Assessing ozone exposure for epidemiological studies in Malmö and Umeå, Sweden

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HIGHLIGHTS

- We measured levels of ozone at 80 sites during three weeks.
- We modelled spatial variation in ozone in two areas of Sweden.
- We modelled the temporal variation in ozone levels in South of Sweden.
- We can, for Umeå, predict the spatial variability of ozone.
- We can predict the temporal variability of ozone in Malmö.

ABSTRACT

Ground level ozone [ozone] is considered a harmful air pollutant but there is a knowledge gap regarding its long term health effects. The main aim of this study is to develop local Land Use Regression [LUR] models that can be used to study long term health effects of ozone. The specific aim is to develop spatial LUR models for two Swedish cities, Umeå and Malmö, as well as a temporal model for Malmö in order to assess ozone exposure for long term epidemiological studies. For the spatial model we measured ozone, using Ogawa passive samplers, as weekly averages at 40 sites in each study area, during three seasons. This data was then inserted in the LUR-model with data on traffic, land use, population density and altitude to develop explanatory models of ozone variation. To develop the temporal model for Malmö, hourly ozone data was aggregated into daily means for two measurement stations in Malmö and one in a rural area outside Malmö. Using regression analyses we inserted meteorological variables into different temporal models and the one that performed best for all three stations was chosen. For Malmö the LUR-model had an adjusted model $R^2$ of 0.40 and cross validation $R^2$ of 0.17. For Umeå the model had an adjusted model $R^2$ of 0.67 and cross validation adjusted $R^2$ of 0.48. When restricting the model to only including measuring sites from urban areas, the Malmö model had adjusted model $R^2$ of 0.51 (cross validation adjusted $R^2$ 0.33) and the Umeå model had adjusted model $R^2$ of 0.81 (validation adjusted $R^2$ of 0.73). The temporal model had adjusted model $R^2$ 0.54 and 0.61 for the two Malmö sites, the cross validation adjusted $R^2$ was 0.42. In conclusion, we can with moderate accuracy, at least for Umeå, predict the spatial variability, and in Malmö the temporal variability in ozone variation.

1. Introduction

1.1. Ozone and health

Ozone is present in the stratosphere but also in the troposphere (ground level up to 10 km) where it can be detrimental to health. Ground level ozone [hereafter referred to as ozone] is due to its
reactive nature a potent oxidant and thus harmful to human health, vegetation and materials. It is one of the most harmful air pollutants in Europe today (EEA, 2011). Ozone can enter the lung and due to its reactive nature cause oxidative stress (Mudway and Kelly, 2004; Koike and Kobayashi, 2004; Sousa et al., 2013). There is evidence that ozone can cause both acute and chronic effects to human health (OECD, 2008; WHO, 2013). There are several studies on the short term effect of ozone on respiratory diseases and mortality (Sousa et al., 2013; WHO, 2008a,b). There is less evidence on long term effects due to lack of accurate exposure data. However some data suggest that there is a long term effect of ozone on respiratory mortality as well and that the effects of ozone have a very low threshold (Brunekreef et al., 2012). It has been suggested that for conducting long term health effects studies of ozone it is crucial to know the local spatial variation to reduce exposure misclassification (WHO, 2008a,b). Although large parts of the ozone measured in Sweden are generated elsewhere, there is a regional and local variation in concentrations. The regional variation of ozone is well known, while the local variation is much less studied (WHO, 2008a,b; SMHI, 2010). WHO have set a health based limit for ozone with a maximum 8-h mean concentration of 100 μg/m³ (WHO, 2008).

Recent studies have focused on the development of more precise exposure assessment models for primary pollutants such as nitrogen oxides [NOx] and particulate matter [PM] (Beelen et al., 2013; Noth et al., 2011; de Hoogh et al., 2013). Less focus has been put on developing models for ozone exposure assessment in epidemiological studies, even if the health risks of ozone exposure is considered as least as serious as primary pollutants (Brunekreef et al., 2012). It has been found that there is a need of ozone modelling since ozone exposure can vary substantially within the area covered by the measurement station (WHO, 2008a,b).

To summarize, there is an obvious and important need to investigate the spatial variation of ozone on the local scale to make it possible to study the long term health effects of ozone.

1.2. Ozone variation in space

Ozone is a secondary pollutant and is formed through chemical reactions including NOx, carbon monoxide [CO] and volatile organic compounds [VOC], collectively called ozone precursors, in the presence of sunlight. The formation of ozone acts over extensive areas, and levels of ozone might be caused by combustion related emissions and photochemical reactions thousands of kilometres away (WHO, 2008a,b). In urban areas on the other hand, emissions of pollutants can reduce ozone levels close to the source, such as road traffic areas (WHO, 2008a,b; Intergovernmental Panel on Climate Change, 2007). This can be explained by freshly emitted nitrogen monoxide [NO] which reacts with ozone and converts it into nitrogen dioxide [NO2] and oxygen [O2]. The implication is that ozone near roadside is scavenged often referred to as the urban decrement (Stedman and Kent, 2008). Ozone precursors can also be carried away from the source and also cause ozone formation in the downwind plume from the city (WHO, 2008a,b; Intergovernmental Panel on Climate Change, 2007). In Europe emissions of ozone precursors have decreased with about 30% from 1990 to 2004, however levels of ozone have not decreased as much (García et al., 2010). An international review project has concluded that there is strong evidence that background ozone concentrations in the northern hemisphere have instead increased by up to 10 μg/m³ per decade over the last 20–30 years (Raes and Hjorth, 2006). Also in many Swedish cities, the trend over the last years is that background ozone is slightly increasing which can partly be explained by a change in the composition of vehicle exhaust (Johansson and Forsberg, 2005). However, episodes of high levels of ozone seem to be fewer and less severe (Jenkin, 2008).

The ozone processes are not only affected by traffic and other sources of combustion gases. During the growing season plants emit highly reactive hydrocarbons, such as isoprene or terpenes, especially when temperatures are higher (Sanderson et al., 2003). These hydrocarbons are so called biogenic VOCs [hereafter referred to as BVOCs] that increase ozone formation. However, in the Scandinavian region it is common with certain tree types that emit BVOCs that could have an opposite effect and decreasing levels of ozone through ozonolysis, i.e. ozone causing the breakage of molecules thru oxidation (Curci et al., 2009).

Spatially, altitude could also be a predictor for ozone concentrations, at lower altitudes ozone levels tend to be lower due to local destruction of ozone with NO (titration) and dry deposition. Whereas in higher altitudes and in coastal regions winds tend to be stronger and there is no formation of shallow boundary layers which affects the atmospheric mixing rates of pollutants (Stedman and Kent, 2008).

To summarize, ozone formation are depending on ozone precursors, mainly emitted by traffic. Near roadside however, traffic exhaust can lower levels of ozone. BVOCs can contribute to both formation and destruction of ozone.

1.3. Ozone variation in time

Ozone has an annual variation with a typical spring maximum due to increased solar radiation (Monks, 2000). Ozone has a daily pattern driven by levels of ozone precursors and solar radiation and tends to build up and peak in the afternoons (García et al., 2010).

2. Aim and outline for this study

The main aim of the study is to develop a Land Use Regression [LUR] model that can be used in epidemiological studies on health effects of ozone exposure. The specific aim is to describe the local spatial variation of ozone in two Swedish cities (Malmö and Umeå) with a LUR model. To be able to use this model in other time windows than the studied period adjustments can be made with stationary measurements. But this data have continuously short periods of missing data. In addition we will therefore also develop a model accounting for temporal variations based on meteorological data (only Malmö) in order to assess ozone levels when there are missing data from the stationary measurements of ozone.

3. Method

3.1. Study areas

We selected two study areas in different parts of Sweden for the measuring campaigns; the Malmö region in the south and the Umeå region in the north of Sweden with a north–south distance of 1250 km between the two areas (Fig. 1). In the Malmö area we included the city of Malmö with surroundings (Fig. 2), with approximately 300 000 inhabitants in the area. The Umeå region includes the city of Umeå and nearby villages with about 150 000 inhabitants. The meteorological definition of spring is when there is a daily temperature above 0 °C during 7 executive days. This period is expected to arrive in the middle of February in the Malmö area and in the middle of April in the Umeå area. However, during the studied year 2012 spring arrived in late February in Malmö and in late March in Umeå. Summer (above 10 °C as a daily temperature for 5 executive days) arrived in late April in Malmö (expected middle of May) and in late May in Umeå (expected beginning of June) (SMHI, 2013). Although there is no strong north–south
gradient in the average ozone levels, episodes with very high levels of ozone are more frequent in the southern parts of Sweden (SMHI, 2010). The Malmö area has a more traffic and thus higher levels of ozone precursors in comparison with the Umeå area.

3.1.1. Measurements for the LUR-model

The week of the measurements was coordinated between the two areas to get as comparable data as possible. The ozone measurements were performed as weekly averages at 40 sites in each study area, during three seasonal measuring periods in 2012; Spring: 16th–24th of April Summer: 28th of May–4th June and Autumn: 20th–27th August.

The measurement stations were categorized, based on their location, as: regional background, urban background and traffic sites according to the criteria by EUROAIRNET (1999). The criteria say that a regional background site is located in an area which is not continuously built up and without influence of nearby industry or traffic. An urban background site is located in a continuously built up area but not heavily influenced by nearby traffic or industry. A traffic site is located so their pollution level is mainly influenced by emission from a nearby road. The distribution of sites between these three classes should also represent the distribution of the population, with most of the sites located in urban background areas and a smaller part in rural areas and in areas heavily influenced by traffic (Table 1). We later on had to exclude one site in Malmö and one in Umeå due to measurement errors.

The Ogawa diffusive sampler (Ogawa & Company, Pompano Beach, FL, USA) was used for all measurements. The sampler consists of a Duralon cylinder containing two nitrite-coated filters. The coated filters were supplied by the manufacturer (Ogawa). Ambient ozone oxidizes nitrite [NO$_2^-$] on filters to nitrate [NO$_3^-$]. The filters are extracted and the accumulated NO$_3^-$ is analysed by ion chromatography. The time-weighted average ozone concentrations for one-week measurement are estimated by the amount of NO$_3^-$ on the filter, the uptake rate of the sampler, and the exposure time. A comparison between parallel measurements with the reference ozone method (UV-monitors) and the Ogawa samplers during eight sets of replicate measurements in Umeå 2011 showed good precision of six samplers (RSD = 1–4%) and the uptake rate was determined to 27.3 ml/min (Hagenbörk-Gustafsson, 2012).

The samplers were placed inside an Ogawa rain and wind shelter at approximately 2–3 m from the ground. All samples were analysed at the Division of Occupational and Environmental medicine, Umeå University, Umeå. Two field blanks were deployed for each study site and at each sampling period. These blanks were run together with the samples, and the blank signal in each run was subtracted from the samples. The ozone concentrations obtained with the Ogawa samplers were compared to continuous measurements by active UV- absorption monitors at two sites included in the Regional Air Quality Monitoring in Malmö during two of the measurement periods (Table 2).

3.1.2. Land use regression model

Briefly, the basis of developing a LUR-model is to collect a large number of digitalized geographical variables, such as road networks and land use, surrounding each of the measuring sites and to include these into a Geographical Information Systems [GIS]. The collected variables should previously be known to have an impact on the pollutant of interest. LUR is a statistical approach to model air pollution, by trying to explain the variation in measured levels between different sites based on differences in geographical variables, hereafter referred to as predictor variables. Levels of pollution may then be predicted for any location, such as individual homes, using the parameter estimates derived from LUR-model. GIS analyses were conducted to derive the values for the predictor variables at each monitoring site.

3.1.3. Predictor variables

We used ArcGIS version 10 (ESRI) to extract the predictor variables. The geographical coordinates of each measurement site were obtained using GPS and checked using digital maps. Around each measurement station we created buffers. The buffer zones used to study influence from traffic were: 25, 50, 100, 300, 500 and 1000 m.
to correspond for both near source emissions and larger areas of traffic density. We intersected the buffers with a digital road network holding information on traffic intensity attached. The digital road network was developed by the Swedish Transport Agency from 2010 for both areas, complemented with road data from the local municipalities. With this data it was possible to collect total traffic load within each buffer zone (calculated as: \(\text{[length of the road segment]} \times \text{[traffic intensity]}\)/road segment for all roads within a buffer zone). In Malmö this was also done separately including only heavy (such as trucks and buses) traffic.

### Table 1

| City     | Site type       | N  | Mean | SD  | Min | Max |
|----------|-----------------|----|------|-----|-----|-----|
| Malmö    | Regional background | 7  | 69.1 | 5.9 | 60.4| 79.0|
|          | Urban background | 15 | 66.7 | 3.2 | 59.9| 73.0|
|          | Traffic         | 18 | 68.4 | 8.0 | 60.0| 83.1|
| Umeå     | Regional background | 5  | 54.9 | 18.3| 27.5| 93.3|
|          | Urban background | 23 | 52.5 | 14.2| 29.4| 71.8|
|          | Traffic         | 12 | 49.3 | 13.5| 26.5| 69.7|

### Table 2

| Measurement site by type and period | Active sampling (µg/m³) 28th of May–4th of June | Passive sampling (µg/m³) 28th of May–4th of June | Active sampling (µg/m³) 20th–27th of Aug | Passive sampling (µg/m³) 20th–27th of Aug |
|------------------------------------|--------------------------------------------------|-------------------------------------------------|------------------------------------------|------------------------------------------|
| Urban Background                   | 73.3                                             | 71.5                                            | 64.6                                     | 68.6                                     |
| Traffic                            | 65.6                                             | 67.6                                            | 57.2                                     | 59.5                                     |

Fig. 2. Study areas and mean ozone concentration at the measuring sites.
The same calculations were also done only including major roads (>5000 vehicles/24 h). Traffic intensity and distance to the nearest road and the nearest major road and other traffic variables was also calculated.

The buffer zones used to study the influence of land use were: 100, 300, 500, 1000 and 5000 m. Within each buffer zone, areas of different land cover were calculated in m². Land use data was collected from the EU initiated Coordination and Information on the environmental programme database [CORINE] from 2006 for Malmö area and 2000 for Umeå area. It compromises 44 land cover types and was regrouped into 6 classes (High density residential areas, Low density residential areas, Industry, Port, Urban Green and Semi natural and forested areas [the latter two hereafter referred to as Natural]). In addition we used locally derived population density data for 2009 in Malmö. In Umeå the population data was derived from Statistics Sweden for the year 2008. We also calculated distance to the sea and altitude.

3.1.4. Model development

The predictor variables are combined in a supervised step wise linear regression model to try to find the combination of variables explaining as much of the spatial variation in pollution levels as possible. We developed a mean of ozone for the three measurement periods (spring, summer and autumn), which were used in the analyses. Regression models that maximises the $R^2$ values, adjusted percentage explained variance were developed to explain the variance in ozone concentration at different sites. First, univariate regression analyses for all predictor variables were conducted. The variable with the highest $R^2$ was chosen as the start model and then the remaining variables were added consecutively. The predictor variables with the highest additional increase in $R^2$ were maintained if they also increased in adjusted $R^2$ more than 1%. If a variable had buffer sizes that were overlapping in the model, the variable was rewritten (and analyses redone) using concentric adjacent rings (so called doughnut rings). Diagnostic tests were applied to the models such as; multicollinearity between included variables by Variance Inflation Factors and influential observations by Cook’s D and QQ-plots were investigated to verify normality assumptions of the residuals. The statistical procedure is further described in the ESCAPE exposure manual (http://www.escapeproject.eu/manuals/ESCAPE_Exposure-manuale9.pdf) and in (Beelen et al., 2013). Depending on the performance, the final model can then be used to predict the levels of ozone at any given location within the area for which the model was developed, by collecting and combining the geographical variables according to the model estimates. There was one major difference in our model building compared to the ESCAPE model; we chose not to have a predefined direction of effect in the model building as ozone is a secondary pollutant involved in many chemical reactions and for all variables we had no clear a priori defined direction of effect. For each area, Malmö and Umeå analyses were conducted separately resulting in local specific predictor variables. The statistical analyses were performed using R (version 2.13.0).

As additional analyses we developed a separate LUR-model, excluding the predefined regional background sites, to see if the fit improved when only including measuring sites in areas where NOx emissions are higher and more local ozone reactions takes place. In the analyses we excluded 12 stations from Umeå and 7 for Malmö and these models are referred to as the urban models.

3.1.5. Model validation

We validated with leave-one-out cross validation, in which a model is developed for all sites and the model structure (with the predictor variables) are applied to n-1 (where n is the number of sites).

3.1.6. Measurements for the temporal model (only Malmö)

Hourly data from 3 measurement stations that continuously measure levels of ozone were collected and aggregated into daily means. The data was collected from one urban background site and one traffic site in Malmö and one regional background site (situated 50 km north of Malmö). The measurement method was active UV-absorption. We used data for the same 3 weeks as in the LUR-analyses and had thus 21 daily measurements from each of the three stations. Meteorological data are collected from the Swedish Meteorological and Hydrological Institute [SMHI] site Heleneholm in Malmö.

3.1.7. Regression analyses for the temporal model (only Malmö)

To investigate the important variables for predicting ozone variations in time, meteorological variables and ozone levels were inserted into a multiple linear regression model. The model that performed best for all three stations was chosen. The meteorological variables were: wind speed, wind direction, wind vector, global radiation, net radiation, temperature, vertical temperature difference (between 2 and 8 m) and vertical temperature difference (between 24 and 8 m). The latter two variables are measures of temperature inversions which can affect atmospheric convection rates.

3.1.8. Validation for the temporal model (only Malmö)

The model was validated by selecting 4 random weeks of daily measurements from the urban background station between the years of 2000 and 2012. The same model (with the same predictor variables) was applied to those measurements. The adjusted $R^2$ for these analyses were used as a validation $R^2$.

4. Results

4.1. Measurements LUR-model

Higher levels of ozone were reported from the Malmö area than the Umeå area. Levels were also highest during spring than during summer and autumn, which is in accordance to cities of this magnitude. The mean levels of ozone for the entire study period in Malmö were 68 µg/m³ (range 60–83 µg/m³) and in Umeå 52 µg/m³ (range 36–63 µg/m³).

4.2. LUR-model

For Malmö the best LUR-model included the predictor variables:

- low density residential areas within a 300 m buffer,
- urban green areas within a 100 m buffer,
- traffic load within a 1000 m buffer and
distance to sea.

All variables had a negative direction of effect which means that the variables explained the ozone decay. This model gave an adjusted $R^2$ of 0.40. The correlations between variables were moderate ($r < 0.4$). The model was validated with “leave one out validation” and had a cross-validation adjusted $R^2$ of 0.17. Levels of ozone in Malmö had a smaller variation than Umeå (Table 1) and this can be a reason why the LUR-model explained less of the variation.

The urban model for Malmö included the following predictor variables:

- low density residential areas within 100 m buffers,
- urban green areas within 100 m buffers and
- heavy traffic load within 1000 m buffers.
All variables had a negative direction of effect and the model adjusted $R^2$ was 0.51 with cross validation adjusted $R^2$ of 0.33. For Umeå, the model that performed best was a model including the following predictor variables:

- traffic load within a 300 m buffer,
- high density residential areas within a 5000 m buffer,
- urban green areas within 500 m buffers and natural areas in a 5000 m buffer.

All directions of effects were negative except urban green areas which explained some of the increase in ozone and the model performed with an adjusted $R^2$ of 0.67. The correlations between variables were high, $(r < 0.6)$, and the model had a cross-validation adjusted $R^2$ of 0.48.

In the urban model for Umeå the following predictor variables were included:

- traffic load within 300 m buffers,
- natural areas within 500 m buffers,
- population density within 5000 and population density within 1000 m buffers.

This model gave an adjusted $R^2$ of 0.81 and cross validation $R^2$ of 0.73. Population density and natural areas had positive and traffic load negative direction of effect. Each variables contribution to the model building is summarized in Table 3 with more in-depth data and residuals in Supplemental materials.

### 4.3. Temporal model for Malmö

The temporal model was the model that best fitted all the sites in the Malmö region. The predictor variables included in the temporal model for best fit were:

- vertical difference in temperature between 24 m and 8 m, net radiation and global radiation.

For the urban background station the adjusted $R^2$ was 0.54, for the traffic site the adjusted $R^2$ was 0.61 and for the regional background site (situated 50 km from Malmö) adjusted $R^2$ was 0.22.

The factors included in the temporal model seem to be factors that are important for local production of ozone such as solar radiation variables (net radiation and global radiation), rather than for in transport such as wind variables. The variables also seem to predict better at the traffic site, where local ozone reaction is higher, than in the urban background site where local ozone production is non-existent due to the lack of ozone precursors, the model performs poorly. When validating the temporal model we used four randomly selected weeks between the years 2000 and 2012 and the model seems to works even in this extended time span with an adjusted validation $R^2$ of 0.42. Thus the model showed that levels of ozone could be predicted by knowing meteorological factors. However the temporal model will not be applicable for the winter season since this period was not studied, yet ozone levels is not of a health concern in this region during winter due to low solar radiation.

### 5. Discussion

#### 5.1. Previous studies

The LUR-model approach is a way of assessing exposure intended for use in epidemiological studies with increased spatial resolution. The intention is to have a better exposure assessment than merely attributing exposure levels from nearest measurement stations which can be several kilometres away. LUR have mostly been used for primary pollutants to explain the relationship between measured levels and different land uses (Beelen et al., 2013; Eeftens et al., 2012; Noth et al., 2011; de Hoogh et al., 2013) and was originally initiated by Briggs et al. (1997). To put our results into perspective Gulliver et al. (2011) considered model $R^2$ of 0.47 to be an acceptable level when they modelled PM$_{10}$ using LUR. Ryan and LeMasters (2007) found $R^2$ in the range of 0.54–0.81 in a review of 12 LUR models for primary pollutants. Hoek et al. (2008) found most $R^2$ of NO$_x$ to be around 0.6–0.7. Beelen et al. (2009) developed a background ozone model for most of Europe (not Sweden) using annual concentration and found $R^2$ for the LUR models between 0.38 and 0.54. Also, applying meteorological variables to explain ozone variation by multiple linear regression analyses have been done previously with $R^2$ of 0.83 (Gvozdic et al., 2011).

A previous study in Sweden found that the urban emissions of NO$_x$ and VOCs do not affect the levels of ozone outside the cities (SMHI, 2010). This study (SMHI, 2010) also showed that high levels of ozone in Sweden tend to come from sources outside of Sweden. Another study conducted at the Swedish west coast showed regional background levels of ozone to be relatively similar to levels measured in urban background, while the levels at heavy traffic sites were considerably lower (Piikki et al., 2008). Decreasing levels of ozone with increasing levels of vehicle exhausts were also seen in the same study when comparing two housing areas situated at different distances from heavy traffic (Piikki et al., 2008). In accordance to this, our results showed that traffic lowered the levels of ozone, and that the model fit improved dramatically in the urban models. The extent of residential areas can also be a measure for urbanity and higher levels of NO$_x$-emissions and thus went into the direction of lowering levels of ozone. All measures of residential areas had in accordance negative effects. In contrary all measures of population density went in the opposite directions with positive direction of effects. In the area population density is not closely linked to traffic density and people tend to live outside the commercial city centre where traffic is dense.

### Table 3

| Predictor variables (buffer) | Adj $R^2$ | Predictor variables (buffer) | Adj $R^2$ |
|-----------------------------|-----------|-------------------------------|-----------|
| Traffic load (300)          | 0.44      | Traffic load (300)             | 0.67      |
| High density Residential areas (5000) | 0.52 | Natural areas (5000)           | 0.75      |
| Natural areas (5000)        | 0.61      | Population density (5000–1000) | 0.79      |
| Urban green areas (5000)    | 0.63      | Population density (1000)      | 0.81      |
| Low density residential areas (300) | 0.19 | Low density residential areas (100) | 0.13      |
| Distance to sea             |           | Urban green areas (100)        | 0.26      |
| Urban green areas (100)     |           | Heavy traffic load (1000)      | 0.51      |
5.2. Modelling experiences

The full Malmö LUR-model performed poorer than the urban LUR-model and corresponding Umeå models. The contribution from the long distance transported ozone is the most likely explanation for the relatively poor LUR-model fit in Malmö. Another possible explanation could be that it was hard to find a good LUR-model since the variation of ozone was smaller in Malmö than in Umeå. The Malmö urban area model performed better than the full model.

One finding that could be investigated further was the variety of ozone levels in Natural areas (Fig. 2). We found low levels of ozone in a regional background station located in a near urban forested area, which can be due to dry deposition to the trees. Another regional background site in the Malmö area was located under a wind power plant situated in an agricultural field which, especially during spring, had high levels of ozone which could not be attributed to higher general wind speed (and thus causing measurement errors by an excess of air generated by movement from the wind power plant entering the sampler) during that period.

We also found that the predictor variable Natural areas both increased and decreased the levels of ozone in Umeå, depending on full or urban model. In the urban model it increased the ozone levels. In the full model it decreased while urban green areas increased levels of ozone. However, in Malmö “urban green areas” decreased the levels of ozone in the urban LUR-model. The role of BVOCs in the local formation of ozone formation in urban areas has been studied recently and it has been shown that contribution can be of great impact but that it is largely dependent on: meteorological factors, if there are high levels of ozone precursors such as NOx present, as well as type of tree species and thus which kind of BVOCs are emitted (Curci et al., 2009; Oderbolz et al., 2013). An example of the latter is that it has been previously found that green areas in Scandinavia lowered levels of ozone due to the ozonolysis of the BVOC terpenes commonly found in Scandinavian forests (Curci et al., 2009). This is especially true in areas with a lot of Betula which have highly reactive terpenes (Oderbolz et al., 2013). In the Umeå area Birch trees from the family Betulaceae is very common and the city is even known as the city of birches. A suggestion for future studies would be to carefully consider regional background sites and surrounding vegetation and to have more detailed information on the BVOC related variables. Altitude did not have any predicting value for our LUR-models even if it can be a predictor in other areas where variation in altitude is larger.

5.3. Limitations

We validated our spatial models using leave one out cross validation. This is the most common validation for LUR-models and standardised in the ESCAPE project (Beelen et al., 2013). It should however be mentioned that this validation method has been criticised for overestimating the predictive ability of LUR-models (Wang et al., 2013). Another possible caveat is the timing of measurements. We decided to measure simultaneously in spring, summer and autumn at both study sites. It can be argued that instead of measuring simultaneously we should have measured according to when the areas are equally green due to spring coming later in the North than in the South. The two studied areas are situated geographically apart (1250 km) and seasons can vary substantially in time. However, in the studied year the spring and summer season in Umeå came earlier than usual which minimized this error. As we developed separate models it should not influence results either. The benefits from using simultaneous measurements should however exceed the limitations as we are studying time periods with similar large scale weather patterns. It should also be noted that we used a standardized model (with the exception that we had no predefined direction of effect), ESCAPE, which was originally developed for primary pollutants. One caveat using this, is that we had very coarse predictor variables for natural areas (all fields and forests lumped into one variable) and thus do not capture fully the role of different BVOCs. There were also pros and cons of using a predefined direction of effect in the model building. We choose not to have a predefined direction of effect as we wanted to explore in what direction each variable went. We did however find that variables went into the direction of effect that could be expected based on previous knowledge.

Another possible limitation is that we used land use data from CORINE for 2000 in Umeå and 2006 in Malmö and it can be argued that the land use might not always be accurate for 2012. By visiting the sites to place and collect the samplers we could at least to some extent make sure that no major change in land use had been done in the vicinity of the measuring sites. The models were built separately for the two areas; using different years of CORINE-data should therefore not have influenced the results.

6. Conclusion

We studied the spatial variation of ozone at two heterogeneous cities (Malmö and Umeå) in Sweden using LUR-models. The LUR-model in Umeå had the best fit with adjusted model R² values of 0.67 whereas the LUR-model in Malmö had adjusted model R² of 0.39. When restricting the models to only urban settings we had a better fit, here the Malmö model had adjusted R² of 0.51 and Umeå a R² of 0.81. A temporal model was also developed for Malmö which performed best in a traffic site with adjusted R² of 0.61. To summarize, this study shows that it is possible to predict the spatial variability of levels of ozone, at least for Umeå. This means that we can assess exposure in Umeå. In order to have exposure data for other time periods than the modelled period we can build our LUR-model using input data from stationary measurements but careful consideration has to be done in order for input data for GIS-analyses still being accurate. We could also predict the temporal ozone variation, adjusted R² of 0.54–0.61 in the Malmö area. Thus, in shorter time periods we need stationary data on ozone levels we can use meteorological data to estimate these levels. Our results should be put in the context of our cities being considerable small and levels of ozone precursors moderate.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.05.038.
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