amount of green spaces was represented
187 by Normalized Difference Vegetation Index (NDVI), derived from the Landsat satellite data collected
188 in 2012. The mesh block count from the Australian Bureau Statistics (ABS) for the year 2011
189 (www.abs.gov.au/websitedbs/censushoe.nsf) was used to derive the population and household density
190 data. Both land use and population/household density variables were measured around the sampling
191 sites within circular buffers with radii of 100m, 300m, 500m, 1000m and 5000m to illustrate dispersion
192 patterns of the pollutants being modelled and to capture the spatial variations of pollutant concentrations
193 at both local and regional scales.

194 Traffic variables were measured in radii of 25m, 50m, 100m, 300m, 500m and 1000 m to capture
195 the local impact of potential traffic sources on air pollutant concentrations. Air pollutant concentrations
196 decline exponentially with the distance to road, and decreases to the background levels behind a row of
197 uninterrupted buildings (Batterman et al., 2010). Therefore, traffic variables without buffers such as
198 traffic intensity on the nearest road and distance to major roads were also determined to account the
199 influence of nearby traffic emissions at the monitoring sites.

200 The traffic-related data such as number and type of vehicles on a given road, hierarchy of capacity
201 of roads, length of roads and location of roads were obtained from the Main Roads WA

(www.mainroads.wa.gov.au), collected for the year 2009. Main Roads WA monitors vehicle counts on selected roads using counters and calculates annual average daily traffic (AADT), the annual average number of vehicles travelling in both directions adjusted for season and time of day. Main Road's WA also classifies the roads based upon its capacity including: primary distributor (>15,000 vehicles per day), distributor A (8,001 – 15,000 vehicles per day), distributor B (6,001 - 8,000 vehicles per day), local distributor (3,001 – 6,000 vehicles per day), and access roads (<3,000 vehicles per day). If no counters were present for a particular road class, the traffic counts from nearby roads of the same class in the road hierarchy were used to estimate the annual average traffic counts within each site's buffer. The nearest distance to a road was obtained by measuring the shortest distance from the monitoring site to the nearest road. Heavy duty vehicles included trucks and buses.

In total, 124 potential predictor variables were measured as summarized in the Supplementary Table S1. The generation of environmental predictor variables used in the development of LUR model have previously been described (Dirgawati, et al., 2015). All GIS work was conducted using ArcGIS version 10.2 (ESRI Inc., 2013).

2.7. Model development

LUR models were developed to estimate the annual average concentrations of PM and the corresponding elements (dependent variable) using predictor variables at all monitoring sites. Prior to the models development, standardized predictor variables were generated by subtracting the mean for each predictor from each individual predictor data point and dividing by the SD. Thus, each standardized variable has a mean of zero and a SD of one. The coefficient generated in the final LUR model estimates the change in concentrations associated with a one SD change in the predictor.

Descriptive summaries and scatter plots between the adjusted annual average pollutant concentrations for each site and each predictor variable were used to develop an initial list of suitable predictors and monitoring sites for inclusion in the modelling.

Suitable predictors for model development were those: (1) with 75% values above zero; and (2) where the resultant slope was in the expected direction, as determined a priori (for example, green space is expected to reduce the particulate concentrations, while traffic activity is expected to elevate

the concentrations). Predictors within smaller buffer sizes including industry and water within buffers of 100m and 300m and traffic within a buffer radii of 25m were found to have a considerable number of zero values (>80%), and thus were excluded from the modelling. Among the 124 potential predictors, the total number of suitable predictors to be considered in the models ranged from 44 to 71 predictors. Limiting the number of predictors considered for LUR models reduce the risk of overfitting that occurs when large number predictors are considered to explain concentrations at relatively small number of monitoring sites (Wang et al., 2013; Wang et al., 2012).

The adjusted annual average concentration of PM_{10}, $PM_{2.5}$, and $PM_{2.5}$Abs at each site were checked to detect potential outliers. If the annual average concentration at a particular site was above the 95% percentile, the value was determined as an outlier. This site was then evaluated to decide whether it should be excluded from the modelling, based upon: (1) the stability of the model, i.e. if the parameter estimates of the model (adjusted R^2, direction effect of the predictors) with and without this site differed considerably; (2) the site location was not representing the site specific environmental exposure. As a result, one monitoring site was determined as an outlier, leaving nineteen sites to develop the LUR models.

The modelling procedure was based on manual stepwise selection techniques, following the procedures outlined by the ESCAPE protocol (http://www.escapeproject.eu/manuals/). Briefly, univariate models were run for all suitable predictor variables and the model with the highest adjusted R^2 and the expected slope direction for the predictor was used as the starting model for generation of the multivariate model. The remaining predictor variables were then added one at a time to the starting model. Variables were included if they complied with the following criteria: (1) increased the adjusted $R^2 \geq 1\%$, (2) the coefficient agreed with the predefined direction of effect, and (3) did not change the direction of effect for predictors already in the model.

During the LUR model development for each air pollutant, the univariate analysis might identify multiple starting models with similar adjusted R^2. This resulting multiple appropriate LUR models for that specified pollutant. Akaike Information Criterion (AIC) and Bayesian Information Criterion (BIC)

values for all models were then reviewed for selecting the final LUR model among the alternate models. The selected final LUR model was the model with smaller AIC and BIC values.

Further, to determine how much of the variability of each predictor contributed to the air pollutant concentrations, the R^2 of the nested model was subtracted from the R^2 of the final LUR model.

2.8. Model evaluation

The final models were reviewed for multicollinearity, influential observations, and autocorrelation using diagnostic statistics for multiple regression models. High multicollinearity for model predictors was determined based on Variance Inflation Factor (VIF) values of more than three. The influential observation was examined to ensure the model was not affected by one or more individual sites using Cook's D value above one. The cut-off and graphical plots between the observed and predicted values were also reviewed. Moran's I analysis was performed to investigate the spatial autocorrelation of the residuals of the final LUR models.

The performance of PM mass and the element models was evaluated using the leave-one-out cross validation. Given the relatively small number of monitoring sites and further application of eligible PM mass models to epidemiological studies in Perth, additional evaluation of PM mass models was conducted to investigate the true predictive ability of the models and stability of the predictors included in the models. The methods used, were hold-out validation and cross-holdout validation.

In the leave-one-out cross-validation method, evaluation models were developed using all but one of the measurement sites and the predicted concentrations were compared with the measured concentrations at the omitted site (Refaeilzadeh et al., 2009). The adjusted R^2 and the root mean square error (RMSE) between the predicted and observed concentrations were then calculated and compared with the original model and the corresponding standard deviation as measures of model performance (de Hoogh, et al., 2013; Eeftens, et al., 2012). Lower RMSE values typically indicate more stable models (Hoek, et al., 2008; Mölter et al., 2010).

The hold-out validation used training and test sets in the evaluation procedure. The total number of suitable monitoring sites for model development (nineteen sites) was equally divided into training and test sets. Ten sites were selected as training dataset for modelling, and the remaining nine sites were

used for prediction outsides the training sites. The sites were randomly selected based on the strata of site types to ensure proportionate distribution of the street, urban background and regional background sites. The selection was repeated nine times to give 10 sets in total. The predictors variables included in the model for all sites were used to develop the training models. Refitting the same predictor in smaller subsets may have changed the direction of effect of predictors in the training model. The same criteria of predictors included in the model was used consistent to obtain the true predictive ability of the training models. Further, the squared Pearson-correlation coefficient, which is equivalent to R^2 between the measured and the predicted concentrations at the test sites was calculated and the stability of LUR model's structure was tested to measure the performance of these models at smaller subsets.

In cross–hold out validation method, one individual site was successively left out, leaving eighteen sites for developing an evaluation model (Wang et al., 2016). The process was repeated nineteen times to obtain nineteen evaluation models. Each of the evaluation model was then used to predict the concentrations at the site that was not included in developing the model. This process was conducted for all evaluation models, resulting the predicted concentrations across nineteen monitoring sites. The true hold out R^2 was calculated as the R^2 between the measured and the predicted concentrations at these nineteen external sites. The R^2 and the true hold out R^2 were then compared with the full sites LUR model. The stability of model's structure was also tested by comparing the predictors included in the full sites model with those included in the training and evaluation models. All statistical analyses were undertaken using the statistical software STATA version 12.1 and 13.1 (StataCorp LP, Texas, USA).

3. Results

3.1. Descriptive statistics of measured air pollutant concentrations

Fifty-one valid samples (85%) were collected from nineteen sites during summer, spring and autumn, 2012. For the elements, the precision of the laboratory method had less than 10% variability demonstrating that the methods were reproducible. Some of elements ($PM_{2.5}$ S, $PM_{2.5}$ Cu, PM_{10} Ni and $PM_{2.5}$ Ni) were unsuitable for LUR modelling. The temporal corrections resulted in poor agreement between the measured and corrected data for S and Cu in $PM_{2.5}$. Both PM_{10} Ni and $PM_{2.5}$ Ni were also

excluded from the modelling as more than 25% of the elements' samples had concentrations below the LOD.

Table 1 provides a summary of the descriptive statistics of the temporally adjusted annual concentrations for all PM and the selected elements for PM_{10} and $PM_{2.5}$ for the remaining nineteen monitoring sites. Summary statistics of the unadjusted measured concentration for the 48 elements of $PM_{2.5}$ and PM_{10} are presented in Supplementary Table S2 and Table S3, respectively.

Table 1

Descriptive Summary of temporally adjusted annual average concentrations for PM_{10}, PMcoarse, $PM_{2.5}$, $PM_{2.5}$Abs, PM_{10} elements and $PM_{2.5}$ elements (N = 19)

Pollutant	Mean	Median	SD	Min	Max
PM mass ($\mu g/m^3$):					
PM_{10}	17.1	16.4	5.0	8.9	30.3
PM_{coarse}	12.4	10.8	5.5	4.2	26.4
$PM_{2.5}$	4.7	4.5	1.6	1.5	7.8
PM absorbance ($10^{-5}m^{-1}$):					
$PM_{2.5}$A	0.7	0.7	0.3	0.2	1.5
PM_{10} elements (ng/m^3):					
Si	3.9×10^{-1}	3.6×10^{-1}	2.4×10^{-1}	1.3×10^{-3}	7.9×10^{-1}
S	4.9×10^{-1}	4.5×10^{-1}	1.3×10^{-1}	3.4×10^{-1}	7.8×10^{-1}
K	1.8×10^{-1}	1.9×10^{-1}	4.8×10^{-2}	9.3×10^{-2}	2.8×10^{-1}
V	1.2×10^{-3}	1.1×10^{-3}	5.0×10^{-4}	6.0×10^{-4}	2.4×10^{-3}
Mn	5.5×10^{-3}	5.1×10^{-1}	3.6×10^{-3}	1.8×10^{-3}	1.7×10^{-2}
Fe	2.9×10^{-1}	2.5×10^{-1}	1.7×10^{-1}	6.4×10^{-2}	6.8×10^{-1}
Cu	6.2×10^{-3}	4.6×10^{-3}	4.4×10^{-3}	1.0×10^{-3}	1.7×10^{-2}
Zn	9.3×10^{-3}	7.5×10^{-3}	5.8×10^{-3}	7.0×10^{-4}	2.3×10^{-2}
$PM_{2.5}$ elements (ng/m^3):					
Si	8.8×10^{-2}	8.8×10^{-2}	5.1×10^{-2}	5.6×10^{-2}	2.4×10^{-1}
K	7.9×10^{-2}	7.9×10^{-2}	2.0×10^{-2}	5.0×10^{-2}	1.2×10^{-1}
V	7.0×10^{-4}	7.0×10^{-4}	3.0×10^{-4}	3.0×10^{-4}	1.2×10^{-3}
Mn	2.4×10^{-3}	2.4×10^{-3}	3.7×10^{-3}	6.0×10^{-4}	1.6×10^{-2}
Fe	8.1×10^{-2}	8.1×10^{-2}	4.6×10^{-2}	2.4×10^{-2}	1.8×10^{-1}
Zn	6.5×10^{-3}	6.5×10^{-3}	5.9×10^{-3}	1.8×10^{-3}	2.9×10^{-2}

The relationships between the annual averages of PM_{10}, PM_{coarse}, $PM_{2.5}$, and $PM_{2.5}$Abs and the selected elements are presented in Supplementary Table S4. A high correlation was observed for the association between PM_{10} and PM_{coarse} ($r = 0.95$), moderate correlation between $PM_{2.5}$ and $PM_{2.5}$ Abs ($r = 0.57$), and poor correlation between the PM_{10} and $PM_{2.5}$ ($r = -0.13$). For the PM_{10} elements, the highest correlation was observed between PM_{10} and S ($r > 0.5$), while poor correlations were observed between PM_{10} and the remaining elements. The $PM_{2.5}$ was moderately correlated with K, V, Mn, Zn, and Fe with $0.45 < r < 0.53$. $PM_{2.5}$Abs was correlated with $PM_{2.5}$ Fe, –Mn, and –Zn; PM_{10} S and –V.

3.2. *LUR Models for $PM_{2.5}$, PM_{10}, $PM_{2.5A}$ and the associated elements*

LUR models were developed to predict the concentrations of $PM_{2.5}$, PM_{10} and their respective elements as well as $PM_{2.5}$Abs using 19 measurements sites across Perth. The LUR models for PM_{coarse}, Cu and PM_{10} Zn had poor predictive ability (adjusted $R^2 < 20\%$) and diagnostics, and are not presented. Figure 2 illustrates the proportion of the spatial variability in the measured concentrations of $PM_{2.5}$, PM_{10}, and $PM_{2.5}$Abs and the selected elements explained by the LUR models.

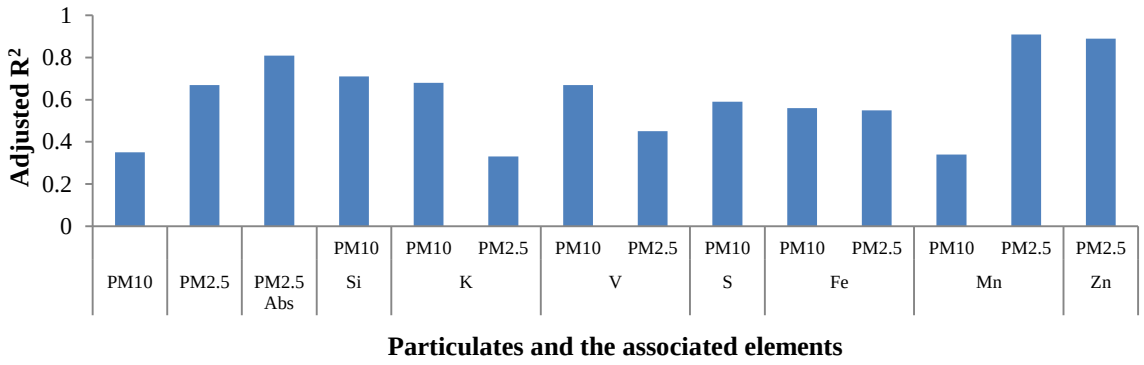

Figure 2 Proportion of the spatial variability in the measured concentrations of $PM_{2.5}$, PM_{10}, and $PM_{2.5}$Abs and selected elements explained by the LUR models

The $PM_{2.5}$ and $PM_{2.5}$Abs models had acceptable predictive ability with adjusted R^2 above 65%, while the PM_{10} model had much lower predictive ability. The LUR models captured greater proportion of the spatial variability in the measured crustal elements (Si and K) and S in PM_{10} fraction than that in $PM_{2.5}$. The $PM_{2.5}$ elemental models explained highest proportions in the spatial variability of industrial related source element (Mn) and vehicle source element (Zn). The variability of industrial/fuel oil combustion-element (V) in PM_{10} was better captured than that in the $PM_{2.5}$. Similar proportion was observed in the spatial variability of non-tailpipe vehicle source (Fe) in both PM_{10} and $PM_{2.5}$.

Complete description of the structure of the LUR models is presented in Supplementary Table S5, the summary of predictor variables captured by each LUR model and the corresponding unique contributions is illustrated in Table 2. For PM_{10}, the most important predictor was green spaces. In contrast, heavy vehicle traffic load was found to be the strongest predictor for $PM_{2.5}$Abs. Traffic

352 variables were observed to be the important contributors for PM_{10} and $PM_{2.5}$ elements in Perth, except
353 for $PM_{2.5}$ V which had distance to coast as the predominant predictors. Open green spaces explained
354 more of the variability in the PM_{10} elements than for $PM_{2.5}$ elements, population density was more
355 important for $PM_{2.5}$ elements than for PM_{10} elements.

Table 2 Predictor variables and their unique contributions included in the LUR models for PM_{10}, $PM_{2.5}$, $PM_{2.5}$ Abs and their associated elements

Predictors Variables	PM_{10}	$PM_{2.5}$	$PM_{2.5}$ Abs	PM_{10} elements					$PM_{2.5}$ elements					
				Si	K	V	S	Fe	Mn	Fe	Mn	V	Zn	K
Open green spaces	5000m (33%)	1000m (13%)		500m (9%)	500m (8%)	5000m (3%)	5000m (9%)	5000m (9%)	1000m (16%)		1000m (2%)			
Population density		100m (8%)	500m (7%)							1000m (15%)				
Housing density								100m (5%)	100m (8%)					
Building	100m (4%)													
Industry			Industry-3[a] 5000 m (11%)								Industry-2[b] 1000m (2%)	Industry-3[a] 1000m (8%)		
Water body		5000m (13%)	1000m (1%)							5000m (9.7%)	5000m (13.8%)		5000m (7%)	5000m (10%)
Distance to water body		Coast (5%)	Coast (1%)			Coast (25%)						Coast(28%) River (1%)		River (5%)
Traffic intensity on nearest road		Dist A[c] (8%)			PDist (7%)					Dist A[c] (14%)				Dist A[c] (6%)
Heavy traffic intensity on nearest road	Any roads (27%)													
Traffic load			Any roads 500m (25%)		Dist B[e] 500m (6.6%)	Any roads 300m (45 %)	PDist 500m (45%)	Dist B[e] 500 m (24%)	Dist B[e] 500m (13%)	PDist[d] 1000m (13%)	Any roads 1000m (77%)		Any roads 1000m (53%)	
Heavy vehicles traffic load		Dist B[e] 500m (3%)					Dist B 1000m (9%)							
Road length				Dist B[e] 500m (15.2%)									PDist[d] 1000m (3%)	Dist B[e] 300m (20%)
Distance to road				PDist[d] (15%)	Dist B[e] (1%) PDist[d] (2%)			PDist[d] (4%)						

a) Industry-3: Industry variable – definition 3; comprised of industrial and commercial
b) Industry-2: Industry variable – definition 2; comprised of industrial, primary/rural and utilities
c) DistA: Distributor A road (8,001 – 15,000 vehicles per day)
d) PDist : Primary Distributor road (>15,000 vehicles per day)
e) DistB: Distributor B road (6,001 - 8,000 vehicles per day)

The model diagnostics for $PM_{2.5}$ and $PM_{2.5}$Abs using the variance inflation factors (VIF), Cook's D and Moran's I were acceptable (Supplementary Table S6). The VIF and the Cook's D values suggest the models do not violate the collinearity and influential observation assumptions. The Moran's I for all PM mass fractions and reflectance ranged from 0.083 to 0.970 with a p-value > 0.05, representing no spatial autocorrelation of the residuals. The elemental models did not violate the general assumptions for the development of the LUR models except for PM_{10} Fe, PM_{10} Mn, and $PM_{2.5}$ V. Influential observation measured it. Based upon evaluation during the model development, it was observed that the parameter estimates of the models for PM_{10} Fe, PM_{10} Mn, and $PM_{2.5}$ V with and without this influential site changed considerably. Therefore, the corresponding site was excluded from modelling for these elements. Among the LUR models for the PM_{10} and $PM_{2.5}$ elements, we found no spatial autocorrelation measured by Moran's I, except for $PM_{2.5}$ Zn.

Table 3 shows the results of the model evaluation procedures, including the leave-one-out cross validation, hold out validation and cross-hold out validation. The differences in the adjusted R^2 between the final models and the leave one out cross validation results for $PM_{2.5}$ was 17% and $PM_{2.5}$Abs was 15%, indicating the spatial predictive ability of both models are relatively good.

Table 3

Summary of $PM_{2.5}$ and $PM_{2.5}$A models evaluation average R^2 Validation results of LUR models for $PM_{2.5}$ and $PM_{2.5}$Absorbance

Air pollutant	Leave one out cross validation (n = 19)	Hold-out validation		Cross-hold-out validation	
		Training sets	Test sets	Training sets	Test sets
		(n = 10)	(n = 9)	(n = 18)	(n = 19)
$PM_{2.5}$	0.50	0.74	0.47	0.69	0.14
$PM_{2.5}$ Abs	0.67	0.74	0.73	0.82	0.61

Plots of the measured and the predicted concentrations for both pollutants are shown in supplementary Figure S1, indicating an agreement between the measured and predicted concentrations.

The hold-out and cross-hold-out validation procedures results also indicated the stability of our $PM_{2.5}$ and $PM_{2.5}$Abs LUR models. The top four predictors in the final $PM_{2.5}$ model such as surface area

of water body, open green spaces, population density, and traffic intensities were dominant in the $PM_{2.5}$ evaluation models (Supplementary Table S7). For $PM_{2.5}$Abs evaluation models, the predictor matched. Traffic load, industry and commercial area, population density, and proximity to coast were consistently captured by all evaluation models (Supplementary Table S8).

4. Discussion

4.1. Air pollutant concentrations

The annual mean concentrations of PM_{10} (17.1 μg/m^3), PM_{coarse} (12.4 μg/m^3), and $PM_{2.5}$ (4.7 μg/m^3) across Perth were below the National Environment Protection Measures (NEPM) for ambient air quality (20 μg/m^3 for PM_{10} and 8 μg/m^3 for $PM_{2.5}$) (NEPC, 1998) and the WHO annual mean air quality guidelines (20 μg/m^3 for PM_{10} and 10 μg/m^3 for $PM_{2.5}$) (WHO, 2005). The mean concentration of $PM_{2.5}$Abs was 0.67 x 10^{-5}m^{-1}. There is no corresponding guideline for this pollutant or for any of the PM elements. The high correlation between PM_{10} and PM_{coarse} reflects the fact that PM_{10} and PM_{coarse} are emitted from similar sources such as non-exhaust emissions and fugitive coarse dust (Keuken et al., 2013), while moderate correlation between $PM_{2.5}$ and $PM_{2.5}$ Abs, possibly reflecting that their concentrations are influenced by complex interaction between local meteorological conditions and the main source of these pollutants such as vehicles emissions and industrial activities (Keuken, et al., 2013).

PM_{10} and PM_{coarse} annual average concentrations were at the lower end of those European cities participating in the ESCAPE study, which ranged from 14.8–43.1 μg/m^3 for PM_{10}; and 6.0–23.6 μg/m^3 for PM_{coarse}. Likewise, the $PM_{2.5}$ concentrations and the $PM_{2.5}$Abs concentrations were both lower than those of the ESCAPE cities, which were 8.3–29.3 μg/m^3 for $PM_{2.5}$; and 0.8 – 3.0 x 10^{-5}m^{-1} for $PM_{2.5}$Abs) (Eeftens, et al., 2012). The median concentrations of the PM elements were typically lower than the ESCAPE cities (de Hoogh, et al., 2013) and the Calgary study (Zhang, et al., 2015). Similarly to the PM mass and the reflectance measures, the concentrations of all PM mass elements in Perth were lower than those measured in European and North America cities (Ross 2007). Possible reasons for these differences between the studies include: (1) air pollutant concentrations in Perth tend to disperse across the airshed due to its flat topography and strong ocean breezes (Yimin, et al., 2003); (2) the traffic

intensities on major roads in Perth (<60,000 vehicles per day) are lower than the cities in the ESCAPE study, which are typically above 100,000 vehicles per day (EEA, 2011); and (3) road networks in the ESCAPE cities are denser than in Perth (EEA, 2011). The $PM_{2.5}/PM_{10}$ ratio is low (0.3), indicating that sea salt and wind blown dust are possibly responsible for the low $PM_{2.5}/PM_{10}$ ratio relative to the European cities (Eeftens, Tsai, et al., 2012).

4.2. Characterisation of surrounding environment and land use

LUR models identify surrounding environmental and land use characteristics that may help explain the variability in pollutant concentrations (Hoek, et al., 2008). Traffic intensity of heavy vehicles in close proximity to the monitoring site was associated with increases in measured PM_{10} concentrations, contributing approximately 27% in the explained variability. More than 90% of the heavy vehicle fleets in Australia have diesel engines. Heavy vehicles account for around 25% of all road transport fuel consumed in Australia, thus they potentially contribute more to traffic exhaust (ABS, 2014). The LUR models for PM_{10} in European cities such as Manchester, London and Ruhr Area, have also included heavy vehicle traffic intensity in their models (Eeftens, et al., 2012).

Traffic variables were included in the all elements models of PM_{10} and $PM_{2.5}$ fraction. Keuken et al., (2013) reported that Zn in particulates found in nearby roads originate from tire ware as its use as galvanised materials in rubber production and re-suspended road dust. Fe was found to be emitted from metal wear in the exhaust systems walls as flakes of iron and it is considered as a good marker for brake wear emissions (Keuken, et al., 2013). Fe can also be emitted from tailpipes, formed within the engine due to gas to particle conversion processes of the ferrocene, an agent to raise octane level of diesel and gasoline fuel (Srimuruganandam et al., 2011). K is related to emissions from the ash fractions of diesel exhausts (Srimuruganandam et al., 2012), and Mn is emitted from brake lining dust (Grigoratos et al., 2015; Srimuruganandam, et al., 2011). Traffic-related emissions are not specific sources for V and S in the PM_{10} fraction, as these elements are also emitted from fossil fuel combustion in industries (Murillo et al., 2013; Srimuruganandam, et al., 2012). Most of the models variables appear attributable to the resuspended road dust.

441 Asphalt roads comprise 95% mineral grains (Ca, Al, Si, Na, K) and 5% filler and binding
442 materials (Srimuruganandam, et al., 2012), suggesting Si may arise from road wear of asphalt roadways.
443 Compared with the other elements' models that included many traffic intensity or traffic load predictors,
444 the LUR model for Si uniquely included distance to major road and length of major road. Therefore,
445 the LUR model indicates the contribution of the re-suspended dust or paved road dusts to the
446 concentrations of crustal elements in PM_{10}.

447 The models for $PM_{2.5}$, and $PM_{2.5}$Abs showed that population density within the smallest buffer
448 sizes (100m and 500m) contributed to the increase in their concentrations. Population and housing
449 density are associated with various anthropogenic sources including residential activities such the use
450 of wood stoves and heaters, as well as tailpipe and non-tailpipe emissions of traffic servicing this area,
451 as reported previously for other areas (Eeftens, et al., 2012; Urman et al., 2014). Wood stoves and
452 heaters are top emission sources of $PM_{2.5}$ in Australia as documented by the National Pollutant
453 Inventory (NPI, 2014).

454 Population and housing density were also included in the Fe – $PM_{2.5}$ and Mn – PM_{10} LUR
455 models, respectively. Fe is related to the dominant vehicle emissions, brake, and road wear, while Mn
456 is mainly attributed to re-suspension of road dust reflecting the contribution of traffic servicing
457 residential area. The results are as expected, given higher number of street sites relative to the urban
458 background and regional sites.

459 Our LUR models identify industrial activities (manufacturing, processing and fabrication) and
460 commercials (shops, retails, offices, entertainment, recreational & cultural activities) located within
461 5000m were modest predictors for $PM_{2.5}$Abs and $PM_{2.5}$ V, and minor predictors for $PM_{2.5}$ Mn. Such
462 results support evidence that traffic variables are the major air pollution source for airborne $PM_{2.5}$ Abs
463 and $PM_{2.5}$ in Perth airshed. Refinery/residual oil combustion for industrial activities has been shown to
464 be indicator of V, and steel making has been the primary contributor of Mn (Chow et al., 2002). Zhang
465 et al., (2015) also identified alternative predictors for Mn including industrial facilities. De Hoogh et
466 al., (2013) identified sources of V to be industrial and fuel oil combustion-related. Both could also be
467 the sources in Perth, given that oil combustion is one of the major air pollution sources in industrial area
468 in Perth (NPI, 2013).

469 The influence of the proportion of green space and water in an area resulted in the reduction of
470 all of the PM and elemental concentrations within a range of buffer sizes. The LUR models for PM_{10}
471 and all the associated elements characterised green spaces predominantly within buffer sizes of 500m,
472 1000m and 5000m. Such associations between greenness and PM mass have also been found in other
473 studies, suggesting plants and trees in open spaces play an important role in improving air quality and
474 reducing the concentrations of particulates (Eeftens, et al., 2012; McDonald et al., 2007). The observed
475 association between the greenness and PM mass concentrations may related to the proportion of green
476 spaces that account for almost 16% of the total area of Perth (ABS, 2012) and possibly lower traffic
477 intensities in open green and water areas. Our PM models are consistent with LUR models for ESCAPE
478 and for North American cities that included urban green and natural land use within large buffer sizes
479 (1000 or 5000 m) (de Hoogh, et al., 2013; Eeftens, et al., 2012; Ross et al., 2007).
480 Off shore shipping are known to be indicators of V (Chow, et al., 2002), thus shipping activity at
481 ports may one of the emission sources for V. Although the variable of ports did not considered in model
482 development because there were 90% zero values around the monitoring sites, the V model included
483 distance to coast, which contributed 25% and 28% to the increase in the concentrations of V in PM_{10}
484 and $PM_{2.5}$ respectively. The LUR models, therefore, indicate the importance of the emission oil
485 combustion of ship movement activities on the nearby coast to the V concentrations.
486
487 *4.3 Spatial variability of air pollutant concentrations*
488 We observed differences in the performance of each LUR model for explaining the spatial
489 variability of particulates and their elements in Perth. The majority of the LUR models were able to
490 explain more than 50% of the pollutant's spatial variability. There were some exceptions which
491 included PM_{10}, PM_{10} Mn, $PM_{2.5}$ V, and $PM_{2.5}$ K which only explained between 30 – 45% of the spatial
492 variability. The absence of specific predictor data for those species may limit the models' performance.
493 For example, fugitive dust to explain the PM_{10} concentrations, refinery/residual oil industries and off
494 shore shipping as the sources of V, steel industries as the primary contributor of Mn, and wood burning
495 for K were not incorporated as potential environmental predictors in the modelling process. Consistent
496 with previous studies in Europe and North America that suggest small variations in the measured

element concentrations, lack of specific predictors and poor precision of measurements in areas with low concentrations are the main reasons for a poor R^2-model and R^2-LOOCV (de Hoogh, et al., 2013; Eeftens, et al., 2012; Ross, et al., 2007)

The model R^2 of $PM_{2.5}$ was comparable with those in the ESCAPE study areas which ranged from 49% to 89% and were slightly higher than those reported in North America (Ross, et al., 2007). The predictive power of the $PM_{2.5}$Abs model was also similar to the ESCAPE results (between 56% and 95%), while for the PM_{10} model, the predictive ability was lower than those in the ESCAPE (50%-90%) (Eeftens, et al., 2012). The PM_{10} elemental models explained between 34% and 71% of the variability with only Fe, Zn and Cu performing poorly in comparison to the average R^2 for the ESCAPE models across all cities. The $PM_{2.5}$ elemental models explained 36% – 90% of the concentration variability. Our Fe and K models performed less well in comparison with ESCAPE but our V and Zn models were able to explain a larger proportion of the variability than ESCAPE (de hoogh et al., 2013).

The stability of our LUR models is also indicated by the results of the evaluation procedures. The differences in adjusted R^2 values between the model and the LOOCV were 17% for $PM_{2.5}$, 9% for PM_{10}, and 15% for $PM_{2.5}$Abs. For PM_{10}, $PM_{2.5}$ and $PM_{2.5}$Abs, the RMSE of LOOCV and models were smaller than the corresponding standard deviation. For both PM_{10} and $PM_{2.5}$ elements, the differences between the adjusted R^2-model and -LOOCV were within 15% suggesting the stability of the models, except for PM_{10} Mn (19%) and $PM_{2.5}$ K (26%). RMSE values that were obtained from LOOCV were found to be higher than those RMSE models. However, when the RMSE were assessed relative to the range of measured air pollutant concentrations, we found small differences between the LOOCV and the models i.e. 9% for $PM_{2.5}$ and 6% for $PM_{2.5}$Abs, demonstrating the stable models.

From the hold out validation method, the $PM_{2.5}$ training models have larger average R^2 compared with the full-sites model, indicating over fitting. The results from the cross hold out validation method showed that the adjusted R^2 of $PM_{2.5}$ models based on the full sites and 18 sites were similar. However, the true hold-out R^2 underestimated the models' predictive ability at the site that was not used for developing the $PM_{2.5}$ models. For $PM_{2.5}$Abs, the average R^2 of training models from the hold out validation and evaluation models from the cross hold out validation were similar to the adjusted R^2 of the full sites models. The training and evaluation models estimated the true predictive ability of the

525 models at the external sites in the acceptable range, consistent with the LOOCV results which
526 demonstrate the stability of the $PM_{2.5}$Abs model.
527
528 *4.4. Limitations and generalizability*
529 Our LUR models used a smaller number of sites compared with previous studies that had suggested
530 a larger number of monitoring sites (>80 sites) were required (Basagaña et al., 2012). However, the
531 number of sites is consistent with the ESCAPE protocol. Overfitting may have occurred because of a
532 large number of predictors (124 predictors) that were included in developing the models relative to the
533 small number of sites (19 sites) (Basagaña, et al., 2012; Wang, et al., 2012). Despite the relatively small
534 number of monitoring sites, this study has developed LUR models at an acceptable performance level
535 with lower risk of overfitting as we implemented selection procedures for suitable predictors prior to
536 modelling.
537 The availability of predictor data may also limit our model generalizability. Data on traffic
538 intensities on relevant roads were obtained by using the available traffic counts. In addition, the
539 concentrations of air pollutants were measured in 2012, while the predictor data such as household
540 density were sourced from 2011 databases and industrial areas were obtained from 2009 databases.
541 Data on predictors that are collected from the same year as when the monitoring occurred can optimize
542 the predictive ability of models and enhance generalizability as the quality of predictor data at specific
543 time periods can affect the results of LUR models (Hoek, et al., 2008). However, the pattern of those
544 predictors in Perth was relatively consistent across the five-year period, suggesting that the final LUR
545 models are generally representative for capturing the spatial variability in air pollutants throughout the
546 year 2012.
547
548 **5. Conclusion**
549 Despite the relatively low concentrations, LUR modelling for $PM_{2.5}$ and $PM_{2.5}$Abs and the
550 elements of PM_{10} and $PM_{2.5}$ is possible for such locations. The LUR models characterised the local
551 traffic related air pollution as the predominant source to explain the spatial variability of airborne
552 particulate matters and the associated elements in Metropolitan Perth. This study represents one of the

few LUR studies investigating PM elements and it will assist researchers in assessing the health impacts of the components of PM as well as PM size fractions.

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