

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

Mila Dirgawati^a, Jane S. Heyworth^a, Amanda J. Wheeler^{a,b}, Kieran A. McCaul^c, David Blake^b, Jonathon Boeyen^b, Martin Cope^d, Bu Beng Yeap^e, Mark Nieuwenhuijsen^f, Bert Brunekreef^g, Andrea Hinwood^b

^aThe University of Western Australia, 35 Stirling Hwy, Crawley, WA 6009, Australia

^bEdith Cowan University, 270 Joondalup Drive, Joondalup, WA 6027, Australia

^cWA Centre for Health & Ageing, The University of Western Australia, Level 6, Ainslie House, 48 Murray St, Perth, WA 6000, Australia

^dCSIRO Marine and Atmospheric Research, PMB1 Aspendale, Vic 3195, Australia

^eSchool of Medicine and Pharmacology, The University of Western Australia, Perth, WA, and Department of Endocrinology and Diabetes, Fremantle and Fiona Stanley Hospitals, Perth, WA, Australia

^fCenter for Research in Environmental Epidemiology (CREAL), Barcelona, Spain

^gInstitute for Risk Assessment Sciences, Utrecht University, P.O. Box 80178, 3508 TD Utrecht, The Netherlands

Abstract

Perth, Western Australia represents an area where pollutant concentrations are considered low compared with international locations. Land Use Regression (LUR) models for PM₁₀, 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 PM₁₀ 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 PM₁₀ 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 PM₁₀ elements than for PM_{2.5} elements, and population density was more important for PM_{2.5} elements than for PM₁₀ elements. The PM_{2.5} and PM_{2.5}Abs LUR models explained 67% and 82% of the variance, respectively, but the PM₁₀ 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. PM₁₀ 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 PM₁₀, 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 10 μ m (PM₁₀) and 2.5 μ m (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).

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

51 There is some evidence that the composition of PM may also be important in determining health
52 effects (Eeftens et al., 2014; Stanek et al., 2011). PM₁₀-associated zinc (Zn) has been associated with
53 the risk of pneumonia among children in seven birth cohort studies in Europe (Fuertes et al., 2014).
54 PM_{2.5}-associated nickel (Ni) and vanadium (V) have also been associated with daily mortality in time-
55 series studies in two northern American cities (Zhou et al., 2010). Small decreases in lung function
56 among young children have also been related with exposure to other PM-associated elements such as
57 Cu, Fe, K, Si, sulfur (S) and Zn (Eeftens, et al., 2014).

58 There are relatively little data available on the intra-urban speciation of PM₁₀ and PM_{2.5} for
59 estimating exposures in large population-based health studies in Australia. Land Use Regression (LUR)
60 models have been used to predict small-scale spatial variations in exposure to air pollutants including
61 species of PM, within cities in other location (Gulliver et al., 2011; Hoek et al., 2008; Zou et al., 2009).
62 The recent European Study of Cohorts for Air Pollution Effects (ESCAPE) developed PM speciated
63 LUR models for eight elements in 15 countries. Most of those LUR models explained a large fraction
64 of the spatial variation within the study area with R² ranging from 50% and 79% (de Hoogh et al., 2013)
65 and were used to investigate the associations with a range of health outcomes (Eeftens, et al., 2014).

66 This paper describes the development of LUR models for PM₁₀, PM_{2.5}, PM_{2.5}Absorbance (PM_{2.5}Abs),
67 and PM-associated elements (Cu, Fe, K, V, S, Si, Zn, Ni, and Mn) for the Perth metropolitan area,
68 Western Australia, an area with lower air pollutant concentrations compared with most European and
69 North American cities. The models were then used to assign exposures to PM and PM elements for a
70 cohort of older men in Perth, the Health in Men Study (HIMS) (Norman et al., 2009).

71

72 **2. Methods**

73 *2.1. Area of study*

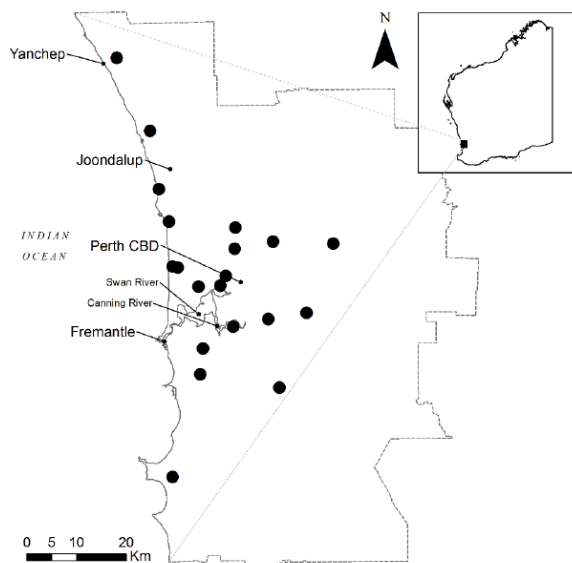
74 The study was conducted in the Perth Metropolitan Area which is the capital of Western
75 Australia. It is located on the Indian Ocean with the Darling Ranges to the East, and with two main
76 waterways, the Swan and Canning rivers. Perth metropolitan area has an area of 6,418 km² and the
77 population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

79

80 *2.2. Sampling sites selection*

81 The monitoring site selection has been described elsewhere (Dirgawati et al., 2015). Twenty
82 sites were selected to measure PM_{2.5} and PM₁₀, comprising two regional background sites; eight urban
83 background sites; and ten street sites based on the criteria describe in the protocol for the ESCAPE
84 study (<http://www.escapeproject.eu/manuals/>). One of the regional background sites was co-located at
85 a monitoring station operated by the Western Australian Department of Environment Regulation for
86 comparison with standard government monitoring. A reference site was also operated continuously
87 throughout the sampling period to adjust for any temporal variability of particle concentrations between
88 seasons. Figure 1 shows a map of the sampling sites.

89



90 **Figure 1** Air sampling sites for PM₁₀, and PM_{2.5} across Perth Metropolitan Area, Western Australia, 2012 (Dotted
91 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 PM₁₀ samples onto Teflon filters (37mm 2µm pore size PALL Life Sciences PTFE
100 Membrane). Sampler flow rates were 10 litres per minute \pm 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 \pm 1°C
108 and relative humidity of 37 \pm 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 PM₁₀ and PM_{2.5}. The PM_{coarse} concentrations were obtained by
112 subtracting PM_{2.5} from PM₁₀ 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.

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

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

130

131 2.4. *Quality assurance*

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

136 The limit of detection (LOD) was generated as three times the standard deviation of the field
137 blanks. The LOD for PM mass was $0.56 \mu\text{g}/\text{m}^3$. The LODs provided by the ED–XRF for V, Si, K, S,
138 Fe, Mn, Zn, and Cu were $2.6 \times 10^{-5} \mu\text{g}/\text{m}^3$, $8.4 \times 10^{-4} \mu\text{g}/\text{m}^3$, $2.1 \times 10^{-4} \mu\text{g}/\text{m}^3$, $1.8 \times 10^{-4} \mu\text{g}/\text{m}^3$, 2.9×10^{-4}
139 $\mu\text{g}/\text{m}^3$, $4.8 \times 10^{-5} \mu\text{g}/\text{m}^3$, and $7.5 \times 10^{-5} \mu\text{g}/\text{m}^3$, and $8.6 \times 10^{-5} \mu\text{g}/\text{m}^3$ respectively.

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

145

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.

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

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

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

216

217 2.7. Model development

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

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

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

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

237 The adjusted annual average concentration of PM₁₀, PM_{2.5}, and PM_{2.5}Abs at each site were
238 checked to detect potential outliers. If the annual average concentration at a particular site was above
239 the 95% percentile, the value was determined as an outlier. This site was then evaluated to decide
240 whether it should be excluded from the modelling, based upon: (1) the stability of the model, i.e. if the
241 parameter estimates of the model (adjusted R², direction effect of the predictors) with and without this
242 site differed considerably; (2) the site location was not representing the site specific environmental
243 exposure. As a result, one monitoring site was determined as an outlier, leaving nineteen sites to
244 develop the LUR models.

245 The modelling procedure was based on manual stepwise selection techniques, following the
246 procedures outlined by the ESCAPE protocol (<http://www.escapeproject.eu/manuals/>). Briefly,
247 univariate models were run for all suitable predictor variables and the model with the highest adjusted
248 R² and the expected slope direction for the predictor was used as the starting model for generation of
249 the multivariate model. The remaining predictor variables were then added one at a time to the starting
250 model. Variables were included if they complied with the following criteria: (1) increased the adjusted
251 R² ≥1%, (2) the coefficient agreed with the predefined direction of effect, and (3) did not change the
252 direction of effect for predictors already in the model.

253 During the LUR model development for each air pollutant, the univariate analysis might identify
254 multiple starting models with similar adjusted R². This resulting multiple appropriate LUR models for
255 that specified pollutant. Akaike Information Criterion (AIC) and Bayesian Information Criterion (BIC)

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

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

260

261 2.8. *Model evaluation*

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

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

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

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

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

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

304

305 **3. Results**

306 *3.1. Descriptive statistics of measured air pollutant concentrations*

307 Fifty-one valid samples (85%) were collected from nineteen sites during summer, spring and
308 autumn, 2012. For the elements, the precision of the laboratory method had less than 10% variability
309 demonstrating that the methods were reproducible. Some of the elements ($PM_{2.5}$ S, $PM_{2.5}$ Cu, PM_{10} Ni and
310 $PM_{2.5}$ Ni) were unsuitable for LUR modelling. The temporal corrections resulted in poor agreement
311 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

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

314 Table 1 provides a summary of the descriptive statistics of the temporally adjusted annual
 315 concentrations for all PM and the selected elements for PM₁₀ and PM_{2.5} for the remaining nineteen
 316 monitoring sites. Summary statistics of the unadjusted measured concentration for the 48 elements of
 317 PM_{2.5} and PM₁₀ are presented in Supplementary Table S2 and Table S3, respectively.

318 **Table 1**

319 Descriptive Summary of temporally adjusted annual average concentrations for PM₁₀, PM_{coarse}, PM_{2.5},
 320 PM_{2.5}Abs, PM₁₀ elements and PM_{2.5} elements (N = 19)

Pollutant	Mean	Median	SD	Min	Max
PM mass (µg/m³):					
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 (10⁻⁵m⁻¹):					
PM _{2.5} A	0.7	0.7	0.3	0.2	1.5
PM₁₀ elements (ng/m³):					
Si	3.9 x 10 ⁻¹	3.6 x 10 ⁻¹	2.4 x 10 ⁻¹	1.3 x 10 ⁻³	7.9 x 10 ⁻¹
S	4.9 x 10 ⁻¹	4.5 x 10 ⁻¹	1.3 x 10 ⁻¹	3.4 x 10 ⁻¹	7.8 x 10 ⁻¹
K	1.8 x 10 ⁻¹	1.9 x 10 ⁻¹	4.8 x 10 ⁻²	9.3 x 10 ⁻²	2.8 x 10 ⁻¹
V	1.2 x 10 ⁻³	1.1 x 10 ⁻³	5.0 x 10 ⁻⁴	6.0 x 10 ⁻⁴	2.4 x 10 ⁻³
Mn	5.5 x 10 ⁻³	5.1 x 10 ⁻¹	3.6 x 10 ⁻³	1.8 x 10 ⁻³	1.7 x 10 ⁻²
Fe	2.9 x 10 ⁻¹	2.5 x 10 ⁻¹	1.7 x 10 ⁻¹	6.4 x 10 ⁻²	6.8 x 10 ⁻¹
Cu	6.2 x 10 ⁻³	4.6 x 10 ⁻³	4.4 x 10 ⁻³	1.0 x 10 ⁻³	1.7 x 10 ⁻²
Zn	9.3 x 10 ⁻³	7.5 x 10 ⁻³	5.8 x 10 ⁻³	7.0 x 10 ⁻⁴	2.3 x 10 ⁻²
PM_{2.5} elements (ng/m³):					
Si	8.8 x 10 ⁻²	8.8 x 10 ⁻²	5.1 x 10 ⁻²	5.6 x 10 ⁻²	2.4 x 10 ⁻¹
K	7.9 x 10 ⁻²	7.9 x 10 ⁻²	2.0 x 10 ⁻²	5.0 x 10 ⁻²	1.2 x 10 ⁻¹
V	7.0 x 10 ⁻⁴	7.0 x 10 ⁻⁴	3.0 x 10 ⁻⁴	3.0 x 10 ⁻⁴	1.2 x 10 ⁻³
Mn	2.4 x 10 ⁻³	2.4 x 10 ⁻³	3.7 x 10 ⁻³	6.0 x 10 ⁻⁴	1.6 x 10 ⁻²
Fe	8.1 x 10 ⁻²	8.1 x 10 ⁻²	4.6 x 10 ⁻²	2.4 x 10 ⁻²	1.8 x 10 ⁻¹
Zn	6.5 x 10 ⁻³	6.5 x 10 ⁻³	5.9 x 10 ⁻³	1.8 x 10 ⁻³	2.9 x 10 ⁻²

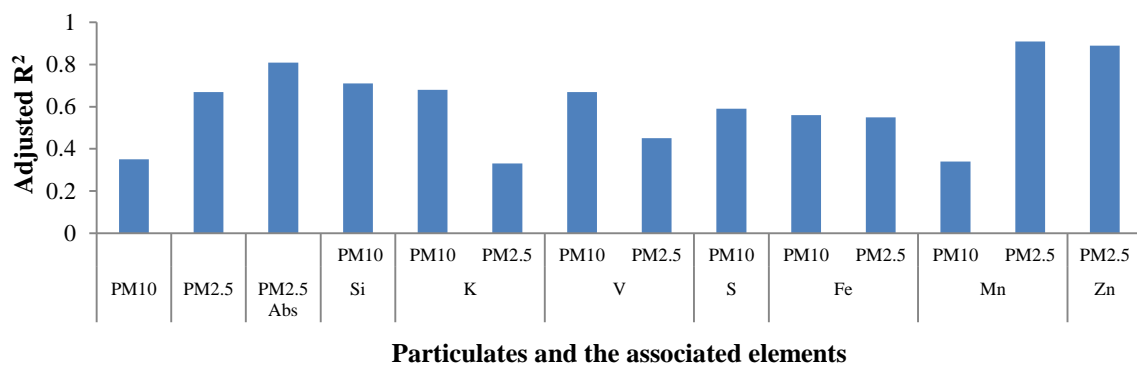
321
 322 The relationships between the annual averages of PM₁₀, PM_{coarse}, PM_{2.5}, and PM_{2.5}Abs and the
 323 selected elements are presented in Supplementary Table S4. A high correlation was observed for the
 324 association between PM₁₀ and PM_{coarse} (r = 0.95), moderate correlation between PM_{2.5} and PM_{2.5} Abs (r
 325 = 0.57), and poor correlation between the PM₁₀ and PM_{2.5} (r = -0.13). For the PM₁₀ elements, the highest
 326 correlation was observed between PM₁₀ and S (r > 0.5), while poor correlations were observed between
 327 PM₁₀ and the remaining elements. The PM_{2.5} was moderately correlated with K, V, Mn, Zn, and Fe
 328 with 0.45 < r < 0.53. PM_{2.5}Abs was correlated with PM_{2.5} Fe, -Mn, and -Zn; PM₁₀ S and -V.

329

330 3.2. LUR Models for PM_{2.5}, PM₁₀, PM_{2.5A} and the associated elements

331 LUR models were developed to predict the concentrations of PM_{2.5}, PM₁₀ and their respective
332 elements as well as PM_{2.5}Abs using 19 measurements sites across Perth. The LUR models for PM_{coarse},
333 Cu and PM₁₀ Zn had poor predictive ability (adjusted R² < 20%) and diagnostics, and are not presented.
334 Figure 2 illustrates the proportion of the spatial variability in the measured concentrations of PM_{2.5},
335 PM₁₀, and PM_{2.5}Abs and the selected elements explained by the LUR models.

336



337

338 **Figure 2** Proportion of the spatial variability in the measured concentrations of PM_{2.5}, PM₁₀, and PM_{2.5}Abs and
339 selected elements explained by the LUR models

340

341 The PM_{2.5} and PM_{2.5}Abs models had acceptable predictive ability with adjusted R² above 65%,
342 while the PM₁₀ model had much lower predictive ability. The LUR models captured greater proportion
343 of the spatial variability in the measured crustal elements (Si and K) and S in PM₁₀ fraction than that in
344 PM_{2.5}. The PM_{2.5} elemental models explained highest proportions in the spatial variability of industrial
345 related source element (Mn) and vehicle source element (Zn). The variability of industrial/fuel oil
346 combustion-element (V) in PM₁₀ was better captured than that in the PM_{2.5}. Similar proportion was
347 observed in the spatial variability of non-tailpipe vehicle source (Fe) in both PM₁₀ and PM_{2.5}.

348 Complete description of the structure of the LUR models is presented in Supplementary Table
349 S5, the summary of predictor variables captured by each LUR model and the corresponding unique
350 contributions is illustrated in Table 2. For PM₁₀, the most important predictor was green spaces. In
351 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.

356 **Table 2** Predictor variables and their unique contributions included in the LUR models for PM₁₀, 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%)						

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

362 The model diagnostics for PM_{2.5} and PM_{2.5}Abs using the variance inflation factors (VIF),
 363 Cook's D and Moran's I were acceptable (Supplementary Table S6). The VIF and the Cook's D values
 364 suggest the models do not violate the collinearity and influential observation assumptions. The Moran's
 365 I for all PM mass fractions and reflectance ranged from 0.083 to 0.970 with a p-value > 0.05,
 366 representing no spatial autocorrelation of the residuals. The elemental models did not violate the general
 367 assumptions for the development of the LUR models except for PM₁₀ Fe, PM₁₀ Mn, and PM_{2.5} V.
 368 Influential observation measured it. Based upon evaluation during the model development, it was
 369 observed that the parameter estimates of the models for PM₁₀ Fe, PM₁₀ Mn, and PM_{2.5} V with and
 370 without this influential site changed considerably. Therefore, the corresponding site was excluded from
 371 modelling for these elements. Among the LUR models for the PM₁₀ and PM_{2.5} elements, we found no
 372 spatial autocorrelation measured by Moran's I, except for PM_{2.5} Zn.

373 Table 3 shows the results of the model evaluation procedures, including the leave-one-out cross
 374 validation, hold out validation and cross-hold out validation. The differences in the adjusted R² between
 375 the final models and the leave one out cross validation results for PM_{2.5} was 17% and PM_{2.5}Abs was
 376 15%, indicating the spatial predictive ability of both models are relatively good.

377

378 **Table 3**

379 Summary of PM_{2.5} and PM_{2.5}A models evaluation average R² Validation results of LUR models for
 380 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 (n = 10)	Test sets (n = 9)	Training sets (n = 18)	Test sets (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

381

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

384 The hold-out and cross-hold-out validation procedures results also indicated the stability of our
 385 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

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

390

391 **4. Discussion**

392 *4.1. Air pollutant concentrations*

393 The annual mean concentrations of PM₁₀ (17.1 µg/m³), PM_{coarse} (12.4 µg/m³), and PM_{2.5} (4.7
394 µg/m³) across Perth were below the National Environment Protection Measures (NEPM) for ambient
395 air quality (20 µg/m³ for PM₁₀ and 8 µg/m³ for PM_{2.5}) (NEPC, 1998) and the WHO annual mean air
396 quality guidelines (20 µg/m³ for PM₁₀ and 10 µg/m³ for PM_{2.5}) (WHO, 2005). The mean concentration
397 of PM_{2.5}Abs was 0.67 x 10⁻⁵m⁻¹. There is no corresponding guideline for this pollutant or for any of the
398 PM elements. The high correlation between PM₁₀ and PM_{coarse} reflects the fact that PM₁₀ and PM_{coarse}
399 are emitted from similar sources such as non-exhaust emissions and fugitive coarse dust (Keuken et al.,
400 2013), while moderate correlation between PM_{2.5} and PM_{2.5} Abs, possibly reflecting that their
401 concentrations are influenced by complex interaction between local meteorological conditions and the
402 main source of these pollutants such as vehicles emissions and industrial activities (Keuken, et al.,
403 2013).

404 PM₁₀ and PM_{coarse} annual average concentrations were at the lower end of those European cities
405 participating in the ESCAPE study, which ranged from 14.8–43.1 µg/m³ for PM₁₀; and 6.0–23.6 µg/m³
406 for PM_{coarse}. Likewise, the PM_{2.5} concentrations and the PM_{2.5}Abs concentrations were both lower than
407 those of the ESCAPE cities, which were 8.3–29.3 µg/m³ for PM_{2.5}; and 0.8 – 3.0 x 10⁻⁵m⁻¹ for PM_{2.5}Abs)
408 (Eeftens, et al., 2012). The median concentrations of the PM elements were typically lower than the
409 ESCAPE cities (de Hoogh, et al., 2013) and the Calgary study (Zhang, et al., 2015). Similarly to the
410 PM mass and the reflectance measures, the concentrations of all PM mass elements in Perth were lower
411 than those measured in European and North America cities (Ross 2007). Possible reasons for these
412 differences between the studies include: (1) air pollutant concentrations in Perth tend to disperse across
413 the airshed due to its flat topography and strong ocean breezes (Yimin, et al., 2003); (2) the traffic

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

419

420 4.2. *Characterisation of surrounding environment and land use*

421 LUR models identify surrounding environmental and land use characteristics that may help
422 explain the variability in pollutant concentrations (Hoek, et al., 2008). Traffic intensity of heavy
423 vehicles in close proximity to the monitoring site was associated with increases in measured PM_{10}
424 concentrations, contributing approximately 27% in the explained variability. More than 90% of the
425 heavy vehicle fleets in Australia have diesel engines. Heavy vehicles account for around 25% of all
426 road transport fuel consumed in Australia, thus they potentially contribute more to traffic exhaust (ABS,
427 2014). The LUR models for PM_{10} in European cities such as Manchester, London and Ruhr Area, have
428 also included heavy vehicle traffic intensity in their models (Eeftens, et al., 2012).

429 Traffic variables were included in the all elements models of PM_{10} and $PM_{2.5}$ fraction. Keuken
430 et al., (2013) reported that Zn in particulates found in nearby roads originate from tire wear as its use
431 as galvanised materials in rubber production and re-suspended road dust. Fe was found to be emitted
432 from metal wear in the exhaust systems walls as flakes of iron and it is considered as a good marker for
433 brake wear emissions (Keuken, et al., 2013). Fe can also be emitted from tailpipes, formed within the
434 engine due to gas to particle conversion processes of the ferrocene, an agent to raise octane level of
435 diesel and gasoline fuel (Srimuruganandam et al., 2011). K is related to emissions from the ash fractions
436 of diesel exhausts (Srimuruganandam et al., 2012), and Mn is emitted from brake lining dust (Grigoratos
437 et al., 2015; Srimuruganandam, et al., 2011). Traffic-related emissions are not specific sources for V
438 and S in the PM_{10} fraction, as these elements are also emitted from fossil fuel combustion in industries
439 (Murillo et al., 2013; Srimuruganandam, et al., 2012). Most of the models variables appear attributable
440 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 PM₁₀.

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 – PM₁₀ 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 PM₁₀
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 PM₁₀
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 PM₁₀, PM₁₀ 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 PM₁₀ 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

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

500 The model R^2 of $PM_{2.5}$ was comparable with those in the ESCAPE study areas which ranged
501 from 49% to 89% and were slightly higher than those reported in North America (Ross, et al., 2007).
502 The predictive power of the $PM_{2.5}$ Abs model was also similar to the ESCAPE results (between 56%
503 and 95%), while for the PM_{10} model, the predictive ability was lower than those in the ESCAPE (50%-
504 90%) (Eeftens, et al., 2012). The PM_{10} elemental models explained between 34% and 71% of the
505 variability with only Fe, Zn and Cu performing poorly in comparison to the average R^2 for the ESCAPE
506 models across all cities. The $PM_{2.5}$ elemental models explained 36% – 90% of the concentration
507 variability. Our Fe and K models performed less well in comparison with ESCAPE but our V and Zn
508 models were able to explain a larger proportion of the variability than ESCAPE (de hoogh et al., 2013).

509 The stability of our LUR models is also indicated by the results of the evaluation procedures. The
510 differences in adjusted R^2 values between the model and the LOOCV were 17% for $PM_{2.5}$, 9% for PM_{10} ,
511 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
512 than the corresponding standard deviation. For both PM_{10} and $PM_{2.5}$ elements, the differences between
513 the adjusted R^2 -model and -LOOCV were within 15% suggesting the stability of the models, except for
514 PM_{10} Mn (19%) and $PM_{2.5}$ K (26%). RMSE values that were obtained from LOOCV were found to be
515 higher than those RMSE models. However, when the RMSE were assessed relative to the range of
516 measured air pollutant concentrations, we found small differences between the LOOCV and the models
517 i.e. 9% for $PM_{2.5}$ and 6% for $PM_{2.5}$ Abs, demonstrating the stable models.

518 From the hold out validation method, the $PM_{2.5}$ training models have larger average R^2 compared
519 with the full-sites model, indicating over fitting. The results from the cross hold out validation method
520 showed that the adjusted R^2 of $PM_{2.5}$ models based on the full sites and 18 sites were similar. However,
521 the true hold-out R^2 underestimated the models' predictive ability at the site that was not used for
522 developing the $PM_{2.5}$ models. For $PM_{2.5}$ Abs, the average R^2 of training models from the hold out
523 validation and evaluation models from the cross hold out validation were similar to the adjusted R^2 of
524 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 PM₁₀ 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

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

555

556 **References**

557

- 558 ABS, Australian Bureau of Statistics,. (2014). Motor Vehicle Census, Australia Retrieved 15 March,
559 2015, from <<http://www.abs.gov.au/ausstats/abs@.nsf/mf/9309.0>>
- 560 Andersen, Z. J., Bønnelykke, K., Hvidberg, M., Jensen, S. S., Ketzel, M., Loft, S., Sørensen, M.,
561 Tjønneland, A., Overvad, K., & Raaschou-Nielsen, O. (2012). Long-term exposure to air
562 pollution and asthma hospitalisations in older adults: a cohort study. *Thorax*, *67*(1), 6-11.
- 563 Basagaña, X., Rivera, M., Aguilera, I., Agis, D., Bouso, L., Elosua, R., Foraster, M., de Nazelle, A.,
564 Nieuwenhuijsen, M., Vila, J., & Künzli, N. (2012). Effect of the number of measurement sites
565 on land use regression models in estimating local air pollution. *Atmospheric Environment*, *54*,
566 634-642. doi: <http://dx.doi.org/10.1016/j.atmosenv.2012.01.064>
- 567 Batterman, S. A., Zhang, K., & Kononowech, R. (2010). Prediction and analysis of near-road
568 concentrations using a reduced-form emission/dispersion model. *Environmental Health*, *9*,
569 29-29. doi: 10.1186/1476-069x-9-29
- 570 Beelen, R., Raaschou-Nielsen, O., Stafoggia, M., Andersen, Z. J., Weinmayr, G., Hoffmann, B., Wolf,
571 K., Samoli, E., Fischer, P., & Nieuwenhuijsen, M. (2014). Effects of long-term exposure to
572 air pollution on natural-cause mortality: an analysis of 22 European cohorts within the
573 multicentre ESCAPE project. *The Lancet*, *383*(9919), 785-795.
- 574 Bukowiecki, N., Lienemann, P., Hill, M., Furger, M., Richard, A., Amato, F., Prévôt, A.,
575 Baltensperger, U., Buchmann, B., & Gehrig, R. (2010). PM10 emission factors for non-
576 exhaust particles generated by road traffic in an urban street canyon and along a freeway in
577 Switzerland. *Atmospheric Environment*, *44*(19), 2330-2340.
- 578 Cesaroni, G., Forastiere, F., Stafoggia, M., Andersen, Z. J., Badaloni, C., Beelen, R., Caracciolo, B.,
579 de Faire, U., Erbel, R., Eriksen, K. T., Fratiglioni, L., Galassi, C., Hampel, R., Heier, M.,
580 Hennig, F., et al. (2014). *Long term exposure to ambient air pollution and incidence of acute
581 coronary events: prospective cohort study and meta-analysis in 11 European cohorts from the
582 ESCAPE Project* (Vol. 348).
- 583 Chan, Y.-C., Cohen, D. D., Hawas, O., Stelcer, E., Simpson, R., Denison, L., Wong, N., Hodge, M.,
584 Comino, E., & Carswell, S. (2008). Apportionment of sources of fine and coarse particles in
585 four major Australian cities by positive matrix factorisation. *Atmospheric Environment*, *42*(2),
586 374-389.
- 587 Chow, J. C., & Watson, J. G. (2002). Review of PM2.5 and PM10 apportionment for fossil fuel
588 combustion and other sources by the chemical mass balance receptor model. *Energy & Fuels*,
589 *16*(2), 222-260.
- 590 de Hoogh, K., Wang, M., Adam, M., Badaloni, C., Beelen, R., Birk, M., Cesaroni, G., Cirach, M.,
591 Declercq, C., & Dedele, A. (2013). Development of land use regression models for particle
592 composition in twenty study areas in Europe. *Environmental Science & Technology*, *47*(11),
593 5778-5786.
- 594 Dirgawati, M., Barnes, R., Wheeler, A. J., Arnold, A.-L., McCaul, K. A., Stuart, A. L., Blake, D.,
595 Hinwood, A., Yeap, B. B., & Heyworth, J. S. (2015). Development of Land Use Regression
596 models for predicting exposure to NO₂ and NO_x in Metropolitan Perth, Western Australia.
597 *Environmental Modelling & Software*, *74*, 258-267.
- 598 EEA, European Environment Agency,. (2011). Size of the vehicle fleet (TERM 032) - Assessment
599 Retrieved 28 April, 2015, from <[http://www.eea.europa.eu/data-and-maps/indicators/size-of-
600 the-vehicle-fleet/size-of-the-vehicle-fleet-2](http://www.eea.europa.eu/data-and-maps/indicators/size-of-the-vehicle-fleet/size-of-the-vehicle-fleet-2)>
- 601 Eeftens, M., Beelen, R., de Hoogh, K., Bellander, T., Cesaroni, G., Cirach, M., Declercq, C., Dèdelè,
602 A., Dons, E., de Nazelle, A., Dimakopoulou, K., Eriksen, K., Falq, G., Fischer, P., Galassi,
603 C., et al. (2012). Development of Land Use Regression Models for PM_{2.5}, PM_{2.5}

604 Absorbance, PM10 and PMcoarse in 20 European Study Areas; Results of the ESCAPE
605 Project. *Environmental Science & Technology*, 46(20), 11195-11205. doi: 10.1021/es301948k

606 Eeftens, M., Hoek, G., Gruzieva, O., Mölter, A., Agius, R., Beelen, R., Brunekreef, B., Custovic, A.,
607 Cyrus, J., & Fuertes, E. (2014). Elemental composition of particulate matter and the
608 association with lung function. *Epidemiology*, 25(5), 648-657.

609 Eeftens, M., Tsai, M.-Y., Ampe, C., Anwander, B., Beelen, R., Bellander, T., Cesaroni, G., Cirach,
610 M., Cyrus, J., de Hoogh, K., De Nazelle, A., de Vocht, F., Declercq, C., Dèdelè, A., Eriksen,
611 K., et al. (2012). Spatial variation of PM2.5, PM10, PM2.5 absorbance and PMcoarse
612 concentrations between and within 20 European study areas and the relationship with NO2 –
613 Results of the ESCAPE project. *Atmospheric Environment*, 62, 303-317. doi:
614 <http://dx.doi.org/10.1016/j.atmosenv.2012.08.038>

615 Fuertes, E., MacIntyre, E., Agius, R., Beelen, R., Brunekreef, B., Bucci, S., Cesaroni, G., Cirach, M.,
616 Cyrus, J., & Forastiere, F. (2014). Associations between particulate matter elements and
617 early-life pneumonia in seven birth cohorts: Results from the ESCAPE and TRANSPHORM
618 projects. *International Journal of Hygiene and Environmental Health*, 217(8), 819-829.

619 Grigoratos, T., & Martini, G. (2015). Brake wear particle emissions: a review. *Environmental Science
620 and Pollution Research International*, 22, 2491-2504. doi: 10.1007/s11356-014-3696-8

621 Gulliver, J., & Briggs, D. (2011). STEMS-Air: A simple GIS-based air pollution dispersion model for
622 city-wide exposure assessment. *Science of The Total Environment*, 409(12), 2419-2429. doi:
623 <http://dx.doi.org/10.1016/j.scitotenv.2011.03.004>

624 Hoek, G., Beelen, R., de Hoogh, K., Vienneau, D., Gulliver, J., Fischer, P., & Briggs, D. (2008). A
625 review of land-use regression models to assess spatial variation of outdoor air pollution.
626 *Atmospheric Environment*, 42(33), 7561-7578.

627 Hoek, G., Krishnan, R. M., Beelen, R., Peters, A., Ostro, B., Brunekreef, B., & Kaufman, J. D.
628 (2013). Long-term air pollution exposure and cardio- respiratory mortality: a review. [journal
629 article]. *Environmental Health*, 12(1), 1-16. doi: 10.1186/1476-069x-12-43

630 Keuken, M. P., Moerman, M., Voogt, M., Blom, M., Weijers, E. P., Röckmann, T., & Dusek, U.
631 (2013). Source contributions to PM2.5 and PM10 at an urban background and a street
632 location. *Atmospheric Environment*, 71, 26-35. doi:
633 <http://dx.doi.org/10.1016/j.atmosenv.2013.01.032>

634 Larson, T., Gould, T., Simpson, C., Liu, L.-J. S., Claiborn, C., & Lewtas, J. (2004). Source
635 apportionment of indoor, outdoor, and personal PM2.5 in Seattle, Washington, using positive
636 matrix factorization. *Journal of the Air & Waste Management Association*, 54(9), 1175-1187.

637 Liang, J. (2013). Chemical Modeling for Air Resources Fundamentals, Applications, and
638 Corroborative Analysis *Chapter 9 - Particulate matter* (pp. 189-219). Burlington: Elsevier
639 Science.

640 McDonald, A. G., Bealey, W. J., Fowler, D., Dragosits, U., Skiba, U., Smith, R. I., Donovan, R. G.,
641 Brett, H. E., Hewitt, C. N., & Nemitz, E. (2007). Quantifying the effect of urban tree planting
642 on concentrations and depositions of PM10 in two UK conurbations. *Atmospheric
643 Environment*, 41(38), 8455-8467. doi: <http://dx.doi.org/10.1016/j.atmosenv.2007.07.025>

644 Mölter, A., Lindley, S., de Vocht, F., Simpson, A., & Agius, R. (2010). Modelling air pollution for
645 epidemiologic research — Part I: A novel approach combining land use regression and air
646 dispersion. *Science of The Total Environment*, 408(23), 5862-5869. doi:
647 <http://dx.doi.org/10.1016/j.scitotenv.2010.08.027>

648 Murillo, J. H., Roman, S. R., Rojas Marin, J. F., Ramos, A. C., Jimenez, S. B., Gonzalez, B. C., &
649 Baumgardner, D. G. (2013). Chemical characterization and source apportionment of PM10
650 and PM2.5 in the metropolitan area of Costa Rica, Central America. *Atmospheric Pollution
651 Research*, 4(2), 181-190. doi: <http://dx.doi.org/10.5094/APR.2013.018>

652 NEPC, National Environment Protection Council. (1998). National Environment Protection (Ambient
653 Air Quality) Measure Retrieved 15 December 2015, from
654 <http://www.scew.gov.au/nepms/ambient-air-quality>

655 Norman, P. E., Flicker, L., Almeida, O. P., Hankey, G. J., Hyde, Z., & Jamrozik, K. (2009). Cohort
656 profile: the health in men study (HIMS). *International Journal of Epidemiology*, 38(1), 48-52.

657 NPI, National Pollutant Inventory,. (2014). 2013/2014 data within Western Australia - All Substances
658 from All Sources Retrieved 3 August 2015, from <http://www.npi.gov.au/npidata/>

- 659 Raaschou-Nielsen, O., Andersen, Z. J., Beelen, R., Samoli, E., Stafoggia, M., Weinmayr, G.,
660 Hoffmann, B., Fischer, P., Nieuwenhuijsen, M. J., Brunekreef, B., Xun, W. W., Katsouyanni,
661 K., Dimakopoulou, K., Sommar, J., Forsberg, B., et al. (2013). Air pollution and lung cancer
662 incidence in 17 European cohorts: prospective analyses from the European Study of Cohorts
663 for Air Pollution Effects (ESCAPE). *The Lancet Oncology*, 14(9), 813-822. doi:
664 [http://dx.doi.org/10.1016/S1470-2045\(13\)70279-1](http://dx.doi.org/10.1016/S1470-2045(13)70279-1)
- 665 Refaeilzadeh, P., Tang, L., & Liu, H. (2009). Cross-validation *Encyclopedia of database systems* (pp.
666 532-538): Springer.
- 667 Ross, Z., Jerrett, M., Ito, K., Tempalski, B., & Thurston, G. D. (2007). A land use regression for
668 predicting fine particulate matter concentrations in the New York City region. *Atmospheric*
669 *Environment*, 41(11), 2255-2269.
- 670 Srimuruganandam, B., & Shiva Nagendra, S. M. (2011). Chemical characterization of PM10 and
671 PM2.5 mass concentrations emitted by heterogeneous traffic. *Science of The Total*
672 *Environment*, 409(17), 3144-3157. doi: <http://dx.doi.org/10.1016/j.scitotenv.2011.04.042>
- 673 Srimuruganandam, B., & Shiva Nagendra, S. M. (2012). Source characterization of PM10 and PM2.5
674 mass using a chemical mass balance model at urban roadside. *Science of The Total*
675 *Environment*, 433, 8-19. doi: <http://dx.doi.org/10.1016/j.scitotenv.2012.05.082>
- 676 Stanek, L. W., Sacks, J. D., Dutton, S. J., & Dubois, J.-J. B. (2011). Attributing health effects to
677 apportioned components and sources of particulate matter: an evaluation of collective results.
678 *Atmospheric Environment*, 45(32), 5655-5663.
- 679 Urman, R., Gauderman, J., Fruin, S., Lurmann, F., Liu, F., Hosseini, R., Franklin, M., Avol, E.,
680 Penfold, B., & Gilliland, F. (2014). Determinants of the spatial distributions of elemental
681 carbon and particulate matter in eight Southern Californian communities. *Atmospheric*
682 *Environment*, 86, 84-92.
- 683 Wang, M., Beelen, R., Basagana, X., Becker, T., Cesaroni, G., de Hoogh, K., Dedele, A., Declercq,
684 C., Dimakopoulou, K., Eeftens, M., Forastiere, F., Galassi, C., Gražulevičienė, R., Hoffmann,
685 B., Heinrich, J., et al. (2013). Evaluation of Land Use Regression Models for NO2 and
686 Particulate Matter in 20 European Study Areas: The ESCAPE Project. *Environmental Science*
687 *& Technology*, 47(9), 4357-4364. doi: 10.1021/es305129t
- 688 Wang, M., Beelen, R., Eeftens, M., Meliefste, K., Hoek, G., & Brunekreef, B. (2012). Systematic
689 Evaluation of Land Use Regression Models for NO2. *Environmental Science & Technology*,
690 46(8), 4481-4489. doi: 10.1021/es204183v
- 691 Wang, M., Brunekreef, B., Gehring, U., Szpiro, A., Hoek, G., & Beelen, R. (2016). A New Technique
692 for Evaluating Land-use Regression Models and Their Impact on Health Effect Estimates.
693 *Epidemiology*, 27(1), 51-56.
- 694 WHO, W. H. O. (2005). WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide
695 and sulfur dioxide.
- 696 Yimin, M., & Lyons, T. J. (2003). Recirculation of coastal urban air pollution under a synoptic scale
697 thermal trough in Perth, Western Australia. *Atmospheric Environment*, 37(4), 443-454. doi:
698 [http://dx.doi.org/10.1016/S1352-2310\(02\)00926-3](http://dx.doi.org/10.1016/S1352-2310(02)00926-3)
- 699 Zhang, J. J., Sun, L., Barrett, O., Bertazzon, S., Underwood, F. E., & Johnson, M. (2015).
700 Development of land-use regression models for metals associated with airborne particulate
701 matter in a North American city. *Atmospheric Environment*, 106, 165-177.
- 702 Zhou, J., Ito, K., Lall, R., Lippmann, M., & Thurston, G. (2010). Time-series analysis of mortality
703 effects of fine particulate matter components in Detroit and Seattle. *Environmental health*
704 *perspectives*, 119(4), 461-466.
- 705 Zou, B., Wilson, J. G., Zhan, F. B., & Zeng, Y. (2009). Air pollution exposure assessment methods
706 utilized in epidemiological studies. *Journal of Environmental Monitoring*, 11(3), 475-490.

707

708

