TITLE

A national satellite-based land-use regression model for air pollution exposure assessment in Australia

AUTHORS

Luke D. Knibbs[1*]

Michael G. Hewson[2]

Matthew J. Bechle[3]

Julian D. Marshall[3]

Adrian G. Barnett[4]

[1] School of Population Health, The University of Queensland, Brisbane, Australia

[2] School of Geography, Planning and Environmental Management, The University of Queensland, Brisbane, Australia

[3] Department of Civil Engineering, The University of Minnesota, Minneapolis, USA

[4] School of Public Health and Social Work & Institute of Health and Biomedical Innovation, Queensland University of Technology, Brisbane, Australia

* Corresponding author

Public Health Building

The University of Queensland

Herston, QLD 4006, Australia

p: +61 7 3365 5409

e: l.knibbs@uq.edu.au

ABSTRACT

Land-use regression (LUR) is a technique that can improve the accuracy of air pollution exposure assessment in epidemiological studies. Most LUR models are developed for single cities, which places limitations on their applicability to other locations. We sought to develop a model to predict nitrogen dioxide (NO_2) concentrations with national coverage of Australia by using satellite observations of tropospheric NO_2 columns combined with other predictor variables. We used a generalised estimating equation (GEE) model to predict annual and monthly average ambient NO_2 concentrations measured by a national monitoring network from 2006 through 2011. The best annual model explained 81% of spatial variation in NO_2 (absolute RMS error = 1.4 ppb), while the best monthly model explained 76% (absolute RMS error = 1.9 ppb). We applied our models to predict NO_2 concentrations at the ~350,000 census mesh blocks across the country (a mesh block is the smallest spatial unit in the Australian census). National population-weighted average concentrations ranged from 7.3 ppb (2006) to 6.3 ppb (2011). We found that a simple approach using tropospheric NO_2 column data yielded models with slightly better predictive ability than those produced using a more involved approach that required simulation of surface-to-column ratios. The models were capable of capturing within-urban variability in NO_2, and offer the ability to estimate ambient NO_2 concentrations at monthly and annual time scales across Australia from 2006-2011. We are making our model predictions freely available for research.

KEYWORDS: nitrogen dioxide; land use regression; exposure; epidemiology; Australia

FUNDING SOURCES

LDK acknowledges an NHMRC Early Career (Australian Public Health) Fellowship (APP1036620). This material is partially based on work supported by the United States National Science Foundation under Grant No. 0853467.

1. INTRODUCTION

Outdoor (ambient) air pollution is a major contributor to the global burden of disease and a leading environmental risk factor for morbidity and mortality (Lim et al., 2012). Accurate estimates of people's exposure to ambient air pollution are required to quantify and understand its effects on health. Exposure estimates have traditionally involved using average measurements from air pollution monitors (e.g. Barnett et al., 2005), assigning exposure using the nearest monitor to a person's residence (e.g. Ritz et al., 2002) or using a proxy like distance to the nearest main road (e.g. Hoffmann et al., 2007). There is potential for exposure misclassification with all these approaches due to their limited ability to capture the spatial variability that characterises some air pollutants (Jerrett et al., 2005; Hoek et al., 2008).

Land-use regression (LUR) is a technique that can improve the accuracy of air pollution exposure estimates. It uses measurements at a set of locations combined with spatial variables to build statistical models that can predict concentrations at unmeasured locations (Hoek et al., 2008). A key limitation of most LUR models is that they are constrained to individual cities, and a model built for one location is not necessarily transferrable to another (Briggs, 2007; Vienneau et al., 2010). However, the recent availability of high quality satellite data has helped address these issues by permitting better representation of large areas in LUR. Satellite-based LUR models for the USA (Novotny et al., 2011), Canada (Hystad et al., 2011) and Western Europe (Vienneau et al., 2013) have been developed, and have similar predictive ability to city models but with national coverage. In some cases, their spatial resolution can rival that of city models (Novotny et al., 2011). These satellite-based LUR models have attractive applications in air pollution epidemiology, environmental justice, and planning studies.

Australia (population ~23 million) is one of the world's least densely populated countries (3 people/km^2) but also one of the most urbanised, as ninety percent of the population live in or near cities (Australian Bureau of Statistics, 2013a,b). There are relatively few regulatory air pollution monitoring sites in Australia. For example, Canada's population is about 50% larger than Australia's, but Canada has about twice the number of monitors (Hystad et al., 2011). Most monitoring sites in Australia are located in and around major cities but are sparsely distributed, which means they are less than ideal for assessing spatial variability in ambient air pollution levels. This can make exposure assessment for the Australian population problematic.

We sought to develop a satellite-based LUR model for Australia that could predict ambient air pollution exposure levels with good accuracy. We aimed to add to the evidence base by investigating the utility of a national satellite-based LUR in a location where ground-based monitors are scant. Most previous national LUR models have focussed on annual concentrations (e.g. Hystad et al., 2011; Vienneau et al., 2013). We aimed to expand the temporal component of our models to include monthly exposure estimates. Having monthly estimates of exposure would be useful for examining health outcomes where exposures within the year are important (such as birth outcomes), and to examine the potential health effects of interactions between seasons and pollution exposure. We sought to produce both monthly and annual exposure estimates over a 6 year period.

2. METHODS

2.1 Predictor variables

Measured NO_2

We focussed on nitrogen dioxide (NO_2) because it is strong marker of traffic and other combustion-derived pollution (e.g. industry, airports) and a key component of ambient air pollution (Briggs et al., 1997, Richter et al., 2005). We obtained hourly average ground-level NO_2 measurements from

January 2006 through December 2011 from the Australian agencies responsible for regulatory ambient air pollution monitoring. NO_2 concentrations were measured using the standard chemiluminescence method, which can be subject to bias due to interference by other nitrogen oxides but is widely used in research and assessing compliance with regulations (Novotny et al., 2011). The measurements had undergone basic quality assurance procedures and we examined them further for completeness and validity. There were 68 monitoring sites across Australia where NO_2 was measured during the study period (supplement, Table S3). The sites' locations ranged from dense urban areas with multiple pollution sources nearby through to rural areas with few local sources.

Land use

We sourced data on natural and anthropogenic features that have a plausible association with measured NO_2 concentrations. Our choice of variables was guided by previous satellite LUR models and data availability (Hystad et al., 2011; Novotny et al., 2011; Vienneau et al., 2013). The variables selected are summarised in Table 1. We incorporated land use data from a range of sources including satellites and the Australian census. Detailed information on data sources is provided in the supplement (tTable S2). We used ArcGIS version 10.0 (ESRI Inc., Redlands, USA) to process our data.

Satellite data: NO_2

The Ozone Monitoring Instrument (OMI) aboard the Aura satellite produces daily global observations of NO_2 tropospheric column abundance at a resolution of 13 × 24 km (nadir) using a differential optical absorption spectroscopy (DOAS) algorithm (Levelt et al., 2006). We obtained the average tropospheric NO_2 columns over Australia for each month from 2006-11. We then produced estimates of ground-level NO_2 by using the Weather Research and Forecasting model

(WRF-Chem) to predict monthly surface-to-column ratios. This approach is a standard method to convert tropospheric column NO_2 abundance (in molecules per cm^2) to ground-level NO_2 concentration (in ppb), and has been described extensively (Lamsal et al., 2008; Bechle et al., 2013). Detailed information on satellite data retrieval and processing is given in the supplement (pages S3-S9).

2.2 Modelling approach

We generated 22 buffers from 100 m to 10 km around each monitoring site (Table 1). This approach was analogous to other national-scale models and aims to capture both proximate and more distant sources of variability in NO_2 concentrations (Novotny et al., 2011; Vienneau et al., 2013). Some variables were calculated within each buffer (e.g., percent tree cover, road length, impervious surface area) using either the average or sum of the variable in each of the 22 buffers (Table 1). Other variables were determined at each monitoring point (e.g., elevation, distance to coast). Detailed information about each variable is presented in the supplement (tTable S2). There were 286 buffer variables (13 variables calculated at 22 buffers each) and 29 point variables, giving a total of 315 independent variables.

Annual model

The dependent variable (measured NO_2) was longitudinal, as measurements were repeated at the 68 monitoring sites over 6 years. We only included years where more than 90% of the daily measurements from a site were non-missing. Selecting the best subset of predictor variables was complex as there were 315 to choose from. Because of the large number of variables and the computational issues this presented, we employed a two-stage variable selection procedure. In the first stage, we narrowed the list of variables by using the lasso method in the 'glmnet' library (Friedman et al., 2010). This places a bound on the sum of absolute coefficient values and

minimises the sum of squared errors (Tibshirani, 1996). Because this method is not suiTable for longitudinal data, we ran separate lasso models for each year. We tabulated the frequencies of selected variables as an indicator of their relative importance over the 6 years (supplement, pages S16-S17). We then used all those variables that were selected at least once in the second stage of variable selection.

In the second stage of variable selection we followed the general approach of Su et al. (2009). This is a forward selection procedure where an independent variable can be added to the model on the conditions that: (1) it is statistically significant at the 5% level, and; (2) the variance inflation factors of all variables in the model remain below five. The second condition is an attempt to avoid co-linearity. For all variables that met these two conditions, we used 10-fold cross-validation with 3 replications using the 'cvTools' library (Alfons, 2012) to choose the variable with the smallest cross-validated root mean square error. We only added a variable to the model if the mean cross-validated error plus the cross-validated standard error was smaller than the previous minimum root mean square error. This criterion aims to create a parsimonious model. These longitudinal models used all available years of data and were fitted by a generalised estimating equation (GEE) model using the 'geepack' library (Højsgaard et al., 2006). This produced one model for predicting annual average concentrations for each year during 2006-2011. We assumed an independent correlation structure for residuals from the same monitoring site.

Model validation

We visually checked the residuals of the final models for outliers, and used Cook's distance and df-beta statistics to test for influential observations. We used five-fold cross-validation with five replications to estimate the prediction error of the final models on an absolute and percentage scale. We examined the importance of individual sites by comparing those with the highest Cook's

distance against 3 randomly selected sites (supplement, pages S20-S21). Additional details on model validation are given in the supplement (pages S13-S14). All modelling was performed using R version 3.0.3 (R Foundation for Statistical Computing, Vienna, Austria).

Monthly model

We used the same approach for our monthly model, except in this case there were up to 72 average concentrations for each monitoring site (12 months by 6 years). We only included months from sites where 25 or more daily pollution measurements were non-missing. The first variable selection stage using the lasso method was run separately for each month. We then used all those variables that were selected 6 or more times in the second stage. We started the second stage variable selection with a model that included month as a factor, as we strongly suspected that this would be an important variable and wanted to avoid potential proxies for month (e.g., solar radiation, rainfall, temperature) being selected unnecessarily. Using the same GEE approach as the annual model, we produced one model for predicting the 72 monthly average concentrations during 2006-2011.

Comparison of different satellite NO_2 estimates

We assessed whether surface NO_2 estimates derived using surface-to-column ratios from WRF-Chem lead to models with better predictive ability for ground level NO_2 than the easier to obtain estimates of tropospheric NO_2 column density. For both our annual and monthly models, we examined two alternatives; one with surface NO_2 estimates as a candidate variable ('surface model') and one with NO_2 column density estimates ('column model'). All other candidate variables were the same across the two models.

Applying the models

We obtained the boundaries of the ~350,000 Australian Bureau of Statistics 'mesh blocks' that cover the entirety of Australia (Australian Bureau of Statistics, 2011). Mesh blocks are the smallest spatial unit used in the Australian census. They contain 62 people on average (range 0 to 2,339), and have a highly variable size (range: 1.0×10^{-4} to 1.7×10^{5} km^2; population-weighted mean size = 26.3 km^2). The majority of populated mesh blocks include between 30 and 60 dwellings (Australian Bureau of Statistics, 2013c). We determined the centroid of every mesh block and used our final models to predict annual average NO_2 concentrations at the centroids for each year during 2006 to 2011 (Hystad et al., 2011; Novotny et al., 2011).

3. RESULTS

The number of air quality monitoring sites that met the inclusion criteria for the annual model ranged from 55 (2006) to 66 (2010) out of a possible 68. There were 358 annual measurements that met the inclusion criteria over the 6-year study period. Between 47 (February 2006) and 67 (May/July 2010) out of 68 sites met the inclusion criteria for the monthly model, and there were 4,371 monthly measurements over the 72 months. The descriptive statistics of measurements used to build the annual and monthly models are shown in Table 2.

The best annual surface model (i.e. model that included surface estimates of NO_2) was capable of explaining 79% of the variability in measured NO_2 concentrations (Table 3). This increased to 81% in the best annual column model (i.e. model that included tropospheric column NO_2 density). The two models had comparable absolute and percentage root mean squared (RMS) prediction errors (Table 3). All other variables in the final models were identical with the exception of summertime mean daily solar exposure, which appeared in the surface model but not the column model. In both

models, the three variables that made the largest contribution to overall R^2 were satellite NO_2, impervious surfaces within 1,200 m, and major roads within 500 m.

The best monthly model that included NO_2 surface estimates explained 73% of the variability in measured NO_2 (Table 4). The best model including NO_2 column measurements explained 76%. The monthly surface and column models had very similar RMS prediction errors (Table 4). Excluding year and month, there were 4 common variables that were in both monthly models (minor roads within 8,000 m, major roads within 100 m, industrial site density within 400 m, industrial land use within 10,000 m). In both models, the variable that made the largest contribution to R^2 was satellite NO_2 (Table 4). The next largest contributors to the column model were minor roads within 8,000m and industrial land use within 10,000 m. The next largest contributors to the surface model were the months of July and August, which is during the Australian winter.

Residuals were approximately normally distributed in all models (supplement, figures S3-S6). For a given variable in the final models some monitoring sites were more influential than others, but after investigation we found no overt undue influence on the models. The results of model checking using df-beta statistics and Cook's distance are presented in the supplement (pages S18-S26). We compared the values of predictors at the monitoring sites with those at the ~350,000 mesh block centroids around Australia and found that they were very similar (supplement, Table S11).

The average NO_2 concentration predicted by the annual surface model for 2008 is in Figure 1, which was selected as a representative example from the 6-year study period. The mostly unpopulated interior of the country had concentrations around 2 ppb. Areas with higher concentrations (from ~5 up to >20 ppb) are the cities and major towns. The inset of Figure 1 focuses on Sydney, Australia's most populous city (4.4 million). Elevated concentrations (> 10

ppb) were predicted on and near major roads. Maximum concentrations (> 20 ppb) were predicted in locations with many nearby major roads and industrial areas. This general pattern was present in the 8 state and territory capital cities around Australia, although levels were highest in the 3 largest cities: Sydney, Melbourne (4 million) and Brisbane (2.1 million).

The annual NO_2 concentrations predicted at the ~350,000 census mesh block centroids by our annual surface model are shown in Figure 2 (Hart et al., 2009). The median concentration predicted across Australia decreased from 6.3 ppb in 2006 to 5.3 ppb in 2011, which was a reduction of 16% over the 6 years. Because about 25% of mesh blocks are uninhabited, we also calculated population-weighted concentrations to indicate the average concentration that Australians are exposed to. These ranged from 7.3 ppb (2006) to 6.3 ppb (2011), a decrease of 14% from 2006 to 2011. The NO_2 levels predicted at each mesh block by the column model were almost identical to those predicted by the surface model (supplement, Table S12).

We also examined predicted NO_2 concentrations in Sydney. Across the ~57,000 mesh blocks that made up the greater Sydney area (Australian Bureau of Statistics, 2011), the population-weighted average annual NO_2 concentration ranged from 9.9 ppb (2006) to 8.7 ppb (2011), a decrease of 12% between 2006 and 2011. A range of statistics on predicted NO_2 levels across Sydney are in the supplement (tTable S13).

4. DISCUSSION

We assessed the ability of satellite-based LUR models to predict monthly and annual average NO_2 concentrations in Australia from 2006-11. We found that the best annual model explained 81% of variation in NO_2, while the best monthly model explained 76%. We applied our models to predict NO_2 concentrations at each of the ~350,000 census mesh blocks across the country and found a

slight but consistent decrease between 2006 and 2011. Predicted concentrations were generally modest compared to studies in the USA and Europe (Beelen et al., 2007; Hart et al., 2009; Vienneau et al., 2013), but were more comparable with those predicted in Canada using similar methods (Hystad et al., 2011).

While it is difficult to comprehensively compare our results to other national satellite-based LUR due to differences in methodology, we found that our models captured a similar or slightly higher amount of variability in NO_2. An annual model for the USA explained 78% of variability in measured NO_2, while a Canadian model explained 73% (Hystad et al., 2011; Novotny et al., 2011). A recent model covering Western Europe explained 60% of measured NO_2 variability (Vienneau et al., 2013). The prediction error of all our models was comparable or slightly lower than other studies, albeit using different validation methods (Novotny et al., 2011; Vienneau et al., 2013; Lee and Koutrakis, 2014).

The variables in our models were generally consistent with those reported in previous studies, with both major and minor roads featuring prominently as well as impervious surface cover (Novotny et al., 2011; Vienneau et al., 2013). Increased roads, impervious surfaces and industrial variables were all associated with higher NO_2. Road traffic is a major source of NO_2, and impervious surfaces are greater in built-up locations and may reflect increased NO_2 sources in these areas. Increased open space (e.g. parklands) and summertime solar exposure were both associated with lower NO_2. Open spaces are relatively free of substantial NO_2 sources, while the presence of summertime solar exposure in the annual surface model may be due to the shorter lifetime of nitrogen oxides in the lower troposphere during the summer months (Lamsal et al., 2010).

Industrial land use and the density of nearby industrial point source NO_X emissions featured in all of our models. Industrial sources are a leading contributor to outdoor NO_X in Australia (Australian Bureau of Statistics, 2012), and the conspicuous presence of industrial variables in our models is in keeping with this. Industrial emissions and industrial land use have been found to be significant predictors of NO_2 in some other national LUR models (Hart et al., 2009; Hystad et al., 2011). Including industrial variables can improve model performance but creates models that are less specific to vehicle emissions (e.g. Novotny et al., 2011). However, it also means that model predictions are able to capture both vehicle and non-vehicle sources of NO_2, both of which contribute to ambient NO_2 and human exposure.

We found that satellite-derived NO_2 estimates from the Ozone Monitoring Instrument added the most predictive ability to 3 of our 4 models. Previous studies have reported pronounced reductions in model performance when satellite NO_2 is excluded (e.g. Novotny et al., 2011; Vienneau et al., 2013). Our findings further confirm the utility of satellite NO_2 in national-scale LUR, and the improvements in exposure assessment that it offers.

Notably, we found that the best annual and monthly models that included NO_2 tropospheric column observations exhibited slightly better predictive ability with comparable error to those that included estimates of surface NO_2 obtained by modelling surface-to-column ratios using WRF-Chem. This could reflect the fact that tropospheric columns are dominated by NO_2 in the part of the atmosphere closest to Earth's surface (i.e. the boundary layer), and are therefore useful proxies of relative ground-level concentrations (Richter et al., 2005). Also, the parameters selected in the process of modelling surface-to-column ratios (supplement, pages S4-S9) may add additional error into NO_2 estimates obtained using this method (Lamsal et al., 2008; Bechle et al., 2013). Modelling surface-to-column ratios is both computationally- and time-intensive and requires technical expertise. Our

findings are promising for those who are interested in less complex approaches to NO_2 exposure assessment. We note, however, that these findings may be specific to the context of our study and are not necessarily applicable beyond that.

Our study has some important limitations. The monitoring data used to build the models came from only 68 sites, which is small when compared with other national LUR studies (e.g. Novotny et al., 2011; Vienneau et al., 2013), particularly when Australia's size is considered (supplement, Table S3; Johnson et al., 2010; Basagaña et al., 2012; Wang et al., 2012). Indeed, it was this paucity of monitoring that provided the initial motivation for our study. However, this means that model predictions may be valid only when applied to environments similar to those where monitoring was performed. We addressed this by comparing the summary statistics of predictors at the monitoring sites with those at the mesh block centroids that covered all of Australia (supplement, Table S11). We found no evidence to indicate that the monitoring sites on which the models were based differed markedly from the broader Australian context that they were applied to. However, the monitoring sites are primarily used for regulatory ambient air monitoring, and so they were not typically sited near substantial emission sources. For this reason, using our models to predict concentrations in pollution 'hot-spots' (such as road intersections or areas with very localised non-road emissions) should be undertaken with caution, and would require additional validation against measurements from the area of interest.

Because we used a generalised estimating equation we were unable to assess spatial autocorrelation in the model residuals. However, all other national satellite-based NO_2 models have reported that spatial autocorrelation was not present in model residuals, and this is also true of many non-satellite urban LUR models (Hoek et al., 2008; Hystad et al., 2011; Novotny et al., 2011; Vienneau et al., 2013). Also, our small data set prevented us from holding out some of the data for an independent

evaluation (Hoek et al., 2008; Novotny et al., 2011; Wang et al., 2012), which has also been the case in other studies with limited monitoring data (e.g. Hystad et al., 2011). We instead used five-fold cross-validation to estimate our models' prediction error. We note that our R^2 values may be higher than would be observed with a new validation data set due to the relatively small number of monitoring sites and large number of predictors (Basagaña et al., 2012; Wang et al., 2012). Finally, we focussed on generating monthly and annual averages rather than daily estimates (e.g. Lee and Koutrakis, 2014). This enabled us to examine all of Australia, rather than a specific region or city.

Satellite-based LUR models hold promise for improving exposure assessment in epidemiological studies and have a diverse range of potential applications. They are particularly useful in locations with sparse or absent ground-based monitoring. Our models included both variables within different buffers (e.g. percent land use type, road length) and point variables (e.g. satellite NO_2, distance to coast). They permit unique predictions for a given set of input points (e.g. residential addresses), and we also presented an example of predictions on a 100 m grid across Australia (Figure 1). Our models are capable of capturing within-urban variability in concentrations, and although we did not aim to capture highly localised effects the models may also be able to capture some near-source (e.g. roads, industry) variability in certain areas (Hoek et al., 2008; Marshall et al., 2008). Because the models spanned the entire country there were no limitations around city-to-city transferability. Our models are the first that we are aware of to offer national coverage of Australia, and add to the growing international evidence regarding the utility of satellite-based LUR.

In summary, our satellite-based LUR models were able to capture ~80% of spatial variability in monthly and annual ambient NO_2 concentrations during 2006-11 across Australia, a country with sparse ground-level monitoring. These models can be used to determine concentrations that

individuals are exposed to at their residential address, or for larger spatial units (e.g. post code or suburb level) if their address is unknown due to confidentiality restrictions. They can also be used to refine and validate estimates of population-level exposures. With these applications in mind we are making our model predictions freely available to those who want to use them for research.

ACKNOWLEDGEMENTS

Please contact the corresponding author if you would like to use the model predictions described in the paper. LDK acknowledges an NHMRC Early Career (Australian Public Health) Fellowship (APP1036620). This material is partially based on work supported by the United States National Science Foundation under Grant No. 0853467. Computational resources and services used in this study were provided by the High Performance Computer and Research Support Unit, Queensland University of Technology, and the Research Computing Centre, The University of Queensland. Analyses and visualisations used in this study were produced with the Giovanni online data system, developed and maintained by the NASA GES DISC. We acknowledge the Aura mission for the production of the data used in this research effort, and the Netherlands Agency for Aerospace Programs in collaboration with the Finnish Meteorological Institute through their contribution to the Aura mission via the Ozone Monitoring Instrument. We thank the state and territory authorities in Australia that freely provided NO_2 monitoring data. We also acknowledge the free use of data provided by the Global Land Cover Facility, NOAA National Geophysical Data Centre, Australian Bureau of Meteorology, Australian Bureau of Statistics, and Australian National Pollutant Inventory. Roads and airports data were purchased from the Public Sector Mapping Agencies (Australia). Elevation data were purchased from Geoscience Australia.

REFERENCES

Alfons A. cvTools: Cross-validation tools for regression models. R package version 0.3.2. Available online: http://CRAN.R-project.org/package=cvTools (accessed June 10, 2014); 2012

Australian Bureau of Statistics. Australian Statistical Geography Standard (ASGS): Volume 1 – Main Structure and Greater Capital City Statistical Areas, Australia. Available online: http://www.ausstats.abs.gov.au/ausstats/subscriber.nsf/0/D3DC26F35A8AF579CA257801000DCD7D/$File/1270055001_july%202011.pdf (accessed June 10, 2014); 2011

Australian Bureau of Statistics. Year Book Australia, 2012. Available online: http://www.ausstats.abs.gov.au/ausstats/subscriber.nsf/LookupAttach/1301.0Publication24.05.121/$file/13010_2012.pdf (accessed 10 June, 2014); 2012

Australian Bureau of Statistics. Regional Population Growth, Australia, 2012. Available online: http://www.abs.gov.au/ausstats/abs@.nsf/Products/3218.0~2012~Main+Features~Main+Features?OpenDocument (accessed June 10, 2014); 2013a

Australian Bureau of Statistics. Australian Social Trends, April 2013. Available online: http://www.abs.gov.au/AUSSTATS/abs@.nsf/Lookup/4102.0Main+Features30April+2013 (accessed June 10, 2014); 2013b

Australian Bureau of Statistics. Mesh Block Counts. Available online: http://www.abs.gov.au/websitedbs/censushome.nsf/home/meshblockcounts (accessed June 10, 2014); 2013c

Barnett, A.G.; Williams, G.M.; Schwartz, J.; Neller, A.H.; Best, T.L.; Petroeschevsky, A.L.; Simpson, R.W. Air Pollution and Child Respiratory Health: A Case-Crossover Study in Australia and New Zealand. Am J Respir Crit Care Med. 171:1272-1278; 2005

Basagaña, X.; Rivera, M.; Aguilera, I.; Agis, D.; Bouso, L.; Elosua, R.; et al. Effect of the Number of Measurement Sites on Land Use Regression Models in Estimating Local Air Pollution. Atmos Environ. 54:634–642;2012

Bechle, M.J.; Millet, D.B.; Marshall, J.D. Remote sensing of exposure to NO2: Satellite versus ground-based measurement in a large urban area. Atmos Environ. 69:345-353; 2013

Beelen, R.; Hoek, G.; Fischer, P.; Brandt, P.A.v.d.; Brunekreef, B. Estimated long-term outdoor air pollution concentrations in a cohort study. Atmos Environ. 41:1343-1358; 2007

Briggs, D.J. The use of GIS to evaluate traffic-related pollution. Occup Environ Med. 64:1-2; 2007

Briggs, D.J.; Collins, S.; Elliott, P.; Fischer, P.; Kingham, S.; Lebret, E., et al. Mapping urban air pollution using GIS: a regression-based approach. International Journal of Geographical Information Science. 11:699 - 718; 1997

Friedman J.; Hastie, T.; Tibshirani, R. Regularization Paths for Generalized Linear Models via Coordinate Descent. J Stat Softw. 33:1–22;2010.

Hart, J.E.; Yanosky, J.D.; Puett, R.C.; Ryan, L.; Dockery, D.W.; Smith, T.J, et al. Spatial Modeling of PM_{10} and NO_2 in the Continential United States, 1985-2000. Environ Health Perspect. 117:1690-1696; 2009.

Hoek, G.; Beelen, R.; de Hoogh, K.; Vienneau, D.; Gulliver, J.; Fischer, P.; Briggs, D. A review of land-use regression models to assess spatial variation of outdoor air pollution. Atmos Environ. 42:7561-7578; 2008

Hoffmann, B.; Moebus, S.; Möhlenkamp, S.; Stang, A.; Lehmann, N.; Dragano, N., et al. Residential Exposure to Traffic Is Associated With Coronary Atherosclerosis. Circulation. 116:489-496; 2007

Højsgaard, S.; Halekoh, U.; Yan J. The R Package geepack for Generalized Estimating Equations. J Stat Softw. 15/2:1–11; 2006.

Hystad, P.; Setton, E.; Carvantes, A.; Poplawski, K.; Deschenes, S.; Brauer, M., et al. Creating National Air Pollution Models for Population Exposure Assessment in Canada. Environ Health Perspect. 119:1123-1129; 2011

Jerrett, M.; Arain, A.; Kanaroglou, P.; Beckerman, B.; Potoglou, D.; Sahsuvaroglu, T., et al. A review and evaluation of intraurban air pollution exposure models. J Expo Anal Environ Epidemiol. 15:185-204; 2005

Johnson, M.; Isakov, V.; Touma, J. S.; Mukerjee, S.; Özkaynak, H. Evaluation of Land-Use Regression Models Used to Predict Air Quality Concentrations in an Urban Area. Atmos Environ. 44:3660–3668; 2010

Lamsal, L.N.; Martin, R.V.; van Donkelaar, A.; Steinbacher, M.; Celarier, E.A.; Bucsela, E., et al. Ground-level nitrogen dioxide concentrations inferred from the satellite-borne Ozone Monitoring Instrument. J Geophys Res. 113:D16308; 2008

Lamsal, L.N.; Martin, R.V.; van Donkelaar, A.; Celarier, E.A.; Bucsela, E.J.; Boersma, K.F., et al. Indirect validation of tropospheric nitrogen dioxide retrieved from the OMI satellite instrument: Insight into the seasonal variation of nitrogen oxides at the northern midlatitudes. J Geophys Res. 115:D05302; 2010

Lee, H.J.; Koutrakis, P. Daily Ambient NO2 Concentration Predictions Using Satellite Ozone Monitoring Instrument NO2 Data and Land Use Regression. Environmental Science & Technology. 48:2305-2311; 2014

Levelt, P.F.; Van den Oord, G.H.J.; Dobber, M.R.; Malkki, A.; Huib, V.; de Vries, J.; Stammes, P.; Lundell, J.O.V.; Saari, H. The ozone monitoring instrument. Geoscience and Remote Sensing, IEEE Transactions on. 44:1093-1101; 2006

Lim, S.S.; Vos, T.; Flaxman, A.D.; Danaei, G.; Shibuya, K.; Adair-Rohani, H., et al. A comparative risk assessment of burden of disease and injury attribuTable to 67 risk factors and risk factor

clusters in 21 regions, 1990–2010: a systematic analysis for the Global Burden of Disease Study 2010. Lancet. 380:2224-2260; 2012

Marshall, J.D.; Nethery, E.; Brauer, M. Within-urban variability in ambient air pollution: Comparison of estimation methods. Atmos Environ. 42:1359-1369; 2008

Novotny, E.V.; Bechle, M.J.; Millet, D.B.; Marshall, J.D. National Satellite-Based Land-Use Regression: NO_2 in the United States. Environmental Science & Technology. 45:4407-4414; 2011

Richter, A.; Burrows, J.P.; Nusz, H.; Granier, C.; Niemeier, U. Increase in tropospheric nitrogen dioxide over China observed from space. Nature. 437:129-132; 2005

Ritz, B.; Yu, F.; Fruin, S.; Chapa, G.; Shaw, G.M.; Harris, J.A. Ambient Air Pollution and Risk of Birth Defects in Southern California. Am J Epidemiol. 155:17-25; 2002

Rose, N.; Cowie, C.; Gillett, R.; Marks, G.B. Validation of a Spatiotemporal Land Use Regression Model Incorporating Fixed Site Monitors. Environmental Science & Technology. 45:294-299; 2011

Su, J.G.; Jerrett, M.; Beckerman, B. A distance-decay variable selection strategy for land use regression modeling of ambient air pollution exposures. Sci Tot Environ. 407:3890-3898; 2009

Tibshirani, R. Regression Shrinkage and Selection via the Lasso. J R Statist Soc B. 58:267-288; 1996

Vienneau, D.; de Hoogh, K.; Bechle, M.J.; Beelen, R.; van Donkelaar, A.; Martin, R.V., et al. Western European Land Use Regression Incorporating Satellite- and Ground-Based Measurements of NO2 and PM10. Environmental Science & Technology. 47:13555-13564; 2013

Vienneau, D.; de Hoogh, K.; Beelen, R.; Fischer, P.; Hoek, G.; Briggs, D. Comparison of land-use regression models between Great Britain and the Netherlands. Atmos Environ. 44:688-696; 2010

Wang, M.; Beelen, R.; Eeftens, M.; Meliefste, K.; Hoek, G.; Brunekreef, B. Systematic Evaluation of Land Use Regression Models for NO_2. Environmental Science & Technology. 46:4481-4489; 2012

Figure captions

Figure 1. Average NO_2 concentration in 2008 predicted at ~350,000 census mesh block centroids by the annual surface model. The inset focuses on the greater Sydney area, Australia's largest city (population ~4.4 million). The figure is displayed using a 100 m grid.

Figure 2. Selected percentiles of annual average NO_2 predicted at ~350,000 census mesh block centroids by the annual surface model, 2006-2011.