Source-Receptor and Inverse Modelling to quantify urban PARTiculate emissions (SRIMPART)

Bruce Denby, Matthias Karl, Herdis Laupsa, Christer Johansson, Mia Pohjola, Ari Karppinen, Jaakko Kukkonen, Matthias Ketzel, Peter Wåhlin

TemaNord 2009:552

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ISBN 978-92-893-1906-5

Printed on environmentally friendly paper
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Nordic Council of Ministers
Store Strandstræde 18
DK-1255 Copenhagen K
Phone (+45) 3396 0200
Fax (+45) 3396 0202

Nordic Council
Store Strandstræde 18
DK-1255 Copenhagen K
Phone (+45) 3396 0400
Fax (+45) 3311 1870

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Contents

Abstract

One of the major emission sources of primary $PM_{2.5}$ in Nordic countries such as Norway, Sweden, Denmark and Finland is wood burning for domestic heating. In Norway alone it is estimated that 80% of $PM_{2.5}$ is emitted through this source. Though direct measurements of wood burning emissions are possible under controlled conditions, emission inventories for domestic heating are difficult to calculate. Emissions vary from stove to stove as well as wood type, wood condition and burning habits. The consumption rate of wood burning is also strongly dependent on meteorological as well as societal conditions. As a result the uncertainty in wood burning emission inventories used in dispersion modelling is considered to be quite high.

As an alternative method for estimating the emissions resulting from wood burning for domestic heating this project makes use of ambient air measurements, chemical analysis of filter samples, receptor models, dispersion models, and simple inverse modelling methods to infer emission strengths. The methodology is applied in three Nordic cities, notably Oslo (Norway), Lycksele (Sweden) and Helsinki (Finland). In two of these cities (Oslo, Lycksele) daily filter samples over several months have been collected. The filter samples have been chemically analysed for a range of elemental and specific markers including OC/EC and Levoglucosan. The chemical analysis has been used as input for a range of receptor models, including UNMIX, PMF (ME-2), PMF-2 and COPREM. From these calculations the source contributions at the measurement sites, with particular emphasis on wood burning, have been estimated.

Such source apportionment studies provide source contributions at the receptor site only. To relate these to emissions, dispersion models are required. The receptor modelling is compared to dispersion models, using the existing emission inventories. This comparison of the dispersion models with the receptor models indicates, for example, that in Oslo and Lycksele the dispersion models overestimate the contribution from wood burning by a factor of 1.5–2. To further assess the differences between the receptor and dispersion modelling a simple inverse modelling technique, using multiple linear regression, is applied to the total $PM_{2.5}$ concentrations, measured at all monitoring stations, to assess the contribution of wood burning. The inverse modelling results have been found to agree with those from the receptor modelling for Oslo and Lycksele. Inverse modelling in Helsinki was inconclusive as no receptor modelling was available there fro comparison. Though both the receptor and inverse modelling point to an overestimation of the wood burning emissions of

$PM_{2.5}$ it is not possible to assign this solely to errors in the emissions inventory as dispersion model error can be significant.

An assessment of the uncertainty in the various methods is made. Uncertainty in the dispersion modelling is found to be of a similar order to the uncertainty in the wood burning emissions inventory and so no firm conclusions concerning the quality of the emissions inventories can be made. It was found that Levoglucosan as a wood burning tracer was important for the identification of the wood burning source. It is recommended to improve plume rise and urban canopy meteorological descriptions in the dispersion models before these models will be of sufficient quality to allow quantitative assessments of emission inventories.

1. Introduction

Fine particles, defined as particles with diameters < 2.5 μm ($PM_{2.5}$), have been associated with numerous adverse health effects (e.g., Pope et al., 2006). Based on the available scientific evidence on the health effects of particles, the European Union has set an Air Quality Directive (EC, 2008). The directive was supported by the Clean Air for Europe (CAFE) program, which estimated that fine particles cause over 300,000 premature deaths annually in Europe and that exposure to fine particles lowers the life-expectancy on the average by 8.6 months (Watkiss et al., 2005).

In order to reduce human exposure to these fine particles it is necessary to assess and understand the emission sources, the formation processes and the transport mechanisms leading to the ambient air concentrations of $PM_{2.5}$. There are a large number of sources of fine particulates and these include:

- Combustion sources such as traffic and industry
- Non-exhaust emissions from traffic such as brake wear, tyre wear and road wear
- Suspended particulate matter from agriculture, soil and other fugitive sources
- Building sites and cement industries
- Domestic and commercial heating sources such as oil and wood burning
- Emissions of aerosol precursors such as sulphur, nitrogen, ammonia and a range of organic compounds
- Non-anthropogenic sources such as sea salt, wind blown dust and wild-land fires

The contribution of these various sources differs both with time of year and with geographical position. For example, the secondary formation of aerosol particles is considered to be a major source in warmer climates during summer, whilst the contribution from traffic and domestic wood burning is a major source in colder climates during winter.

Generally the high concentrations in winter are due to both adverse meteorological conditions, atmospheric stagnation with strong inversions and low wind speeds, as well as the enhanced emissions from wood burning (Yttri et al, 2005) or suspended dust and salt from road traffic. This last is especially caused by the use of studded tyres and road salting activities (Aarnio et al., 2008; Larssen et al, 2007; Normann and Johansson, 2006; Tiitta et al., 2002, Larssen and Hagen, 1997). In regard to meteorology, Kukkonen et al. (2005) analysed 21 episodes involving substantially

high concentrations of PM_{10} from seven cities in six countries; they found that the best meteorological prediction variables were the temporal evolution of the temperature inversions and atmospheric stability and, in some of the cases, wind speed. Strong ground-based or slightly elevated temperature inversions prevailed in the course of the episodes for instance in Oslo and Helsinki, and their occurrence coincided with the highest PM_{10} concentrations.

The annual mean $PM_{2.5}$ concentrations in the Nordic cities are commonly relatively low compared with those in other major Central or Southern European cities, as can be seen in the reported values to the European Environment Agency (EEA, 2007; EMEP/CCC, 2007). However, high short-term levels (characteristically hours or a few days) of both PM_{10} and $PM_{2.5}$ concentrations are observed in Nordic countries in the winter. Substantially high concentrations of $PM_{2.5}$ are also commonly measured in the course of regional or long-range episodes of wild-land fire plumes in spring or summer (e.g., Aarnio et al., 2008; Saarikoski et al., 2007).

The significance of wood burning as a pollutant source on the European scale must not be underestimated. The most recent EMEP emissions database for 2004 estimates that 33% of the emissions of total carbon mass in Europe are the result of domestic wood burning (Kupiainen and Klimont, 2004). In the recent WHO report on ‘Health risks of particulate matter from long-range transboundary air pollution’ the relative contribution of domestic wood burning to $PM_{2.5}$ emissions are expected to increase from the 2000 levels of 25% to 38% in 2020.

Wood burning is known to be a significant source of fine fraction particle matter in a number of cities throughout Scandinavia and Europe. The major source of these particles is from domestic home heaters. Estimates of the contribution of domestic home heating to $PM_{2.5}$ emissions varies from city to city but Oslo has an estimated contribution of up to 80% from wood combustion (Larssen et al., 2005). In Denmark wood burning accounts for around 47% of all Danish $PM_{2.5}$ emissions (Palmgren et al. 2005). Cities such as Helsinki have less significant contributions with wood burning estimated to account for around 24% of the local emissions (Kauhaniemi et al., 2008). However, in smaller cities in Finland (e.g. Kuopio), Sweden (e.g. Lycksele and Gävle) and even Denmark (e.g. Gundsømagle) wood burning is estimated to be the dominating emissions source.

Current emission rates of PM from wood burning are based on estimates of consumption and the application of emission factors. These factors are taken from measurements of direct emissions from a variety of wood burning ovens (Sternhufvud et al., 2004; Finstad et al., 2004). Consumption can be calculated from surveys, from total wood consumption distributed over the population but also information concerning wood burning may be obtained from chimney sweepers. There is a large uncertainty in these estimates, for both the consumption of wood and the emission fac-

tors. Efforts to harmonise the consumption based emission factors within the Nordic countries were undertaken in a project funded by the Nordic Council of Ministers (NMR) that was running parallel to our project (Illerup et al. 2009).

Methods for estimating emissions of wood burning generally follow the above methodology. However, the actual emissions will depend greatly on local heating habits, the quality of stoves and wood as well as on meteorological conditions. Pragmatically, the only in situ way to assess the integrated emissions of PM from a large number of wood burning stoves is to measure the ambient PM concentrations in the field, and then use some inversion technique to infer the actual emissions. If only the relative contribution to ambient air concentrations of some source is to be determined then source-receptor modelling methods may be applied. This involves identifying compounds that are indicative of the particulate source, measuring these compounds at representative monitoring sites and then applying source-receptor modelling. These models allow, given a sufficient number of samples, the identification of source chemical profiles and by further analysis their relative contribution to the measured concentrations. Reviews of receptor modelling techniques can be found in e.g. Bruinen de Bruin et al. (2006), Watson and Chow (2004) and Hopke (1991). Such methods have already been successfully applied in many urban areas, e.g. Buzcu-Guven et al. (2007), Vallius et al. (2003) and Hueglin et al. (2000), to assign source contributions of both PM_{10} and $PM_{2.5}$. Examples of such models include PMF, Positive Matrix Factorisation (Paatero and Tapper, 1994; Paatero, 1997)) or COPREM, Constrained Physical Receptor Model (Wåhlin, 2003).

Knowing the relative contributions does not, however, provide information on the emission strengths. To determine this dispersion models are required that link the ambient concentrations to the emissions. On the simplest level, calculations using dispersion models can be compared to the receptor modelling at the receptor sites and the appropriateness of the emissions used in the dispersion modelling may be estimated. At a more complex level some form of inverse modelling may be applied at either the receptor site, where the receptor model is applied, or indeed at any measurement site. However, the urban environment is generally so complex and the number of observations so limited that most inverse modelling methods cannot be effectively applied to establish both the temporal and spatial distributions of emissions. Simpler inverse modelling techniques, using variational methods such as multiple linear regression (MLR), may be applied instead (Laupsa et al., 2008). These determine the optimal fit of the modelled source contributions, effectively providing scaling factors for the emission rates given the assumed spatial distributions.

The aim of this project is to determine if the emission rates of $PM_{2.5}$ from domestic wood burning can be estimated through indirect means, i.e. not through the estimation of emission factors and consumption rates.

These indirect methods include the use of ambient air measurements of $PM_{2.5}$, the chemical analysis of ambient air samples of $PM_{2.5}$, receptor modelling, dispersion modelling and inverse modelling. Integral to this aim is the need to determine the uncertainty of the methods. This is necessary if any meaningful comparison of the direct and indirect methods is to be carried out.

The indirect methods discussed above are applied in this study to three Nordic cities, where relevant data and modelling activities have been carried out. These are the cities of Oslo (Norway), Lycksele (Sweden) and Helsinki (Finland).

This report will firstly provide an overview of the selected case study cities and the available data and modelling carried out in these cities. It will then present the methodological elements required for the application of the indirect methods, including a description of the current emission inventories, receptor modelling, dispersion modelling and the inverse modelling. It will then present the results, looking at the individual elements of the methodology in regard to the cases studied and including an analysis of the uncertainties in the methods. It will then combine and compare these results to assess the results in regard to both methodology and location. It will finally discuss and conclude on the extent to which indirect methods can be applied and the further developments needed to improve on these.

2. Case study selection and description of sites

Two Nordic cities and one combination of two cities, where both measurement and modelling activities have been undertaken, are selected for this paper. These are Oslo (Norway), Helsinki (Finland) and Lycksele and Gävle (Sweden). In regard to the last the city of Gävle, this is only included in the study for the uncertainty assessment of the dispersion modelling as no measurements of $PM_{2.5}$ are available there. All of these cities have been found in previous studies (Hedberg et al., 2006; Krecl, et al. 2007; Krecl et al, 2008a; Krecl et al. 2008b; Kauhaniemi et al., 2008, Laupsa et al., 2008; Larssen et al. 2007) to have a significant proportion of their $PM_{2.5}$ contribution originating in domestic wood burning. A brief description is provided in this section of the cities, the study periods and the available data for use in this paper.

The cities considered in this study are located in geographic and climatic regions of Northern Europe. These areas represent a maritime climate (Oslo), a partly maritime-influenced and partly continental climate (Helsinki), and a continental climate (Lycksele and Gävle). One city is located in complex terrain (Oslo) and the other cities are situated in fairly flat areas (Helsinki, Lycksele and Gävle). The populations of both the metropolitan areas of Oslo and Helsinki are approaching one million, while Lycksele and Gävle are smaller conurbations with populations of 8,600 and 92,000, respectively.

The cities investigated have a number of similarities as well as differences that are important for their assessment. In regard to the sources of wood burning emissions, domestic wood burning in individual dwellings (in both ovens and fire places) is a major contributor to the total wood burning emissions in Oslo but to a lesser extent Helsinki. In the Swedish cities wood burning is often centralised at boilers and wood burning in individual dwellings is less frequent than in for instance Oslo. The meteorological conditions in these selected cities varies depending on the latitude, marine influence and terrain. However, all selected cities are subject to strong stagnant winter time conditions where low wind speeds and strong temperature inversions can lead to severe episodic pollution events.

2.1 Oslo

The city of Oslo is located at the northern end of the Oslo fjord, surrounded by a large topographical pot formation. The topographical features of the area tend to worsen the dispersion conditions, capturing pollutants emitted within the urban airshed. The most important local sources of PM in Oslo are domestic wood-burning in stoves that are used for wintertime house heating, and vehicular traffic (Laupsa and Slørdal, 2003). The influence of wood-burning PM emissions is most dominant in the densely populated central city area, where a large fraction of the flats are equipped with strongly polluting old stoves.

The winter period in Oslo from January–May 2004 and December–April 2005 is selected for this study. During this period measurements at four continuous monitoring stations (Aker Hospital, Kirkeveien, Løren and RV4) in Oslo were available, along with a regional background station (Birkenes) located approximately 300 km south-west of Oslo. At all of these sites other compounds in addition to $PM_{2.5}$, such as PM_{10} and NO_x, were also measured. At RV4 a targeted measurement campaign was undertaken during the Oslo study period where 80 12 hour filter samples were taken for analysis using a Kleinfiltergerat (KFG), 40 in each winter period. In addition to hourly measurements of air pollutants (PM_{10}, $PM_{2.5}$, NO_x, NO_2), traffic volume, speed and the composition of the vehicle fleet were measured.

The filter samples were analysed with respect to major anions and cations using ion chromatography, and 30 elements were analysed using ICP-MS (Inductively Coupled Plasma Mass Spectroscopy). Thermal Optical analysis (TOT) was used to quantify the sample content of elemental (EC) and organic carbon (OC). (Birch and Cary, 1996), whereas levoglucosan (Dye and Yttri, 2005; Yttri et al., 2005) and NCBA (N-cyclohexylbenzothiazolamin) were quantified using HPLC/HRMS (High Performance Liquid Chromatography/High-Resolution Mass Spectrometry). The primary objective of this targeted measurement campaign was to determine the contribution of non-exhaust traffic emissions but the study also provided information on the other source contributions.

Meteorological data was measured at a 25 m mast (Valle Hovin) approximately 2 km from the air quality site. A map showing the position of the stations is provided in figure 2.1. More information on the general conditions and measurement data available in Oslo are described in Larssen et al, 2007 and Laupsa et al. (2008). The receptor modelling carried out in this paper is based on the measurements from both winter periods. The dispersion and inverse modelling is only applied for the first winter period.

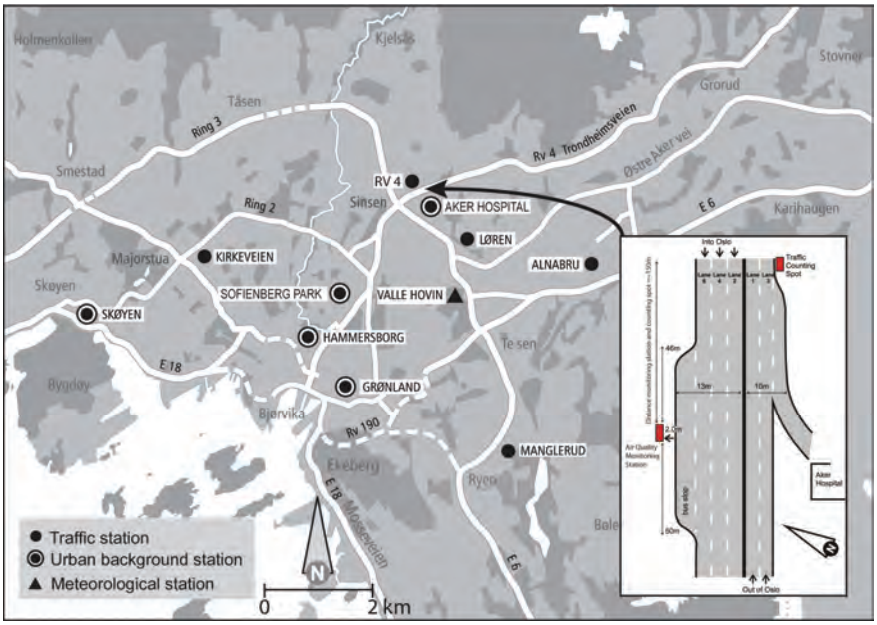


Figure 2.1. Position of the meas urement stations in Oslo. $PM_{2.5}$ data was availa ble from the stations Aker Hospital, Kirkeveien, Løren and RV4. At RV4 filter samples were taken and used for the chemical analysis and receptor modelling.

2.2 Lycksele and Gävle

Measurements were performed in Lycksele, Sweden (64.55°N, 18.72°E, 240 m a.m.s.l., population 8600) during January–March 2002 (Johansson et al., 2004). A monitoring station was placed between two family houses *situated* in a residential area (Forsdala), where residential wood burning is common. Twelve hour filter samples were collected using an automatic sampler (SAM, Hansson and Nyman, 1985). Sampling took place during 15 January–9 March 2002. A total of 103 inorganic samples were analyzed for elements using PIXE (Johansson & Campbell, 1988). In addition ionic species (IC) and levoglucosan were analysed. Total PM_{10} mass concentrations were measured at 1.9 m above ground level with a Tapered Element Oscillating Microbalance (TEOM 1400a, Rupprecht & Patashnick Inc., USA). Meteorological measurements were carried out at ~2 km from the aerosol sampling site. Wind speed WS and wind direction WD were determined by a 2-D WindSonic anemometer (Gill Instruments Inc., USA) at 13 m height. The absolute air temperature was measured at 2 m with a PRT Pt 100 probe, whereas the differential temperature between 2 and 13 m height was measured with a resistance temperature device mounted in an aspirated radiation shield (Gill Instruments Inc., USA). The measurements and other data have been described in detail by Johansson et al. (2004) and by Hedberg et al. (2006). Measurements of

$PM_{2.5}$ were also made at Norrmalm (site “N” on the map) using TEOM, though results form this site are not presented in this report.

The city of Gävle is situated along the east coast in Middle of Sweden. It has a population of 92,000 and previous studies (Lövenheim, 2006) have shown annual mean contributions from wood burning of up to 4 µg/m^3 PM_{10}. No monitoring data is available in Gävle. In this paper Gävle is used as a case study for assessing the uncertainty of the dispersion models used to calculate the contribution of wood burning emissions to $PM_{2.5}$ concentrations. This is because a very detailed emissions inventory for domestic wood burning is available in Gävle, providing a solid foundation for the sensitivity tests carried out. The main local sources of $PM_{2.5}$ in Gävle are industry, road traffic and residential wood burning. The industrial emissions are large but occur in high stacks and contribute relatively little to the air quality in central Gävle, where road traffic is the main local source.

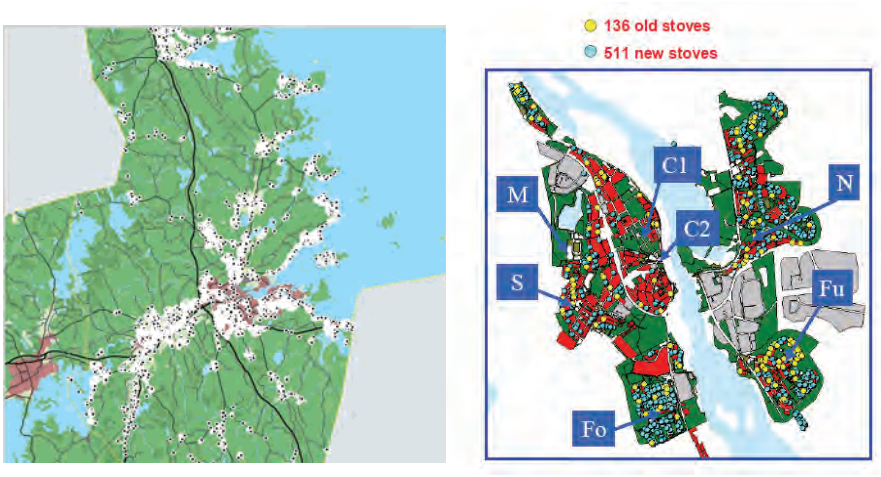


Figure 2 .2. Left the city of Gävle show ing the position of the domestic wood brunign sources. Right the town of Lycksele show ing the positions of stoves and the various measurement sitesused in the campaign. Th e site “Fo” is Forsdala where the filt er samples were taken for chemical analysis and at stations “C” (Centrla) a nd “N” (Norrmalm) PM were also measured.

2.3 Helsinki

The city of Helsinki and its surrounding region is situated in a fairly flat coastal area. The $PM_{2.5}$ concentrations at street level are dominated by long-range transport, local vehicular traffic, and to a lesser extent domestic home heating (e.g., Kauhaniemi et al., 2008).

No chemical analysis or receptor modelling data are available for this study from Helsinki. Monitoring data and dispersion calculations for the two sites of Vallila and Kallio in central Helsinki, during the 2002 winter period, are the only data used in this study (Kauhaniemi et al, 2008). These stations are operated by the Helsinki Metropolitan Area Council (YTV, 2006). The station of Vallila represents urban roadside conditions; the

station of Kallio is an urban background station. The measurement height at both stations is 4.0 m. The station of Vallila is situated in a park at a distance of 14m from the edge of the Hämeentie road. The average weekday traffic volume of Hämeentie was 13,000 vehicles/day in 2001. The heights of the buildings in the vicinity of the station, at the other side of the Hämeentie road and surrounding the park, range from 10 to 15 m. The Hämeentie road is fairly wide; there are four lanes for cars and additionally two lanes for trams. The station of Kallio is located at the edge of a sports ground. The busiest streets in the vicinity of the station are Helsinginkatu at a distance of 80m and Sturenkatu at a distance of 300 m. The average weekday traffic volume of Helsinginkatu was 7,800 vehicles/day in 2001. The station of Kallio is expected to represent the exposures that are characteristic of the centre of Helsinki. At both of these urban stations, the concentrations of $PM_{2.5}$ were measured with Eberline FH 62 I-R that is based on b-attenuation method. The flow rates of continuous instruments were calibrated twice a year with mass flow meters (Bronchorst model F-112AC-HA-00-V). The mass measurement of both Eberlines was calibrated by calibration foils. The site locations are presented in figure 2.2.

Figure 2.3. The location of the EMEP sites (Jokioinen, Ähtäri and Virolahti), urban air quality monitoring sites Kallio and Vallila, and the meteorological stations (Helsinki-Vantaa airport, Helsinki-Isosaari and Jokioinen) used in this study.

The sites and available data and applications are summarised below in table 2.1.

Table 2.1. Summary of available data and air quality situation of the study areas.

Study area	Oslo	Lycksele	Gävle	Helsinki
Monitoring sites $PM_{2.5}$	4 continuous monitoring stations	3 continuous monitoring stations	No measurement data is included here.	2 continuous monitoring sites
Chemical analysis of filter samples	80 twelve hour filter samples at traffic site (RV4)	100 twelve hour samples (day/night) in residential area (Forsdala).	None	None
Receptor modelling	PMF-2, UNMIX, PMF (ME-2), COPREM	PMF-2, COPREM	None	None
Dispersion modelling	AirQUIS modelling system	Airviro modelling system	Airviro modelling system	UDM-FMI
Estimated contribution of local domestic wood burning emissions to urban background concentrations in the winter	30–50%	40–80%	5%–30%	14%
Other major sources	Long-range transport and traffic (exhaust and non-exhaust)	Long-range transport and road traffic (exhaust and non-exhaust) (Krecl et al., 2008b)	Long-range transport road traffic (exhaust and non-exhaust)	Long-range transport and traffic (exhaust and non-exhaust)
Annual mean $PM_{2.5}$ concentrations observed	11.0–21.0 µg/m^3 (2000- 2006). 13.3 µg/m^3 during the study period (Winter 2004)	No annual means available. 6.8 µg/m^3 during the study period (winter 2002)	No measurements available. Annual mean PM_{10} modelled is 12–18 µg/m^3	8.6 µg/m^3 at the station of Kallio in 2002.

3. Methodology

As previously stated the aim of this paper is to determine if dispersion, receptor and inverse modelling techniques can be used to provide an independent estimate of domestic wood burning emissions of $PM_{2.5}$ and its contribution to urban concentrations of $PM_{2.5}$. To achieve this aim a number of models and methodologies are applied and a description of these are provided in this chapter, including methods for estimating uncertainty. These methods are applied to the case study cities, each with a different array of available measurements and modelling capabilities.

Two indirect methods are used for estimating wood burning emissions of $PM_{2.5}$. The first is based on a direct comparison between the results of dispersion models and receptor models. This will enable a direct comparison of the source contributions as calculated using the emissions inventories and dispersion modelling, compared to the chemical analysis and receptor modelling. This comparison can only be made at the receptor site, where the filter sampling is carried out. The second method applies multiple linear regression (MLR), as an inverse modelling technique, to the calculated dispersion model source contributions at a number of continuously measuring $PM_{2.5}$ sites as well as at the receptor site.

Uncertainty in the methods is determined in several ways. For the receptor modelling, uncertainties in the chemical analysis are propagated into the receptor model. For each receptor model statistical uncertainties related to the identification of source profiles can be obtained. In addition the variability between receptor models is also assessed for the cases where more than one receptor model is used. In the case of dispersion modelling the uncertainty is based on sensitivity analysis, expert knowledge and direct comparison to observations. Uncertainty in the inverse modelling is quantified by use of boot strapping techniques. This uncertainty provides an indication of the significance of the inverse modelling calculation, based on the sample size and as such must be seen as a minimum uncertainty of the inverse modelling. Though efforts have been made to quantify the uncertainty in the methods there must still remain a range of unquantifiable uncertainties. In this regard the uncertainties reported here will be indicative of the minimum uncertainty estimates.

3.1 Receptor modelling

Receptor models are mathematical or statistical procedures for identifying and quantifying the sources of air pollutants at a receptor location. Most receptor models used for source apportionment are in principle the

same model based on the assumption that the receptor site concentrations can be adequately explained by a linear combination of contributions from various relevant sources with constant source profiles. The approach to solve the mathematical problem is quite different in the two main branches of receptor models, i.e. Chemical Mass Balance (CMB) models and multivariate factor analytic models. It is the last of these two types that are applied in this paper.

Factor analysis (e.g. PCA, PMF, UNMIX) attempts to apportion the sources and determine their composition on the basis of a series of observations at the receptor site. Factor analysis is a commonly used tool, because it does not demand special user expertise or knowledge about the source profiles. The choice of the model dimension and the search for non-negative solutions by axis rotations can be based entirely on mathematical criteria. Nevertheless, it is a fundamental problem that factor analysis attempts to get more information out of the atmospheric data than is really there (Henry, 1987).

In general the PM concentrations in urban environments are the result of a large number of contributing sources. Some of these sources are not primary, but secondary particle formation in the atmosphere. The most important contributions are ammonium sulphate, ammonium bisulphate, ammonium nitrate, sea salt, reaction products of sea salt (e.g. sodium nitrate), desert or other soil dust, OC/EC from biomass combustion, OC/EC from coal and oil combustion, OC/EC from diesel exhaust, road- and tyre dust, brake dust, in some cases road salt and fugitive dust from local industry. Factor analysis cannot handle such a large number of sources without mixing everything in factors that are sometimes difficult to interpret. If the dimension of the factor analytic model is smaller than the actual number of independent sources, different sources with some degree of co-variation will be bundled in common factors (e.g. “traffic”, “secondary” or “continental air”, “oceanic air”). Often the factors are hard to interpret and may be given speculative names by the user on a completely subjective basis. In complex urban environments the solutions may not reliable and may not be suitable for source apportionment. However, some sources may be much more clearly defined than others. This will depend on the uniqueness of the chemical markers used in the analysis. A good example of this is levoglucosan, which is known to be a marker for bio-mass burning and is not found in other combustion sources.

A way out of the problems with pure CMB and pure factor analysis is the use of hybrid models that unify qualities from factor analytic models and chemical mass balance models. As an example, confirmatory factor analysis may be applied, which offers some control of the solutions by “fixing” or “freeing” specific parameters such as the factor correlation coefficients. These parameters are set according to the theoretical expectation of the researcher (Gleser, 1997; Christensen and Sain, 2002). With COPREM (Wåhlin, 2003) the bi-linear equation is solved iteratively. An

initial profile matrix is set up in which the columns have the main characteristics of known sources, and constraints are set up to maintain these characteristics, or to prevent the profiles from mixing together during the iteration. In this way any a priori knowledge about the character of the sources can be used to achieve a polarised solution with a sufficient number of sources. The Multilinear Engine (Paatero, 1999) is a programming tool that can solve multi-linear problems with the possibility of implementing many kinds of constraints using a script language.

In this paper four different receptor models have been applied. These are UNMIX, PMF (ME-2), PMF-2 and COPREM. These are listed in table 3.1.

3.1.1 Uncertainty in the source apportionment of the receptor models

In COPREM a calculation of the uncertainties (expressed as the standard deviation) of the elements in the source profiles is implemented. The calculation is based on a weighted multi-linear regression analysis. The results depend particularly on the uncertainties of the fitted data and should be considered as lower limits of the uncertainties. In principle, the upper limit of the uncertainty interval cannot be defined in a scientific way, because the result depends upon the user's subjective decisions about the number of sources and the constraints. Within the model (in which we presume that the chosen preconditions for the model are correct) a recent (beta) version of the COPREM software has a facility for the study of the rotational ambiguity of the solution. In this way a kind of uncertainty interval can be estimated. The problem defining the uncertainty of source apportionments is general for receptor modelling.

The UNMIX software version 6.0 for Windows has been released by the U.S. Environmental Protecti/products/unmix/unmix.htm). PMF Multilinear Engine (ME-2) is also available as Windows software, currently version 3.0, from the U.S. EP/pmf/pmf.htm). The variability of the source profiles obtained from UNMIX and PMF Multi-linear Engine (ME-2) are estimated using a block bootstrap technique which is implemented in both programs. The source profile is considered robust or stable, if a small change to the input data produces proportionally small change in the results. The bootstrapping technique helps to measure the variability in the source profile with respect to the variability in the input concentration data. It is important to note that variability and uncertainty are not equivalent. Uncertainty associated with a source profile can only be constructed if the underlying uncertainty distribution is known. Thus, running multiple block bootstrap runs on the same source profile is necessary to construct source profile uncertainties. Bootstrap data sets are constructed by sampling, with replacement, from the original input data set.

For UNMIX bootstrap datasets are created and run until 100 feasible solutions are obtained. They are then used to calculate the standard devia-

tion and percentile distribution of the source compositions and to generate source profile variability plots. For PMF (ME-2) the bootstrapping method is combined with a method to account for the rotational freedom in the solution. 200 bootstrap runs are carried out using a minimum correlation value of 0.6 and a random seed value of 50.

A range of receptor models have been applied to the selected cases and their application is summarised in table 3.1. All of these receptor models function on similar principles, as outlined above, however their application in different software packages and by different uses will lead to differing results. Several receptor models have been employed in order to provide an indication of the uncertainty of individual receptor modelling results and also to try to improve the final results of the modelling by combining and assessing different techniques. In table 3.1 the different users are also indicated as this can play a role in the resulting calculation.

Table 3.1. Receptor models applied in the study

Receptor model	Short description	Uncertainty assessment	Application city	Reference	Applied by *
UNMIX	Self-modelling curve resolution	Boot strapping	Oslo	Henry (2003)	User 1
PMF (ME-2)	Weighted least squares using the Multi-linear Engine	Boot strapping	Oslo	Paatero (1999)	User 1
PMF-2	Weighted least squares using non-negativity constraint	Based on the statistical uncertainty obtained from the model	Oslo, Lycksele	Paatero (1993), Paatero and Tapper (1993; 1994), Polissar et al. (1998)	User 2 User 4
COPREM	Confirmatory factor analysis. The user can pre-define part , or all, of the source chemical profiles. Solved iteratively	Statistical assessment of multiple linear regression of the resulting source profiles	Oslo, Lycksele	Wåhlin (2003)	User 1 User 3 User 4

*User 1: NILU (MK); User 2.: NILU (JS); User 3: NERI (PW); User 4: ITM (CJ)

3.2 Dispersion modelling

Three different dispersion models are applied for the four different cities. The fundamental differences between the models lie in the use of either Gaussian or Eulerian dispersion model types. In addition to this difference meteorological data may also be applied in different ways and, in the case of wood burning emissions, different methods are used to describe the vertical distribution of emissions. Table 3.2 provides an overview of the different models used in this paper.

3.2.1 AirQUIS

For Oslo the model suite available in AirQUIS (AirQUIS, 2008; Slørdal et al., 2003; Slørdal et al., 2008) are applied. Wood burning emissions are dispersed using a Eulerian model with a horizontal resolution of 1 x 1 km and a vertical resolution of 10 layers, with the lowest layer at 14 m. Wood burning emissions are introduced directly into the lowest 3 grid layers (up to 72 m). Sensitivity tests carried out later in this paper address the sensitivity of the model to both the vertical and horizontal distribution of the wood burning emissions. Emissions from roads are dispersed using the line source model HIWAY-2 (Petersen, 1980), which is a Gaussian line source model. Long-range transport is taken from observations at Birkenes, a rural background station approximately 300 km south-west of Oslo.

3.2.2 UDM-FMI

For Helsinki the transport and dispersion of wood burning emission is calculated with the Gaussian dispersion model UDM-FMI (Karppinen et al., 2000b) with 1 x 1 km emission source resolution. The emission inventories include wood combustion emissions from the whole of Finland within a 20 km radius and traffic emissions from the Helsinki Metropolitan Area. The wood burning source height is set at 7.5 m (containing emission height plus initial plume rise). The wood burning source was assumed to be in the centre of a square calculation grid of size 40 x 40 km^2 for domestic wood combustion emissions, the interval of grid points being 1 km, and road traffic emissions were calculated from the network of roads and streets. The Helsinki traffic emission inventory includes exhaust emissions, cold starts and driving, and non-exhaust vehicular emissions, for the network of roads and streets within Helsinki Metropolitan Area. The emissions were modelled dependent on vehicle travel velocity based on the EMME/2 transportation planning system (INRO, 1994) and nationally conducted vehicle emission measurements (Laurikko, 1998). Included in the traffic emissions is a correction for resuspended road dust and salt, used to scale the exhaust emissions by a factor of 2.6. The contribution of long-range transport to the concentrations is evaluated using the Ion Sum method (Karppinen et al., 2004). Model concentrations were calculated at the height of 2.0 m.

3.2.3 Airviro

For both Lycksele and Gävle a Lagrangian Gaussian and a Eulerian grid model, which are part of the Airviro-system are used (http://www.airviro.smhi.se). The Airviro software is a GIS-based Air Quality Management system. It includes modules for dispersion calculations, emission data administration and measurement data storing and presentation. Air

quality modelling studies using the Airviro system have been published in scientific journals (Johansson et al., 2007; Murkherjee et al., 2000; Murkherjee and Viswanathan, 2001; Namdeo et al., 2002; Nyberg et al., 2000; Bellander et al., 2001; Rosenlund et al., 2006). The Gaussian model has been used in a number of projects in the Stockholm region, mainly with the objective of describing the exposure of the population (Johansson et al., 1999; Nyberg et al., 2000; Bellander et al., 2001; Rosenlund et al., 2006; Eneroth and Johansson, 2006; Johansson and Eneroth, 2007; Lövenheim et al., 2007; Johansson et al., 2008). Using the Lagrangian Gaussian model the simulation area should be relatively flat and not too large since stationary conditions are assumed. The Eulerian grid model allows dispersion in complex topography and over large areas. The Lagrangian Gaussian model is the model chiefly applied in this study, with the exception of a number of sensitivity tests.

Chemical and physical transformation processes of particles as well as dry and wet deposition are neglected in the current model calculations. In both models, the buildings in the city are parameterised as surface roughness, individual buildings are thus not resolved. Brief descriptions of the models are given below.

In the Airviro Gaussian model the advection of the polluted air follows trajectories in the wind field. For each emission point source, such a trajectory will constitute the plume centre line, i.e. a transformation to a Lagrangian coordinate system is made. Depending on the emission characteristics, an initial distribution of the pollutant is defined by the initial (at X = 0) dispersion length scales of σ_y and σ_z. For point sources, introduced either in the Lagrangian or the Eulerian grid model, σ_y and σ_z at X = 0 are set equal to external stack radius. For area sources introduced individually the initial horizontal dispersion, σ_y and σ_z are directly related to the grid size.

The Airviro Eulerian model is based on the three-dimensional advection-diffusion equation. For details on numerical solutions, parameterisation of the turbulent vertical exchange coefficient and the terrain-following co-ordinate system see the specifications in the documentation at http://www.airviro.smhi.se. In the grid model the plume is initially treated as a Gaussian puff following the trajectory of the wind field. When the plume extends over a magnitude comparable with the horizontal grid size it is released into the grid cells.

3.2.4 Uncertainty in the dispersion modelling

There is a wide range of uncertainty sources in dispersion models. These include inherent errors in the dispersion parameterisations used, the limited numerical resolution of the models and missing or poor process descriptions. There are also uncertainties in input data, such a meteorological fields, as well as the emissions. Generally direct comparisons of modelled and observed concentrations are made to assess the total model

uncertainty (Borrego et al., 2007; Chang and Hanna, 2004). However, such assessments also include uncertainties in the emissions and it is difficult, if not impossible, to separate the model uncertainties from the emission uncertainties. Only under controlled and usually simplified conditions is it possible to separate the individual sources of uncertainty. Such conditions or experiments are not available for urban areas.

An indirect method for ascertaining the uncertainty of a model, with respect to the inaccuracies of input data or the selection of various submodels and parameterisations, is through sensitivity studies. By varying parameters or process descriptions that are considered to be within the range of likely values then an estimate of their influence on the final modelled concentrations can be made. This provides some information concerning the range of model outcomes and can be implemented independent of emissions.

In this study the uncertainty of the dispersion models is assessed in three ways:

- Through direct comparison with measurements to provide a statistical indication of the uncertainty in the total model calculations.
- Through sensitivity analysis of model calculations to a selected set of parameters. In this case we concentrate on the emission height and vertical profile from wood burning sources.
- Through inter-comparison of models or model types. In this case through a comparison of Gaussian and Eulerian models.

The results of uncertainty analysis will be presented as part of the dispersion modelling results.

Table 3.2. Overview of the dispersion models used in this paper

Study area	Oslo	Lycksele	Gavle	Helsinki
Model name	AirQUIS-EPISODE (Slørdal, 2003)	Airviro	Airviro	UDM-FMI
Model type	Eulerian model for area emissions with an embedded Gaussian line source model for traffic emissions (Petersen, 1980)	Gaussian line, area and point source model (www.airviro.smhi.se)	Gaussian line, area and point source model and Eulerian model (www.airviro.smhi.se)	Gaussian model for stationary point, area and volume sources + line sources
Meteorological model	Diagnostic wind field model (Foster et al., 1995)	Diagnostic wind field model (Danard et al. 1977)	Diagnostic wind field model (Danard et al. 1977)	MPP-FMI (Karppinen et al, 2000a)
Vertical emission profile for wood burning	Wood burning emissions evenly distributed in the lowest three model layers (< 72m)	Every single family house is described as a point source with chimney height, exhaust gas speed and temperature.	See text for the different tests carried out.	Emission height plus initial plume rise 7.5m
Model resolution	1 x 1 km horizontal resolution, 10 vertical layers starting at 14 m	50 meter horizontal resolution.	100, 250, 500 and 1000 meter horizontal resolution used in different scenarios.	1 x 1 km horizontal, concentrations calculated for height of 2 m
Uncertainty assessment	- Comparison to observations - Sensitivity to vertical emission profiles - Sensitivity to horizontal resolution	Comparison to observations	- Sensitivity to initial plume height - Sensitivity to model type	Comparison to observations

3.3 Wood burning emission inventories

Wood burning emission inventories are collected and applied in the dispersion models in different ways, dependent on the type of source and the city. A brief description is provided here and summarised inn table 3.3.

3.3.1 Oslo

In Oslo the spatial and temporal emission inventory for wood burning is based on a survey of home heating habits, carried out in the autumn of 2002 (Finstad et al., 2004) and the total emissions are based on consumption data related to the sale of wood in the Oslo region. Maximum wood burning emissions occur between 17:00 and 22:00, with higher emissions on weekend days, and the weekly time variation reaches a maximum in the last week of January, since wood burning emissions are distributed according to the climatological temperature variation. In the dispersion

model calculations presented here for Oslo, no daily dependence on actual temperature is used in the emission calculations.

3.3.2 Lycksele

The emission inventory for Lycksele is described by Johansson et al. (2004). The sampling site was located in a residential area with 391 local conventional-type wood stoves and 13 older, non-environmentally approved high emitting old wood boilers without heat storage tanks. Even though local conventional-type wood stoves are the most frequent, they are not expected to be used as the main heating source but more for pleasure heating. Other heating systems in the area include wood boilers connected to heat storage tanks, multi-fuel boilers and electrical radiators. Old multi-fuel boilers without heat storage tanks are estimated to dominate the emissions of PM (Johansson et al., 2004). Local traffic within the area is limited. The closest major road is located 200 m from the site, ~3000 vehicles/day.

3.3.3 Gävle

A detailed emission inventory for domestic wood burning in Gävle has been performed by the local environmental administration of the municipality (Ekman, 2007). The emissions due to domestic wood burning in Gävle are based on information on the household's appliances. The emission depends on the type of appliance and the required energy consumption. In the database there are 21,909 appliances. The appliances range from small pleasure heating and cooking stoves and fire places to large wood or oil boilers. In a boiler, fuel is burnt to heat water. The heated water is either used for direct heating of a house through a piping system, or to heat water stored in a heat storage tank, which in turn is circulated through the piping system to heat a house.

3.3.4 Helsinki

For Helsinki, wood burning emission estimates were based on source-receptor matrices for inert particles, obtained from the Finnish Regional Emission Scenario (FRES) model (Karvosenoja, 2008; Karvosenoja et al., 2008). The size of each source is 1 x 1 km^2, and the spatial allocation of emissions in the FRES model is determined according to weighting factors from the building and dwelling register and municipality level data. The diurnal and seasonal variations in wood burning (from questionnaires) were taken into account in generating the hourly time series for the emissions. For domestic wood combustion, the emission height was assumed to be 7.5 m including both source height and initial plume rise.

3.3.5 "Review, improvement and harmonisation of the Nordic PM Air Emission Inventories"

Parallel to this project the Nordic Council of Ministers (NMR) funded also a project on "Review, improvement and harmonisation of the Nordic PM Air Emission Inventories". Since the topics are related a close co-operation between the two NMR projects was started. Several times persons participated in project meetings of the other project, giving short reports on progress in each others projects, exchanging information, references and emission data.

The fraction of emission from combustion in residential plants (small scale combustion) on total PM emissions reported in 2005 in the Nordic countries is in average 39% for PM_{10} and 50% for $PM_{2.5}$ (Illerup et al. 2009). For Norway up to 80% of the PM emissions are reported from residential plants.

For the emission inventories so far only the total mass (total TSP, $PM_{2.5}$ or PM_{10}) is estimated based on emission factors and fuel consumption. The chemical specification, source profiles for other trace elements as estimated in receptor analysis for wood emission will be useful to derive emissions for other compounds in future emission inventories.

The emission factors used in the inventories are based on fuel or energy consumption (g per GJ) and depend very much on the type of fuel and age of stove used. Modern stoves using pellets have emissions as low as 20 g/GJ while conventional fire places using wood logs emit about 2,000 g/GJ (Illerup et al. 2009). This information about emission factors and their variability and uncertainty is very useful for dispersion modelling studies as reported in this study (see section 5).

Table 3.3. Summary information concerning the domestic wood burning emission inventories

Study area	Oslo	Lycksele	Gävle	Helsinki
Short description	Based on surveys of wood burning habits and indicative sales of wood	Based on detailed inventories of individual appliances	Based on detailed inventories of individual appliances	Data on locations as a combination from building & dwelling register, and municipality level data, temporally according to month of year and time of day
Temperature dependence	Only climatological, no daily dependence	Daily	Daily	Climatological
Reference	Finstad et al. (2004)	Johansson et al. (2004)	Ekman (2007)	Karvosenoja et al. (2008)
Estimated uncertainty of daily emissions	Unknown	Unknown	Unknown	Unknown
Estimated uncertainty of annual emissions	50 %	50%	50%	-37% to + 50%

3.4 Inverse modelling using multiple linear regression

The aim of the inverse modelling is to provide an indication of the average contribution of particular source sectors to the total observed $PM_{2.5}$ concentration. We consider that there is no chemical or physical transformation and these sources are additive in the following manner:

$$C(x,y,t) = \sum_{i=1}^{n} c_i(x,y,t) \qquad (1)$$

where C is the total concentration and c_i indicates the contributions from the n source sectors contributing to the total $PM_{2.5}$ concentration. The true source contributions at any particular site are unknown from the total $PM_{2.5}$ mass concentrations and so we use a simple linear model to describe these given by

$$C_{obs}(x,y,t) = \sum_{i=1}^{n} a_i c_{\mathrm{mod}\,i}(x,y,t) + \varepsilon_i(x,y,t) \qquad (2)$$

where ε is the error and the coefficients a are time independent. Writing it in this way infers that each individual source contribution can be scaled by the factor a_i to minimise the error ε. The factors a_i can be determined by minimising the mean square error. This is equivalent to MLR without a bias offset, i.e. the intercept is forced to pass through 0. Given a number of observations in time and space, the factors a_i can be determined and can be interpreted to be emission scaling factors, though they may also be the result of model error and bias.

The methodology is generally applicable to any set of sources but the following conditions will apply for its application:

- The contribution of the different source sectors should not be highly correlated. MLR will not be able to distinguish between the sources in such a case.
- There is an assumption of linearity in regard to the modelled and real source contributions when using MLR. If this is not the case then MLR will not work optimally.
- The methodology is best applied when the source contributions are of the same order of magnitude. The methodology will not provide useful results for a particular source when it is significantly smaller than the others.

Uncertainty in the MLR analysis: To assess the uncertainty in the MLR method, bootstrapping methods are used to provide standard deviations of the regression slope parameters. 10,000 realisations are used to determine the standard deviations of the scaling factors a_i. This type of uncertainty analysis provides an indication of the uncertainty due to the limited samples available for the regression. This must be considered to be the minimum uncertainty in the determination of the linear regression coefficients a_i.

4. Results of the receptor modelling

The results of the receptor modelling studies are presented here for two case study cities (Oslo and Lyksele). The focus is on the results of the wood burning contribution but other contributions will also be discussed. The main emphasis here is on the chemical profiles and total source contributions determined by the various methods. A number of results are reported based on different receptor models and on different assumptions (users) when applying the models. This “ensemble” of models and users is used, along with uncertainty estimates from the individual receptor models, to indicate the uncertainty of the receptor modelling results.

4.1 Oslo

For the Oslo case five different receptor results are analysed, using various numbers of sources and different software packages. Table 4.1 provides an overview of these model applications.

Table 4.1. List of receptor models and number of sources applied for the Oslo case.

Receptor model	User	Number of sources
COPREM	User 1: NILU(MK)	13
COPREM	User 3: NERI(PW)	14
UNMIX	User 1: NILU(MK)	4
PMF2	User 2: NILU(JS)	6
PMF (ME-2)	User 1: NILU(MK)	6

The 5 main sources that are included in most of the model applications are:

- Biomass/Wood burning
- Regional background/long range transport
- Vehicle exhaust
- Road and tyre wear/resuspended road dust
- Road salt

Four source profiles were identified in the UNMIX analysis (Regional background, Road salt, Wood burning, Vehicle exhaust). Two parameters determine the reliability of a solution found by UNMIX: the minimum $R^2$and the signal-to-noise ratio. The R^2 value is related to the proportion of variance of each species explained by the source factors. In general, it is recommended that the minimum R^2 is greater than 0.8 and the mini-

mum signal-to-noise ratio is greater than 2. For the Oslo $PM_{2.5}$ dataset, a solution with a minimum R^2 of 0.84 and a minimum signal-to-noise ratio of 3.88 was found. UNMIX does not use uncertainty estimates for the input data.

PMF(ME-2) was first run with for 4 sources and similar source profiles as with UNMIX were obtained. Increasing the number of sources to 5 could reduce the convergence criterion Q to about 2,300–2,500. With 6 sources, an additional source that contributed less than 3% to total $PM_{2.5}$ was found and Q was about 1,900, very close to the statistically expected value of Q (= number of samples × number of species).

Measurement uncertainties used in the COPREM analysis from NILU were the same as in the PMF (ME-2) analysis. The initial source profiles for Vehicle exhaust and Wood burning were constrained by the corresponding source profiles from the PMF (ME-2) solution. The COPREM model is able, due to the possibility of specifying detailed constrains, to separate a number of correlated sources such as brakes and smaller sub-fractions of the regional background contribution.

The obtained source contributions from the range of receptor modelling results are shown in figure 4.1. All models are consistent in giving wood burning the highest contribution followed by “Regional background” and “Road Exhaust”. Largest deviations are observed for the “salt” source, while the other sources agree reasonably well. Note that the 9 sources from “$(NH_4)HSO_4$” to “Oil” should be compared as the sum to the “Regional background”. As well “Exhaust” and “Brakes” should be added before comparing between the models.

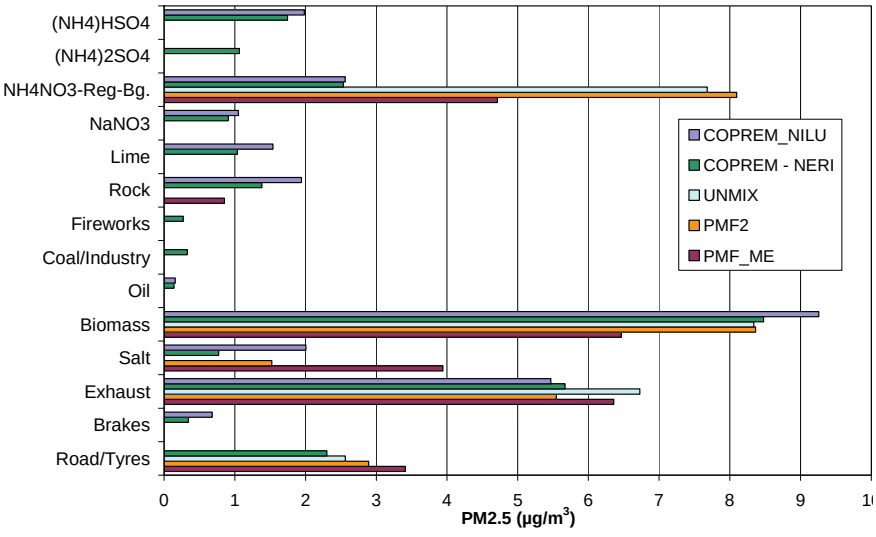


Figure 4.1. Source contributions for the 5 receptor model applications in the Oslo case (table 4.1). Note the different number of sources in the models, and therefore the subdivision of the contributions from “Regional background” and “Exhaust” in the COPREM results.

Source profiles for wood burning are compared in figure 4.2. The models agree relatively well (within a factor of 2) in the source contributions to OC, EC, LG, K and Zn, except from COPREM (NERI) which is missing the Elemental Carbon (EC). For other components much larger deviations are found. Some of the minor contributions might be considered as "noise" since only COPREM is able to force some "non-physical compounds" to exactly zero while the other models will often end up with some minor contributions in secondary elements.

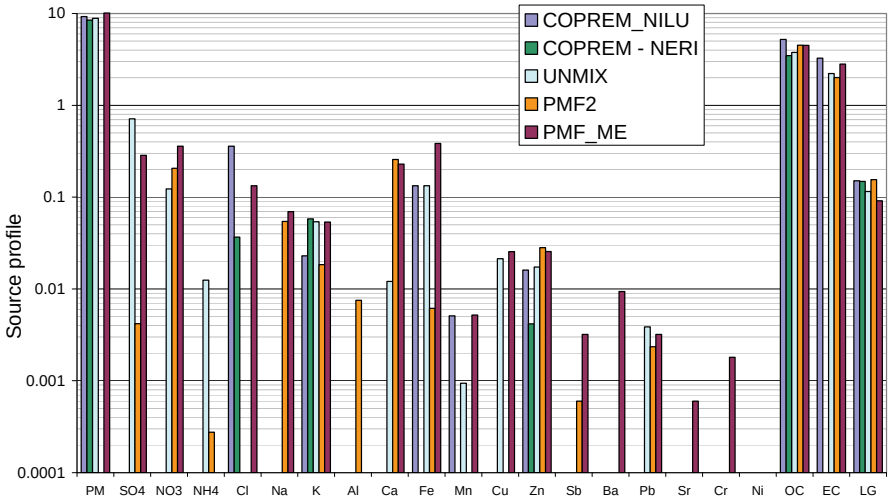


Figure 4.2. Chemical profiles of the source classified as wood burning using the 5 receptor model applications applied to the Oslo case (Table 4.1).

The receptor model provides source contributions for each sampling period. An example for the COPREM (User 1) result is shown in figure 4.3 below.

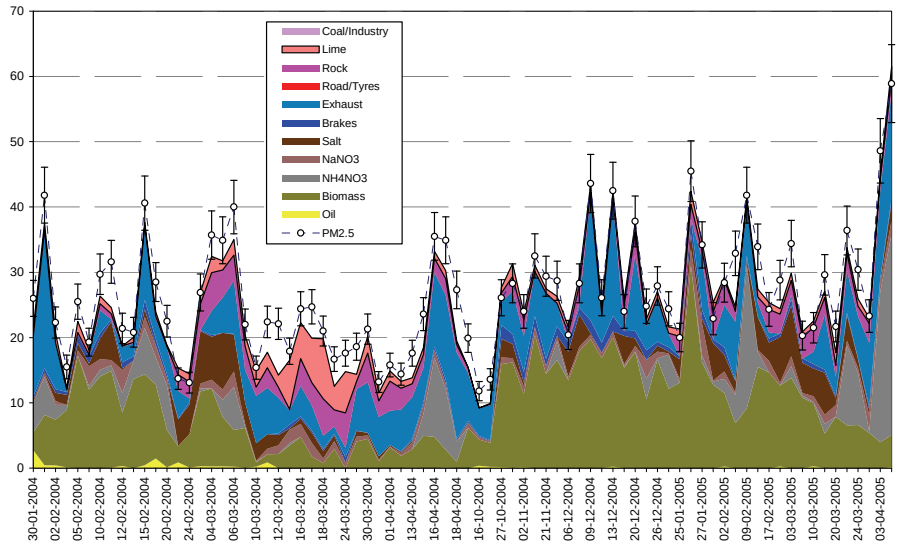


Figure 4.3. Showing the time series dependen ce of the source contributions for Oslo for both the winter of 2004 and 2005.

4.1.1 Uncertainties

As described in section 3.1 the methods for determining uncertainty in the receptor model results vary from model to model and package to package. For UNMIX and PMF (ME-2) bootstrapping methods are applied, for COPREM the standard error is used of the source distributions calculated from a weighted multi-linear regression analysis. In PMF-2 uncertainty from the chemical specification is propagated through the model to give uncertainties for individual elements in the various sources. It is thus difficult to directly compare the uncertainty estimates from different models.

In figure 4.4 output plots from UNMIX and PMF (ME-2) are shown to indicate how the uncertainty assessment is visualised in the software packages used. In this case the chemical profile for wood burning is shown.

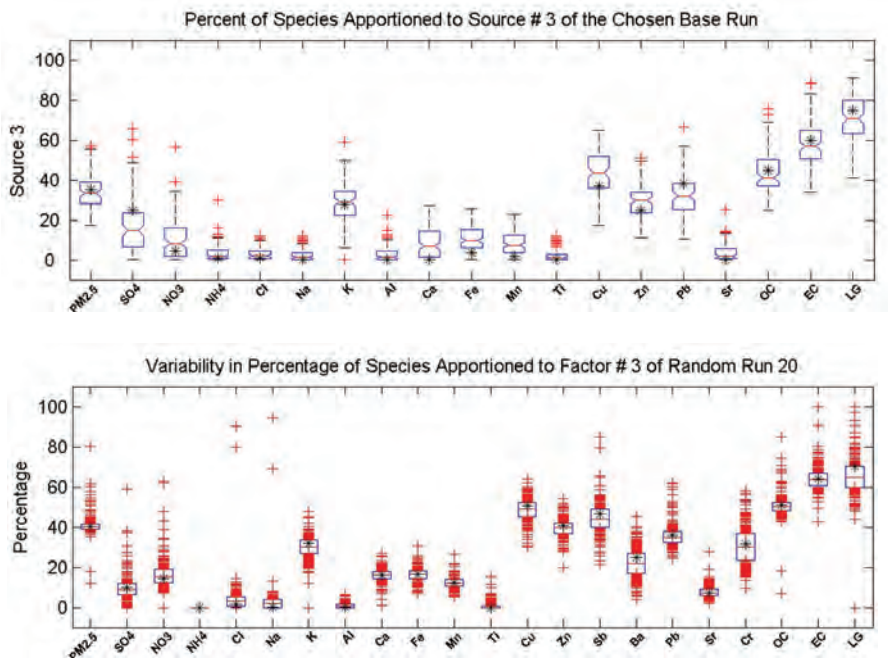


Figure 4.4. Top: Variability of the wood burning source profile in the UNMIX solution showing the percentage of species apportioned to the wood burning factor. Blue boxes denote the range where 50% of the bootstrap values lie. Black asterisks denote values from the base model run (normalized by the bootstraps row sum). Red pluses represent the outliers of the bootstraps analysis. Bottom: Variability of the wood burning source profile in the PMF (ME-2) solution obtained from 200 bootstraps runs. Percentage of species apportioned to the wood burning factor. Blue boxes denote the range where 50% of the bootstrap values lie. Clustered red pluses that lie close to the box indicate good reproducibility.

Uncertainties obtained from each of the receptor models represent standard deviations of the receptor model fit to the observed data and should be considered as lower limits of the real uncertainties associated with the receptor model solution. In principle, the upper limit of the uncertainty

interval cannot be defined in a scientific way, because the result depends upon the user's subjective decisions about the number of sources and the constraints (a priori information on sources, non-negativity, etc.).

4.2 Lycksele

For the Lycksele case only 3 receptor model applications are available: two using COPREM and one using PMF-2. 5 sources were resolved by these models as shown in figure 4.5. All models show good agreement in attributing the highest source contribution to the wood burning source.

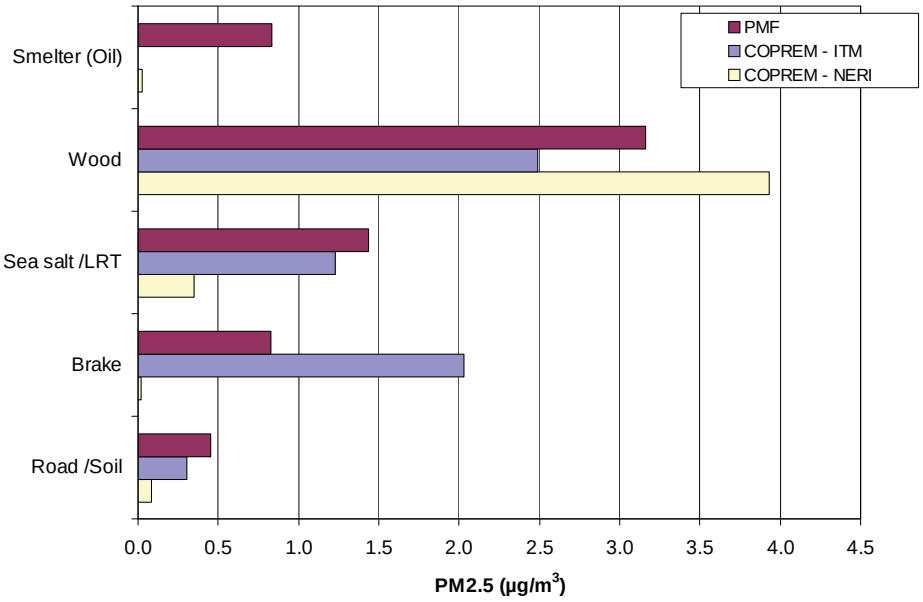


Figure 4.5. Comparing the source con tribution to $PM_{2.5}$ for PMF-2 and the two different COPREM runs.

The plots in figure 4.6 show the source contributions on a daily basis for the three model results. These of course reflect the differences in the total source contributions, as shown in figure 4.5.

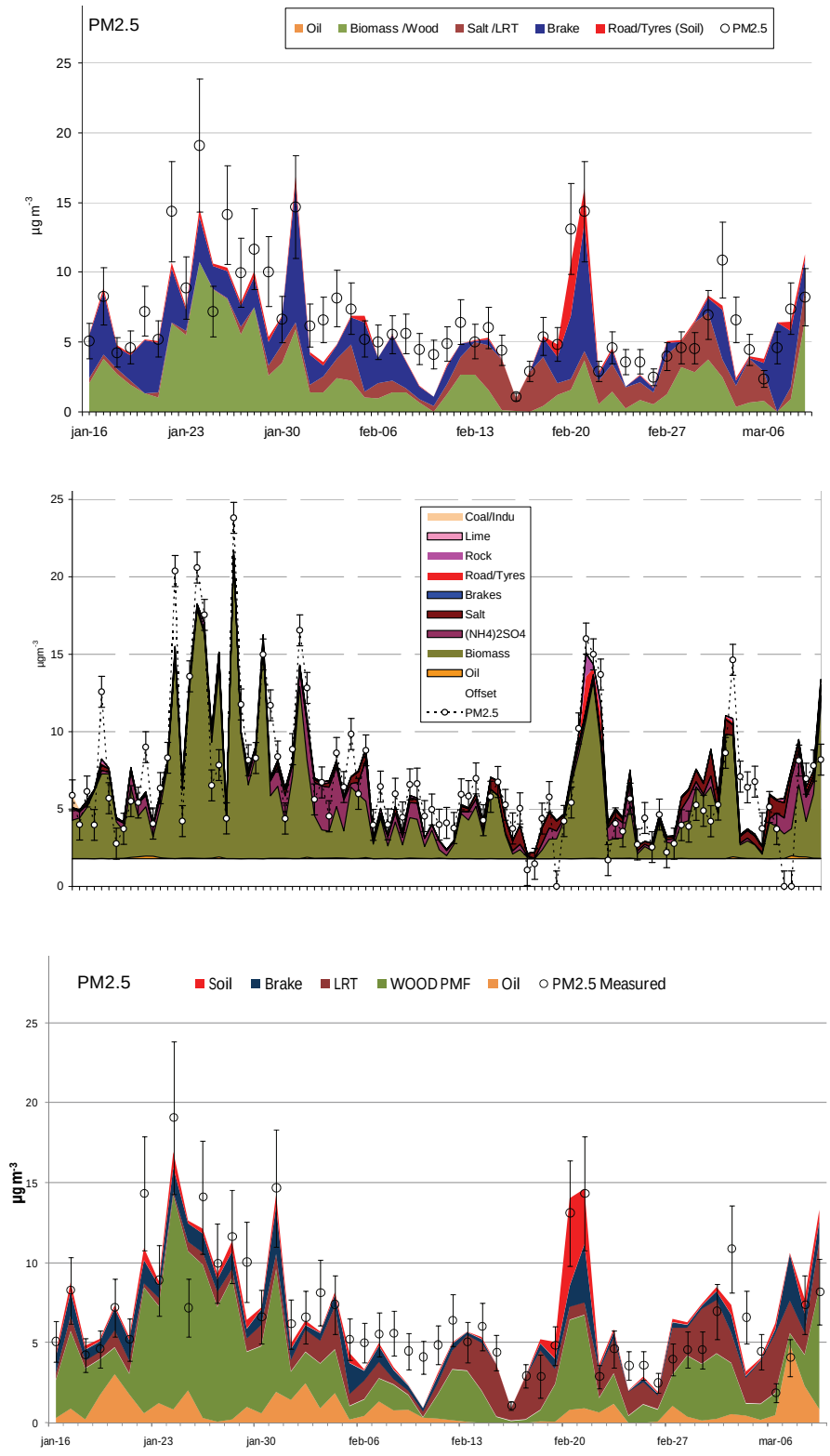


Figure 4.6. Source apportionment of PM $_{2.5}$ according to COPREM (User 4) (top), COPREM (User 3,(Middle) and PMF-2 (User 4) for Lycksele. Note the higher time resolution (12-hour) for the COPREM (User 3), others show daily means.

4.2.1 Source profiles

The input source profiles for COPREM are shown in table 4.2. Source profiles for brake wear were taken from measurements by Hjortenkrans et al. (2007; 2006). The main marker used is Cu. To achieve a separation from road wear, Ti was set to zero in the brake wear source profile. For residential wood burning (RWC), the wood stove emission measurements of Hedberg et al. (2002) were applied with K, Fe, Cu and Rb fixed. For oil Ni was used as indicator and Cu was fixed to zero. For sea salt Br was used as an indicator and Cu and Ti were set to zero. PMF-2 was run as described by Hedberg et al. (2006), with the difference that $PM_{2.5}$ was included in the PMF-2 calculation together with the elements, not via regression afterwards.

Table 4.2. Initial source profiles used for COPREM (user 4).

	Road wear	Brake wear	Sea salt	RWC	Oil
$PM_{2.5}$	0	0	0	1300	0
As	0	0	0	0	0
Br	0.0000395	0	**1**	0.19	0
Ca	0.0978	0	0	0.43	0
Cu	0	**0.687**	**0**	**0.07**	**0**
Fe	0.0431	0	0	**0.21**	0
K	0.0135	0	0	**27.4**	0
Mn	0.00197	0	0	0.14	0
Ni	0.0000903	0	0	0.06	**1**
Pb	0.000165	0.00666	0	0.34	0
Rb	0	0	0	**0.15**	0
S	0	**0**	0	6.55	0
Ti	**0.00405**	**0**	**0**	0	0
Zn	0.00269	0.198	0	7.5	0

Values in bold are fixed using the form-matrix as described by Wåhlin (2003). Other values (non bold) are free to change during the COPREM run, see table 4.3.

Calculated source contributions for $PM_{2.5}$ and the related elements are given in table 4.3 and table 4.4 for COPREM and PMF, respectively. A comparison of the source contributions based on COPREM and PMF is shown in figure 4.7. For the wood burning the agreement is excellent, possibly due to the fact that this source dominates the total mass contribution.

Table 4.3. Calculated source contributions using COPREM (User 4).

	Road Wear	Brake wear	Sea salt	RWC	Oil	Total con-centration predicted	Residue
$PM_{2.5}$	0.31	2.03	1.23	2.49	0.00	6.05	0.72
As	0.16	0.0075	0.00	0.14	0.056	0.36	0.16
Br	0.00	0.00	0.63	0.023	0.069	0.72	0.092
Ca	12.1	0.00	4.5	0	1.3	18.0	1.9
Cu	0.0050	0.79	0.00	0.22	0.00	1.0	0.041
Fe	15.4	5.97	0.031	0.65	0.00	22.0	2.12
K	16.5	0.00	7.8	85.1	8.4	117.8	1.6
Mn	1.14	0.27	0.00	0.11	0.00	1.51	0.21
Ni	0.0042	0.00	0.00	0.0076	0.40	0.41	0.014
Pb	0	0.31	0.13	0.025	0.17	0.64	0.60
Rb	0.10	0.00	0.027	0.47	0.040	0.63	0.023
S	8.13	0.00	39.4	51.4	60.7	159	51.9
Ti	0.64	0.00	0.00	0.16	0.094	0.90	0.23
Zn	1.46	0.64	0.28	13.8	0.80	17.0	0.36

Residue is calculated as the difference between measured and calculated. Unit: All elements are expressed in ng m^{-3}; $PM_{2.5}$ is given in µg m^{-3}.

Table 4.4. Calculated source contributions using PMF-2 (User 4).

	Road Wear	Brake wear	Sea salt	RWC	Oil	Pred PMF	Residue
$PM_{2.5}$	0.45	0.83	1.44	3.16	0.83	6.71	0.09
As	0.08	0.05	0.05	0.17	0.07	0.42	0.10
Br	0.00	0.00	0.73	0.00	0.00	0.73	0.06
Ca	8.71	0.09	6.29	0.79	0.45	16.33	3.77
Cu	0.02	1.05	0.00	0.01	0.01	1.09	0.01
Fe	18.58	1.31	0.29	0.71	1.69	22.59	1.87
K	5.12	9.09	7.69	91.53	5.20	118.63	0.90
Mn	1.22	0.11	0.01	0.15	0.03	1.52	0.25
Ni	0.00	0.00	0.00	0.00	0.30	0.30	0.13
Pb	0.01	0.17	0.25	0.47	0.35	1.25	0.29
Rb	0.03	0.09	0.08	0.50	0.01	0.70	-0.04
S	0.06	1.16	47.39	36.53	98.27	183.41	29.84
Ti	0.62	0.13	0.23	0.10	0.04	1.12	0.01
Zn	0.11	2.10	0.00	14.23	0.22	16.67	0.82

Residue is calculated as the difference between measured and calculated. Unit: All elements are expressed in ng m^{-3}; $PM_{2.5}$ is given in µg m^{-3}.

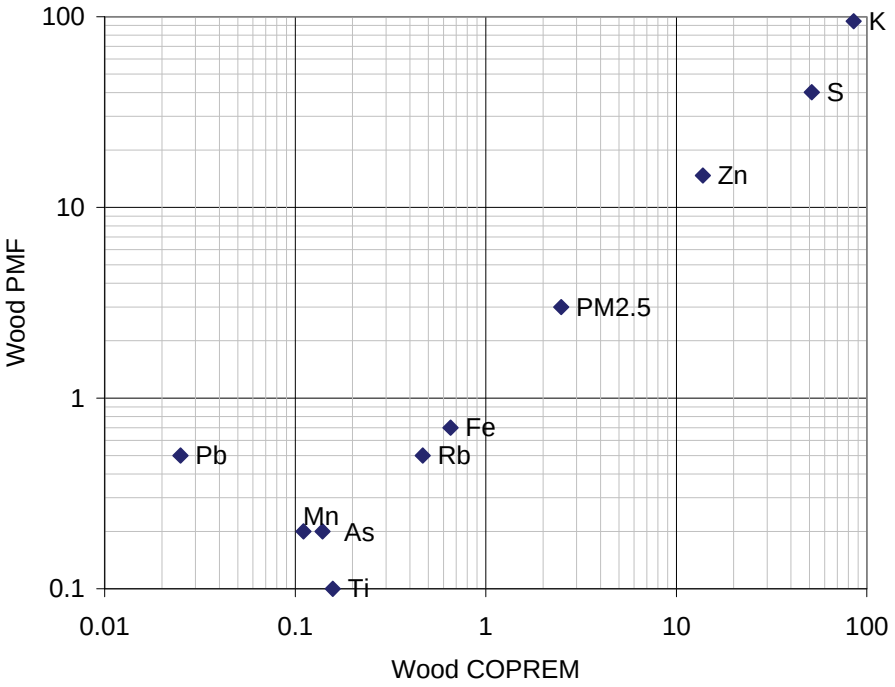


Figure 4.7. Co mparison of source apportionmen t of the different elements using CO-PREM (User 4) and PMF-2 (User 4) for the wood burning source.

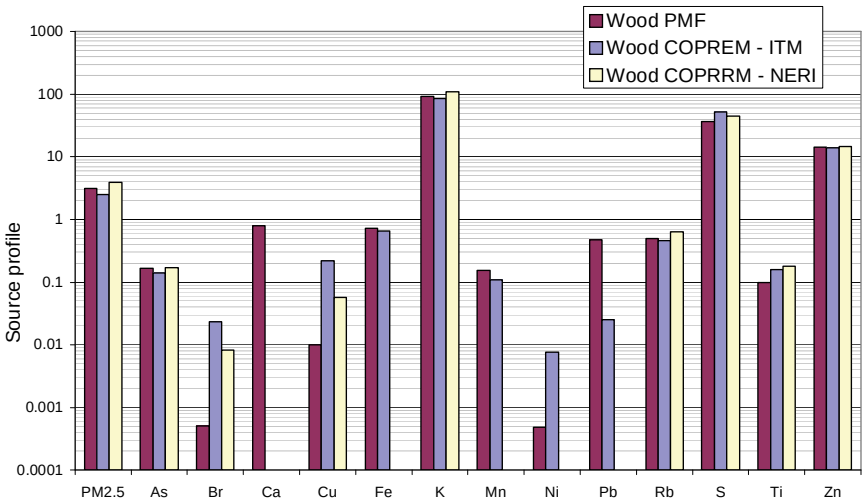
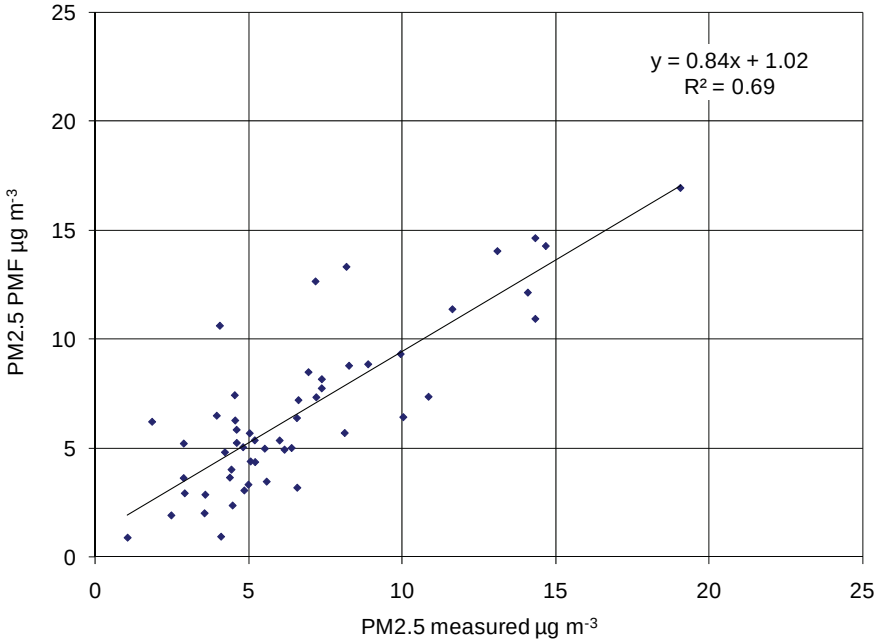
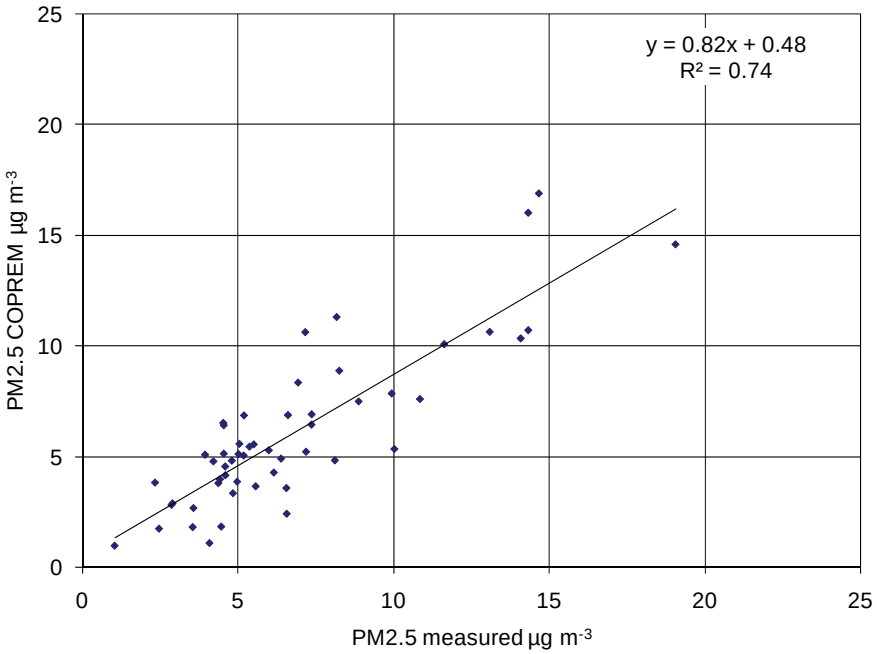


Figure 4.8. Comparing the wood source profile for PMF and two different COPREM runs

4.2 Scatter plots of total $PM_{2.5}$

The R^2 values for calculated versus measured $PM_{2.5}$ concentrations are 0.69, 0.74 and 0.75 for PMF2, COPREM (User 4 and 3), respectively (see figure 4.9). This shows the very similar performance of the models for this case.





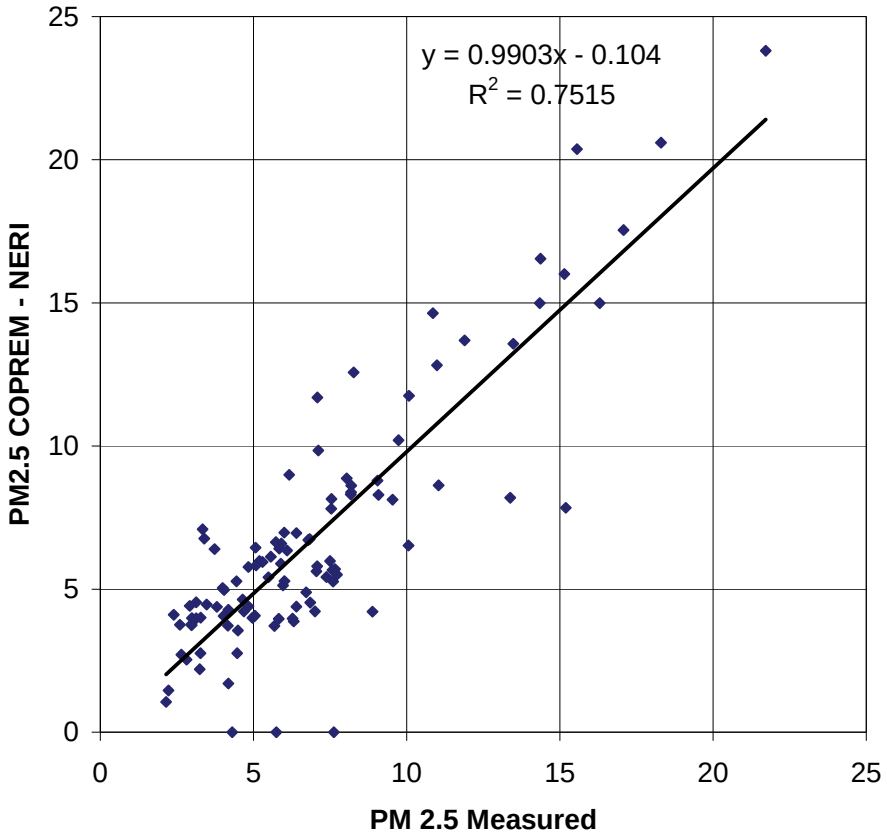


Figure 4.9 Scatter plots of measured $PM_{2.5}$ versus calculated using PMF2 (upper left) and COPREM (User 4 – upper right) and COPREM (User 3 – lower right).

4.3 Temporal variation and temperature dependence

Wood burning concentrations are expected to be dependent on not only wind speed and stability but also the temperature which directly effects the emissions (Hedberg et al., 2006). Though temperature may be correlated with emissions it is also related to stability and so the two effects can amplify the contribution of wood burning. To investigate this a comparison of the calculated time series of total $PM_{2.5}$ concentration due to emissions from wood burning using COPREM, for both Oslo and Lycksele, is shown in figure 4.10. There is a clear correspondence in Lycksele between ambient air temperature and wood burning contributions. The correspondence is less well defined, but still present, for Oslo. The highest contributions occur during the cold periods reflecting the increased emissions as well as more stable atmospheric conditions

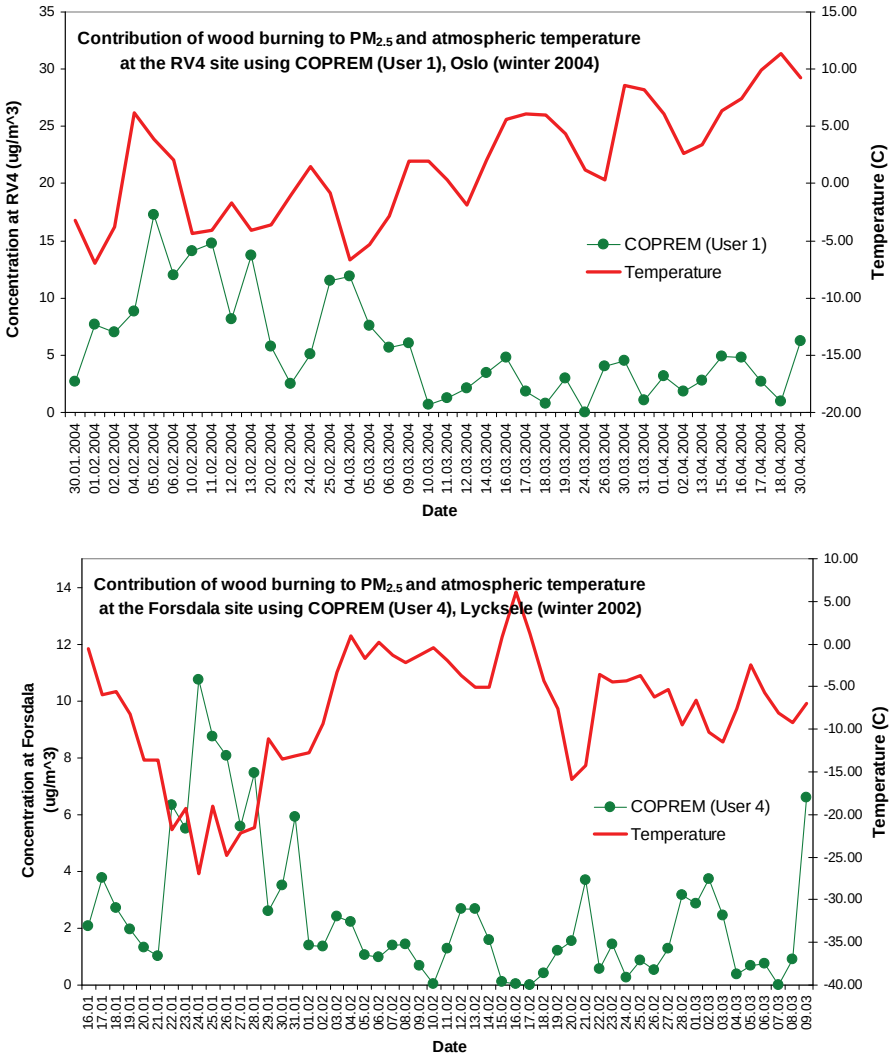


Figure 4.10. Time series of ambient temperature and the contribution to $PM_{2.5}$ from biomass burning according to COPREM for the receptor point at RV4, Oslo in the winter period 2004 (Top) and Forsdala, Lycksele winter 2002 (Bottom).

5. Results of the dispersion modelling

In this section the results of the dispersion modelling, as well as sensitivity analysis of the models, are reported. Dispersion models have been applied in Oslo, Lycksele, Gävle and Helsinki. At all of these sites, with the exception of Gävle, monitoring data from other measurement sites were also available for assessment of the model and these are additionally used here. These observational data may not necessarily cover the same period as the receptor modelling. The results presented here are based on all the available $PM_{2.5}$ data.

It is important to note that in this, as well as the other studies carried out, that there are significant uncertainties associated with the PM measurements themselves, up to 30% for hourly values. These uncertainties depend on the chemical composition of the particles and on the method used.

5.1 Oslo

Dispersion modelling was carried out for the period January to May 2004 in Oslo. Figure 5.1 shows the results of the hourly model calculations and observations for $PM_{2.5}$ concentrations at the RV4 site. The model can be seen to severely over predict $PM_{2.5}$ concentrations on a number of occasions, these being mostly in the January–February period. These over predictions are seen to occur when wind speeds are low, i.e. < 2 m/s as measured at 25 m, and the peaks are chiefly due to wood burning contributions. In the period from March and onwards, outside of the wood burning season, there is a slight tendency to under predict the $PM_{2.5}$ concentrations.

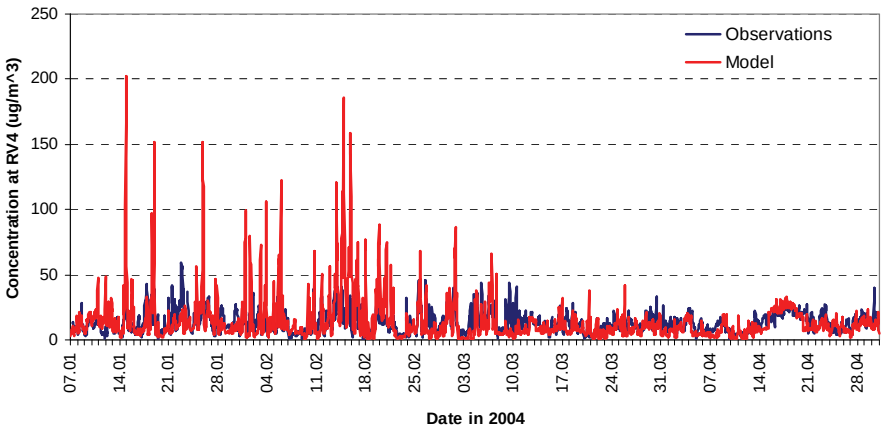


Figure 5.1. Hourly mean concentrations from the model calculations (red) and observations (blue) for $PM_{2.5}$ at the RV4 station in Oslo for the winter period of 2004.

Table 5.1 provides an overview of the performance of the model at all four sites. The statistics presented in this table are based on daily average concentrations at the various sites. The results are fairly consistent from one station to the next. Despite the overestimates seen in figure 5.1 there is a general tendency for the model to slightly under predict the $PM_{2.5}$ concentrations, from -3% to -27%. The model is found to explain between 32 % and 44% of the variability (correlation) and the root mean square error (RMSE) is seen to be around 60% of the observed average concentrations for all stations.

Table 5.1. Summary assessment of model performance at the four $PM_{2.5}$ sites in Oslo for daily mean concentrations.

Station	All stations		RV4		Aker Hospital		Kirkeveien		Løren	
Jan–May (103 days)	Obs	Mod	Obs	Mod	Obs	Mod	Obs	Mod	Obs	Mod
MEAN (µg/m^3)	13.9	12.3	16.3	12.8	10.6	10.7	13.2	12.8	15.3	13.0
STD (µg/m^3)	6.2	9.7	6.6	9.1	4.8	9.1	5.6	10.3	6.2	10.1
REL BIAS (%)		-12		-27		+1		-3		-18
Correlation (r^2)		0.36		0.32		0.32		0.44		0.43
RMSE (µg/m^3)		7.9		8.4		7.5		7.7		7.9
Contribution from wood burning		40%		37%		43%		48%		35%

Measured (TEOM) and estimated (based on the dispersion model) $PM_{2.5}$ concentrations from January 2004–May 2004 (103 days). Included are the results from the observations (Obs), the dispersion model (Mod).

Though the model results on a daily mean basis can be considered to be acceptable for the $PM_{2.5}$ concentrations, the hourly concentrations have been found to be over sensitive to wind speeds. When these are low the concentrations can become unrealistically high. Also included in table 5.1 are the relative contribution of wood burning, as calculated by the model, for the different stations. This is quite large, at 40% for all stations. This can be compared to the estimated contribution of long-range transport that is around 25% for the same period.

5.2 Lycksele

Model calculations have been carried out for Lycksele for the period 16.01.2002 to 09.03.2002 which corresponds to the period in which daily filter samples were taken. Two stations, Forsdala and Norrmalm are available for comparison with the model. Figure 5.2 shows the calculated and observed daily mean $PM_{2.5}$ concentrations at Forsdala during this period. There is a tendency for the model to overestimate concentrations but otherwise the model is performing relatively well.

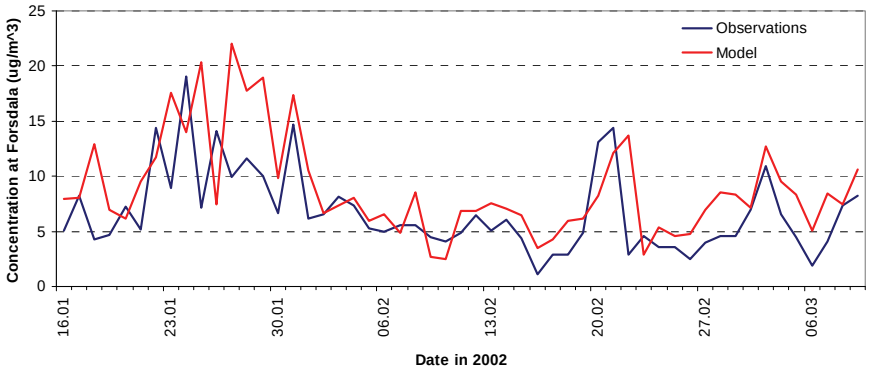


Figure 5.2. Daily mean concentrations from the model calculations (red) and observations (blue) for $PM_{2.5}$ at the Forsdala site station in Lycksele for the period 16.01.2002 to 09.03.2002.

The statistical assessment of the model performance for this period is presented in table 5.2. The total contribution of wood burning from the model is seen to be quite high, at 67%. The remaining contribution is due almost entirely to regional background contributions. Road traffic exhaust emissions at the Forsdala site contributes to less than 1% of the total $PM_{2.5}$ concentration. It should be reminded that the road, brake and tire wear is not included in the model and these sources make the most important contributions for $PM_{2.5}$ from road traffic as shown by Krecl et al., (2008b).

Table 5.2. Summary assessment of model performance at the two $PM_{2.5}$ sites in Lycksele (Forsdala and site R) for daily mean concentrations.

Station	Forsdala		Norrmale	
16.01.2002–09.03.2002	Obs	Mod	Obs	Mod
MEAN (µg/m^3)	6.7	8.9	6.4	8.4
STD (µg/m^3)	3.7	4.5	2.4	3.8
REL BIAS (%)		+24		+23
Correlation (r^2)		0.32		0.10
RMSE (µg/m^3)		4.4		4.26
Contribution from wood burning		67%		61%

Results from the period 16.01.2002 to 09.03.2002 (50 days) are show. Included are the results from the observations (Obs) and the dispersion model (Mod).

5.3 Helsinki

Model calculations have been carried out for Helsinki for the entire year. Two stations, Vallila and Kallio, are available for comparison with the model. Figure 5.3 shows the calculated and observed $PM_{2.5}$ concentrations for Vallila, a traffic station. There is a tendency for the model to overestimate concentrations in the winter months but otherwise the model is performing relatively well.

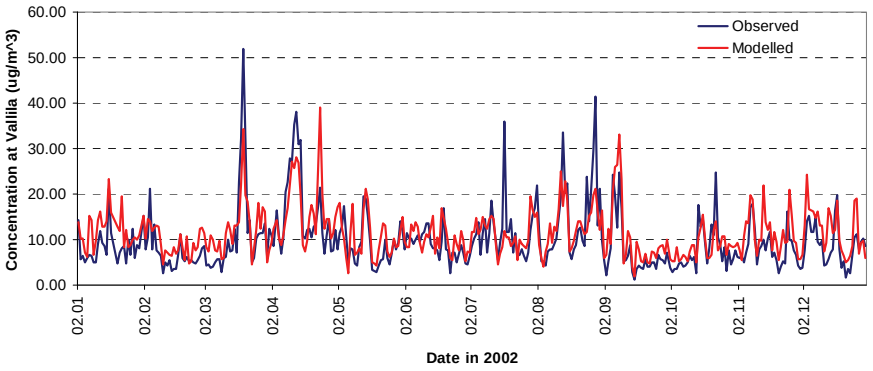


Figure 5.3. Daily mean concentr ations from the model calc ulations (red) and ob servations (blue) for $PM_{2.5}$ at the Vallila site station in Helsinki for the entire year 2002.

The statistical assessment of the model performance for the five winter months of November–March is presented in table 5.3. The total contribution of wood burning is seen to be quite low, however the contribution of long-range transport to the concentrations at these two sites is estimated to be around 75%.

Table 5.3. Summary assessment of model performance at the two $PM_{2.5}$ sites in Helsinki (Kallio and Vallila)

Station	Kallio		Vallila	
Nov, Dec, Jan, Feb, Mar	Obs	Mod	Obs	Mod
MEAN (µg/m^3)	7.1	7.9	8.2	11.2
STD (µg/m^3)	5.3	4.2	6.0	4.7
REL BIAS (%)		+ 10		+ 26
Correlation (r^2)		0.62		0.43
RMSE (µg/m^3)		2.6		4.5
Contribution from wood burning		5%		4%

Based on daily mean concentrations for the 5 winter months of January–March, November–December 2002. Included are the results from the observations (Obs), the dispersion model (Mod).

5.4 Uncertainty assessment and sensitivity of the dispersion models

5.4.1 Oslo

In previous studies it has been found that the model calculations for wood burning were most sensitive to the vertical distribution of the wood burning emissions in the Eulerian model. To further assess the sensitivity of the model to this a number of sensitivity runs were carried out in which the wood burning emissions were distributed differently in the lowest 3 layers of the model. These lowest 3 layers have depths of 14 m, 14 m and 43 m. In the standard model runs the wood burning emissions are evenly distributed into all 3 layers. The sensitivity runs redistributed the emissions through these three layers in different ways. The resulting changes, relative to the standard model runs, in average concentrations are presented in table 5.4.

Table 5.4. Results of the sensitivity analysis of AirQUIS to changes in the vertical distribution of wood burning emission.

Value	Layer 1	Layer 2	Layer 3	Layer 1+2	Layer 2+3	Layer 1+2+3 (Standard)
Average ($\mu g/m^3$)	4.16	3.12	1.77	3.64	2.10	2.51
Relative change compared to the standard model	1.66	1.25	0.71	1.45	0.84	1.00

Results are presented as the average over the entire model domain (22 x 18 km).

The spatial distribution of this sensitivity is shown in figure 5.4, where the relative change in concentration is shown for the case where the emissions were distributed in the lowest two layers (layer 1+2)

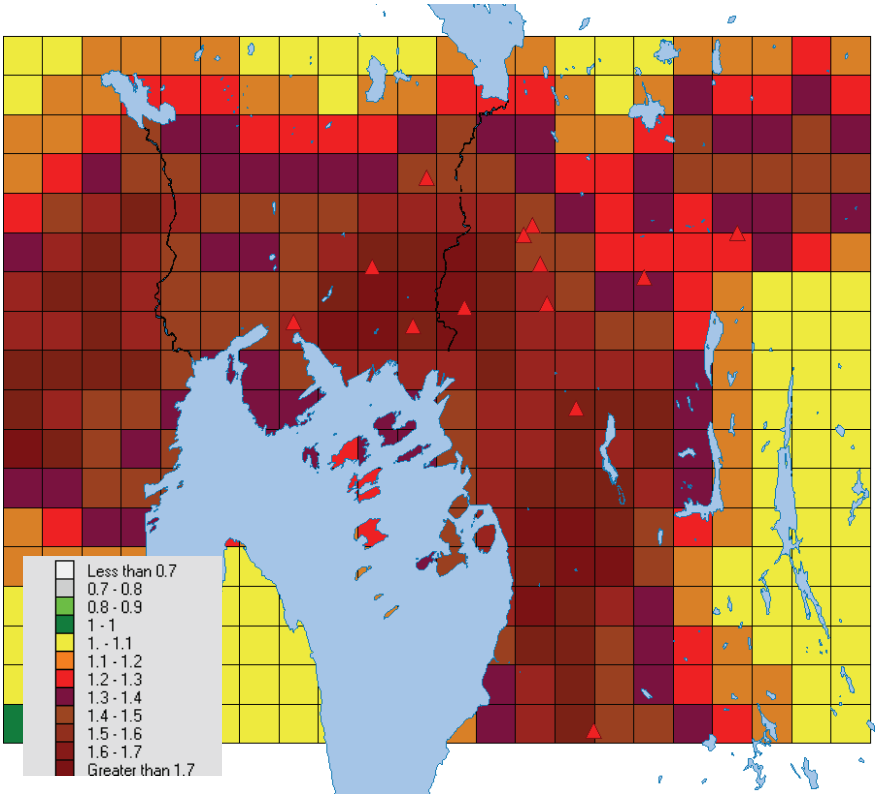


Figure 5.4. Spatial distribution of the relative change in concentrations due to the vertical redistribution of the wood burning emissions. In this case the emissions were evenly distributed in the lowest 2 layers (layer 1+2) instead of the standard run where they are distributed in the lowest 3 layers (layers 1+2+3).

This sensitivity study alone indicates that significant bias can be introduced into the model by the selection of this one parameterisation. The current choice of a 3 layer distribution is based, to a large extent, on a comparison with observations. However, if there is an emission bias then this model parameterisation choice will introduce a correcting bias.

Based on the comparison with observations and on the sensitivity of the model to vertical distributions we conclude that the model uncertainty, in long term mean concentrations, is of the order of 50% or more for the wood burning contribution.

5.4.2 Gävle

Airviro can apply either Eulerian models or Gaussian models for simulating the transport and emissions associated with wood burning. In this study, the sensitivity of the results to the following are assessed:

- The type of dispersion model (Lagrangian Gaussian or Eulerian grid model)
- Horizontal resolution of the calculations
- If emissions are input as area sources without plume lift or as point sources with plume lift.

The assessment of the importance on the calculated concentrations is based either on a time series of meteorological data (January 2004 or the whole year of 2004) or on the climatology, which consist of 360 weather conditions (see above). The reason for performing the assessment on different meteorological data sets is to ensure that the conclusions are not biased due to meteorology. NO_x emissions are used for the sensitivity analysis.

5.4.3 Assessment based on real time meteorology for one winter month:

Calculated NO_x concentrations for January 2004 from the two source types using the grid and Gaussian model are compared in table 5.5. Identical calculation areas with 176 x 122 grid cells and a horizontal resolution of 250 meters have been used in these calculations.

The ratios of the concentrations as obtained from the two different models are presented in table 5.6. This shows that the grid model gives on average higher values by a factor 2.41 as compared to the Gaussian model when the source type is an area type. But when the source type is a point source (with plume behaviour) the opposite is true, the grid model gives 24% lower concentrations. It can also be seen that the same tendency is true also for all higher values in the calculation domain, i.e. when looking at the 75 and 95-percentile values and the maximum value. These higher values represent grid cells that are closest to the wood combustion sources (since wood combustion is the only source included in the emissions).

Table 5.5. Calculated monthly average NO_x concentrations for January 2004.

Source type	Model	Average of all grid cells[1)]	90-percentile of all grid cells[2)]
Area	Grid	0.18	0.40
Area	Gauss	0.073	0.17
Point	Grid	0.055	0.13
Point	Gauss	0.073	0.18

Horizontal resolution is 250 meters. Note that all values are monthly averages. Thus, “Maximum” means the maximum monthly average of all grid cells in the calculation domain and so on.
[1)] Average of all grid cells in calculation area (176x122 cells).
[2)] The 90-percentile of all grid cells represents receptor points close to the emissions.

Table 5.6. Ratio of grid and Gaussian modelled averages of the concentrations presented in table 5.5 for the area and point source type respectively.

Source type	Average	90-percentile
Area	2.41	2.33
Point	0.76	0.74

Table 5.7 shows the ratios of the concentrations obtained based on area versus point source. It is seen that the grid model gives much higher concentrations when the emission is input as an area source as compared to point source (3.19 times higher on average and 3.02 for the 90-percentile). This is very different when the Gaussian model is used. Then the concentrations are about equal for the area and point source type.

Table 5.7. Ratio of Area and point source type results of the concentrations presented in Table 5.5 for the grid and Gaussian model, respectively.

Model	Average	90-percentile
Grid	3.19	3.02
Gauss	1.00	0.96

5.4.6 Assessment based on climatology:

In this section model calculations are based on the climatological meteorological data. Then the values correspond to annual averages. Table 5.8 shows comparisons of calculations based on different geographic resolutions (500 and 1,000 meters horizontal grid), emission types (area or point), and different models (Gaussian or grid) using the climatology as input for the calculations. Looking at the highest values (75 and 90-percentile values), which are closest to the emissions, the following conclusions can be drawn:

- *Horizontal resolution:* Increasing horizontal resolution from 1,000 to 500 meters gives somewhat higher values in the case of the Gaussian model for both point and area source type. In the case of the grid model similar values are obtained in the case of the area source but around 40% higher values are seen with point sources.
- *Source type:* Switching from representing the emissions as point sources with plume lift to area sources does not affect the results when the Gauss model is used, but substantially higher concentrations are seen with the Eulerian grid model.
- *Model type:* Comparing the two different model types it is seen that the grid model gives higher concentrations than the Gaussian model when the emissions are represented as point sources, but the opposite is true when the emissions are represented as area sources.

These results are thus consistent with the results when only January 2004 is used as input for the calculations.

Table 5.8. Ratios of concentrations for different geographic resolution, different source types, different models.

Resolution:			**Average of all cells**	**The 90-percentile highest cells***
Gauss	Area	500/1000	1.09	1.11
	Point	500/1000	1.12	1.17
Grid	Area	500/1000	1.00	0.96
	Point	500/1000	1.34	1.44
Emission type:				
Gauss	area/point	500	0.90	0.88
	area/point	1000	0.93	0.93
Grid	area/point	500	4.77	5.02
	area/point	1000	6.40	7.54
Model:				
Gauss/grid	area	500	0.37	0.35
	area	1000	0.34	0.30
	point	500	1.95	2.00
	point	1000	2.32	2.46

Calculations are based on the climatology. NOTE that the area source in the grid model is 5 m
* This is close to the source since the highest concentrations are obtained near emissions.

5.5 Conclusion

Considering the emissions as point sources with plume lift or simply as area sources introduces uncertainties depending on what type of model is used:

- With a grid model there can be a factor of 3 or more difference
- With a Gaussian model it is of less importance
- The Airviro grid model gives
- Much higher concentrations than the Gaussian model if area sources are used
- Similar concentrations if point sources with plume lift are used.

It seems that Gaussian models are better suited for describing concentrations and exposure due to wood smoke emissions in residential areas with many point sources (plumes) and where a high spatial resolution is required in order to capture the gradients in the concentrations. As a result of the sensitivity study, and experience with other applications of the model, then the uncertainty of the dispersion model calculations for wood burning sources can be defined as being large, factor of 3, when near the source but diminishing with distance from the source to an estimated 50%.

6. Inverse modelling using multiple linear regression

Multiple Linear Regression (MLR), as described in section 3.4, of the modelled source contributions was applied to the observed total $PM_{2.5}$ concentrations for the three cities of Oslo, Lycksele and Helsinki. The application was not limited in Oslo or Lycksele to just the filter data used in the receptor modelling but was also applied to other available data. The results of the inverse modelling results are described here.

6.1 Oslo

6.1.1 Application to all available stations for the 103 day winter period:

For Oslo four sources described as the regional background, traffic induced suspension, wood burning and other area sources are included in the MLR. $PM_{2.5}$ from traffic exhaust is not included as it is highly correlated, r^2=0.84 in the model, with the traffic induced suspension source. Traffic induced suspension was chosen, over traffic exhaust, for two reasons. Firstly $PM_{2.5}$ emissions from exhaust are better defined than those from suspension and secondly the receptor modelling carried out indicates a large discrepancy between the modelled and observed $PM_{2.5}$ contribution to traffic induced suspension. An alternative to choosing just the one traffic source is to lump them in a single source, however, this does not change the result of the regression to any significant extend.

MLR is carried out on all four stations simultaneously for the 103 day winter period. Any model source contributions not included in the MLR are subtracted from the total $PM_{2.5}$ concentration before the regression and added again when the regression model is calculated. The results of the regression in terms of the calculated regression coefficients, including their uncertainty, are shown in table 6.1 for two different cases. The first where the regression factors are determined for all four selected sources, and the second where only the two most significant sources, i.e. wood burning and traffic induced suspension (road dust and salt), are fitted. In both cases the regression indicates that the model is overestimating the contribution from wood burning and significantly underestimating the contribution from traffic induced suspension. The source contributions averaged over the four stations and for the 103 day period are also shown in figure 6.1.

Table 6.1. Multiple linear regression slopes determined for the various model sources of $PM_{2.5}$ for the 103 days in the period January 2004–May 2004 as well as the 38 filter days (based upon daily mean values).

	103 day winter period		38 filter days	
Emission sources	4 sources	2 sources	4 sources	2 sources
Regional background	1.22 ± 0.07		0.93 ± 0.06	
Traffic induced suspension	7.6 ± 1.0	8.6 ± 0.76	9.8 ± 1.7	7.8 ± 1.1
Wood burning	0.30 ± 0.06	0.29 ± 0.05	0.41 ± 0.12	0.33 ± 0.09
Other area sources	0.75 ± 0.42		-0.03 ± 0.83	

All four available stations are included in the analysis. Uncertainty estimates show standard deviations of the slope parameters using bootstrapping methods.

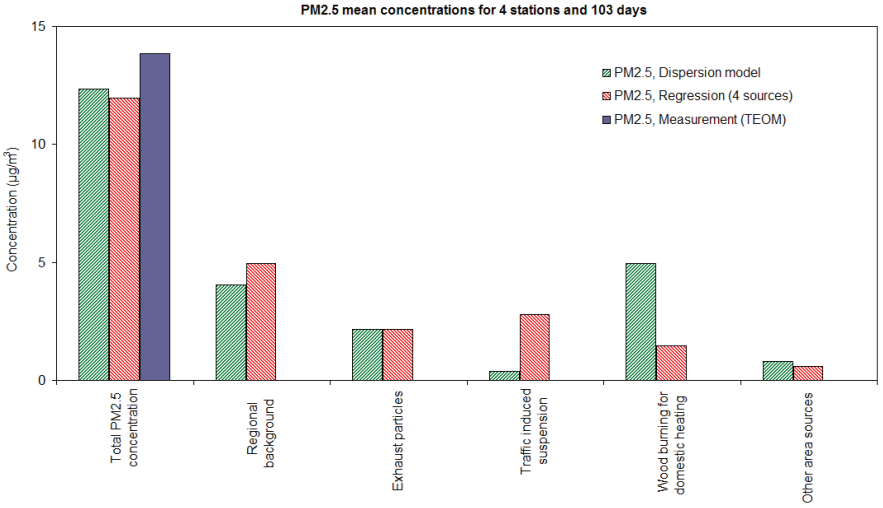


Figure 6.1. Estimated mean contributions to $PM_{2.5}$ ($\mu g\ m^{-3}$) from the different source categories using data from all four monitoring stations over the 103 day period. Shown are the dispersion model (green), the multiple linear regression (red) and the observed total $PM_{2.5}$ (blue).

Scatter plots showing the results of both the model calculations and the MLR calculations are shown in figure 6.2 to indicate the effect of the application of MLR on the data. The results of the MLR for the 103 day period can be summarized as follows:

- The most dominant source in the regression is wood burning, having the highest coefficient of determination, followed by traffic induced suspension. There is little improvement when regional background contributions and other area sources are included in the regression, either in correlation or RMSE.
- Correlation (r^2) increases from 0.36 to 0.50 with the application of MLR and the RMSE decreases from 7.9 μgm^{-3} to 5.7 μgm^{-3}
- The uncertainty assessment in the regression slope indicates that the regional background, wood burning and traffic induced suspension are quite well statistically defined. There is a large uncertainty in the regression slope for the other area sources.

- The results for the entire measurement period (103 days) are also consistent with the 38 day filter period indicating that these 38 days are representative of the entire study period.

6.1.2 Application to the available filter days:

In order to compare the results of the MLR with the receptor models, Chapter 4, MLR was also applied to the 38 filter days using all the available stations. The results are also shown in Table 6.1 and are similar to the results for the 103 day period except that the uncertainties are around twice as large due to the reduced sampling set. The modelling confirms the differences found in the previous section for the 103 day period, i.e. a significant under prediction of the traffic induced suspension and an over prediction of the wood burning contribution by the dispersion model.

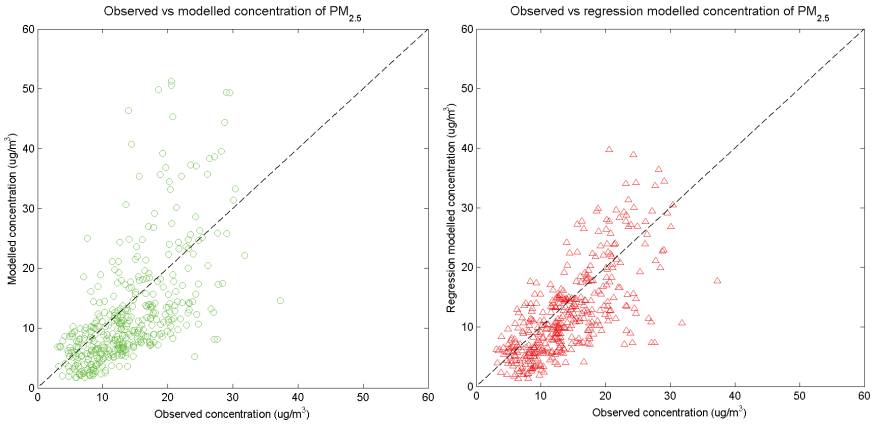
Conclusions:

Although the measured and modelled total $PM_{2.5}$ concentrations are, on the average, in good agreement at all sites in Oslo, MLR as an inverse modelling technique has shown large deviations for individual sources that compensate when combined together. The largest deviations are revealed for wood burning and traffic induced suspension where the optimal contributions differ from the dispersion model by a factor of 0.30 and 7.6, respectively.

The difference between the modelled and observed daily mean concentrations can be accounted for by either errors in the emission inventories, in the model formulation or in the meteorological input data. For the case regarding wood burning, which is modelled using the Eulerian grid model, it is not strictly possible to distinguish between these two uncertainties and it may well be that model formulation, e.g. vertical dispersion, initial emission heights or wind speeds are partly or wholly responsible for the differences found. Indeed, the days when measured concentrations are strongly over-predicted by the dispersion model due to wood burning contributions are also days characterised by measured wind speeds of < 2 ms^{-1}. Sensitivity studies concerning the vertical distribution of the wood burning emissions in the dispersion model also show large variations in model concentrations depending on the height at which emissions are introduced into the model grid. Another important source of uncertainty is the meteorological field generated by the diagnostic model, particularly in an urbanised area. Based on the current knowledge and available observational data, particularly meteorological, it is not possible to come to any firm conclusions regarding the cause of the differences found for the wood burning contribution.

For the case of traffic induced suspension the model, a Gaussian line source model, is less affected by uncertainty in meteorological conditions relating to dispersion or emission heights compared to the grid model. In

this regard there is more confidence in the results of the line source model than the grid model. It should also be noted that the regression analysis cannot distinguish between exhaust emissions and traffic induced suspension due to the high correlation of these two sources in the model. In the results presented here we have assumed that the exhaust contribution is correct and any deviation is due to the traffic induced suspension.



a) b)

Figure 6.2. Dispersion model calculations verses observations of daily mean $PM_{2.5}$ using the four stations for the 103 day period. a) Model calculations without adjustment, b) Results after the multiple linear regression, as given in table 6.1.

6.2 Lycksele

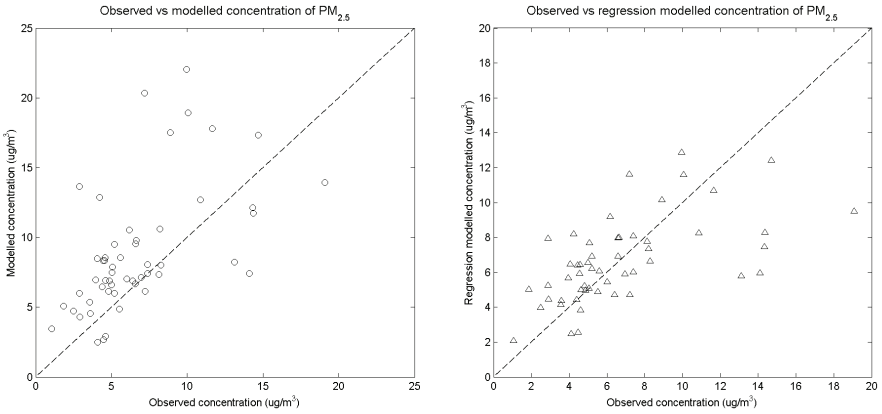
The methodology adopted for the Oslo filter samples was also applied to the Lycksele filter sample set from Forsdala. In this case model source categories included only domestic wood burning and traffic exhaust. Background concentrations for the model were taken from the near by Vindeln site (a monitoring station of the Cooperative Program for Monitoring and Evaluation of the Long-Range Transmissions of Air Pollutants in Europe (EMEP), situated in a forest 65 km southeast of Lycksele). The filter measurements of $PM_{2.5}$ used in the receptor modelling from the Forsdala site were also used for the MLR. Hourly automatic measurements carried out at site R were averaged to provide daily means for the same days. The modelled traffic exhaust PM contribution was very low < 1% at both stations, which are in residential areas. Higher traffic contributions are expected if road and brake wear were be considered. The fitting procedure was extremely uncertain for the traffic contributions at this station and so the traffic element was dropped from further analysis at this site.

Table 6.2 presents the results of this assessment and figure 6.3 provides the scatter plots of the model and regression calculations.

Table 6.2. Multiple linear regression slopes determined for the various model sources of $PM_{2.5}$ for the period 16.01.2002 to 09.03.2002 for the Forsdala station in Lycksele (50 daily mean samples)

Emission sources	Forsdala
Regional background	1.41 ± 0.19
Traffic	–
Wood burning	0.43 ± 0.11

Uncertainty estimates show standard deviations of the slope parameters using bootstrapping methods.



a) b)

Figure 6.3. Dispersion model calculations verses observations of daily mean $PM_{2.5}$ for the 50 sample days at the Forsdala site. a) Model calculations without adjustment, b) Results after the multiple linear regression, as given in table 6.2.

The results of the Lycksle MLR can be summarised as follows:

- The observed background and modelled local wood burning contribution are quite well correlated with each other ($r^2 = 0.61$). This indicates that the background contribution is influenced by the same meteorology as the local contributions. I.e. that the background is likely to be of a more local origin rather than being the result of long range transport. This is in contrast to the Helsinki and Oslo results that indicate very little correlation of the model with the background observations as these are significantly more distant.
- The model appears to be overestimating the wood burning contribution by a factor of 2.3 (regression slope of 0.43) with an associated uncertainty of around 25%.

- The regression also indicates that the regional background contribution, estimated from the site at Vindeln, is underestimated by around 30%, with a regression slope of 1.4. Uncertainty in this parameter, based on the bootstrapping assessment, is around 15%.

6.3 Helsinki

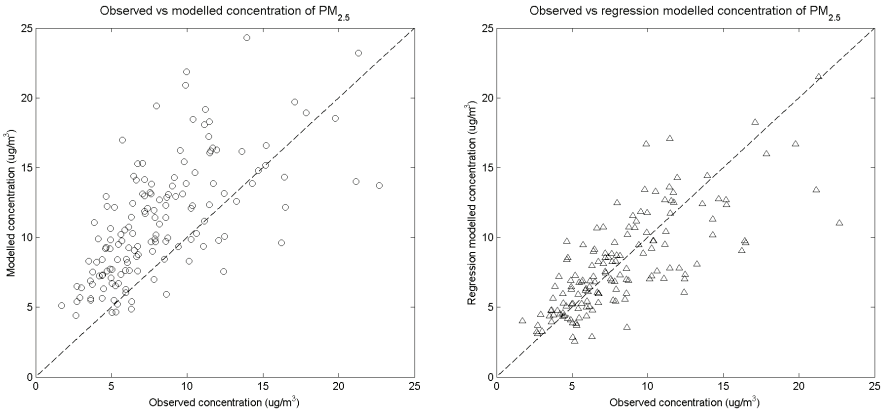
MLR was applied to the daily mean $PM_{2.5}$ data for the two stations in Helsinki, Vallila and Kallio, for the 5 winter months for November–March. Model concentrations from the three sources of long range transport, wood burning and traffic were assessed. The application followed very much the same lines as that for Oslo described in section 6.1.

Table 6.3 presents the results of this assessment and figure 6.4 provides the scatter plots of the model and regression calculations.

Table 6.3. Multiple linear regression slopes determined for the various model sources of $PM_{2.5}$ for the period January 2002–March 2002 and November 2002–December 2002 for the two stations in Helsinki, Vallila and Kallio.

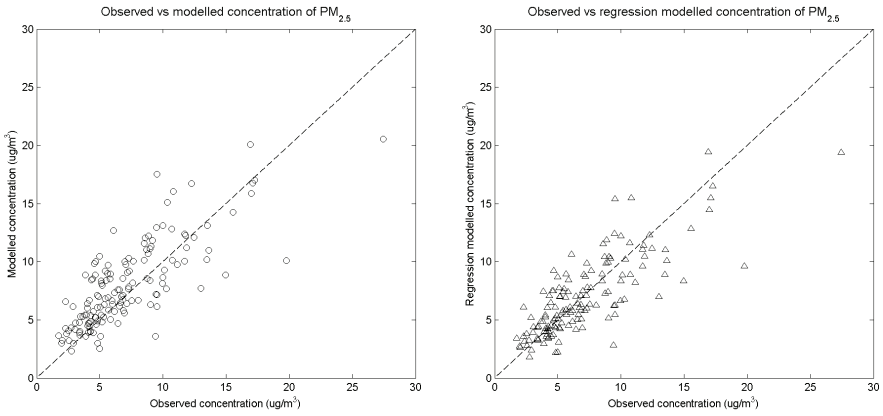
Emission sources	Vallila	Kallio
Regional background	1.06 ± 0.07	0.99 ± 0.05
Traffic	0.22 ± 0.09	0.38 ± 0.19
Wood burning	1.11 ± 0.45	0.72 ± 0.35

Uncertainty estimates show standard deviations of the slope parameters using bootstrapping methods.



a) b)

Vallila



c) d)

Kallio

Figure 6.4. Dispersion model calculations verses observations of daily mean $PM_{2.5}$ for the two stations Vallila and Kallio in Helsinki. Left (a. and c.) are the model calculations, and right (b. and d.) are after the applicat ion of the multiple lin ear regression, as g iven in table 6.3.

The results of the MLR in Helsinki can be summarised as follows:

- The three model sources are poorly correlated with each other. This is surprising for the traffic and wood burning emissions (r^2 = 0.05 and 0.12 for Kallio and Vallila respectively) since both wood burning and traffic emissions are subject to the same meteorology and model processes. Stronger correlations, (r^2 = 0.3–0.6) were found in the Oslo and Lycksele datasets. It is possible that the different daily cycle of the two emission sources in Helsinki, which occur at different times of the day, are poorly correlated because of a large variability in the meteorological cycle during the day.
- The background contribution dominates the local air quality at these sites with a contribution of around 25% at both sites, see section 5.3. The deduced regression slope for the background contribution is close to 1 with little error for both sites. This confirms the parameterisation developed by Kauhaniemi et al. (2008) using data from the year 2000.
- The traffic contribution adds a significant level of variability to the regression, at least in the Vallila case, but there remains an uncertainty in the slope of around 50%. However, at both sites the modelled traffic contribution seems to be overestimated by a factor of 3–5. This overestimation may be due to the re-suspension scaling factor for traffic emissions. Exhaust emissions are enhanced by a factor of 2.6 to include the effect of suspended road dust and salt emissions. This may lead to an overestimate of the traffic contribution by the model if conditions are not relevant for re-suspended road dust.

- The wood burning contribution is very small (5% of the total) and has a high uncertainty in the determined slope of around 50%. This indicates that wood burning may be smaller than the original estimates of 14%. However the uncertainty in this assessment is high and no concrete conclusions can be made based on this analysis.
- The large contribution of background concentrations makes the regression difficult to apply. The requirement when carrying out MLR (section 3.4), that the contributions from the different sources should be of a similar order, is not met in the Helsinki case.

7. Comparison of receptor, dispersion and inverse modelling

In this chapter we collect the results presented in chapters 4–6 in order to compare the estimated wood burning contribution at each of the sites using the receptor, dispersion and inverse modelling. In addition the results from the different sites are qualitatively compared. In Table 7.1 the results of the entire study are summarised. The various estimates of wood burning contributions are presented in both absolute and relative (%) terms.

Table 7.1. Summary of the mean wood burning contributions to $PM_{2.5}$ for the three case cities and periods.

Source	Oslo All days, all sites	Oslo Filter days, RV4 site	Lycksele	Helsinki Vallila	Helsinki Kallio
Total $PM_{2.5}$ mass (μgm^{-3})					
Observed	14.9	23.3	6.7	8.2	7.1
Modelled	12.3	18.9	8.9	11.1	7.9
Contribution from wood burning to $PM_{2.5}$ mass (μgm^{-3})					
UNMIX		8.1 ± 1.9 [U1] (36%)			
PMF (ME-2)		5.2 ±1.4 [U1] (24%)			
PMF-2		3.9 ± 1.6 [U2] (18%)	3.2 ± 2.9* [U4] (48%)		
COPREM		5.6 ± 0.3 [U1] (25%) 6.0 ± 0.9 [U3] (28%)	2.5 ± 2.5* [U4] (37%) 3.9 ± ** [U3] (55%)		
Dispersion modelling	5.00 ± 2.5 (40%)	7.3 ± 3.7 (39%)	6.0 ± 4.0* (67%)	0.5 ± ** (4%)	0.4 ± ** (5%)
Inverse modelling	1.7 ± 0.4 (13%)	3.0 ± 0.8 (14%)	2.5 ± 0.6 (37%)	0.5 ± 0.25 (4%)	0.3 ± 0.15 (4%)

Presented are results for the receptor site and all available sites. Also included are the percentage, in brackets, contribution from wood burning to the total concentration as determined by that particular method. The Oslo results are shown for the 2004 winter period only.

* Standard deviation based on variability rather than uncertainty

** No uncertainty assessment carried out

[U1] Applied by User 1.

[U2] Applied by User 2.

[U3] Applied by User 3.

[U4] Applied by User 4.

7.1 Comparison of wood burning source apportionment in Oslo

The Oslo receptor modelling presents a range of values for the contribution of wood burning to the measured $PM_{2.5}$ concentration at the RV4 site. The highest estimate is obtained using UNMIX and the lowest was obtained using PMF-2. It is interesting to note that even though PMF-2 should be working on a similar principle to PMF (ME-2) the two models were applied at different times, by two different users and with two different measurement uncertainty estimates. The difference between the two results reflects these factors. The COPREM model, which is partly based on a-priori knowledge of the source profiles, is probably the most well based estimate.

The dispersion model calculations for Oslo overestimate the contribution of wood burning in comparison to all the receptor models. Its uncertainty is considered to be high at around 50%. Application of the MLR to the dispersion model, at both independent sites and the RV4 receptor site, leads to an underestimate of the wood burning contribution in respect to the receptor modelling. This was previously explained, in section 6.1, as being partially the result of model error in regard to the effects of wind speed and vertical distributions of wood burning emissions. The uncertainty estimate for the MLR must be regarded as the minimum uncertainty of the technique.

In figure 7.1 the daily variation of the wood burning contribution, as calculated by the range of receptor, dispersion and inverse models is presented. There is a significant amount of variability between the various methods on a daily basis.

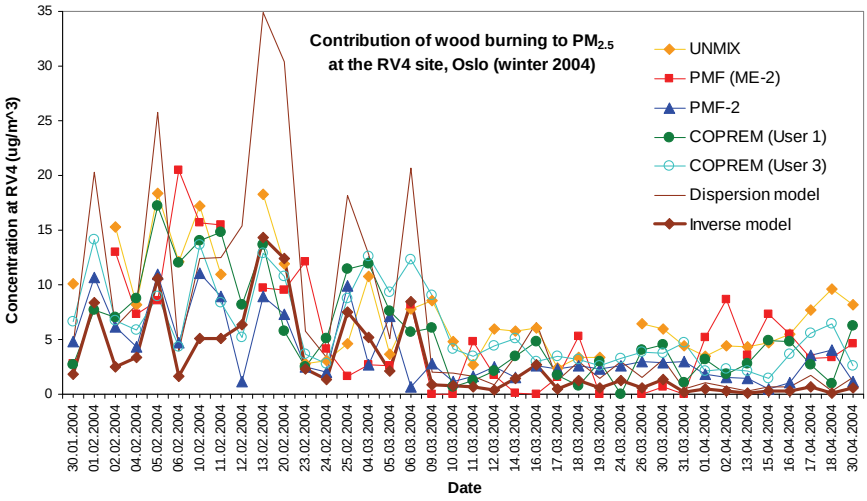


Figure 7.1. Plot showing the different estimates for the temporal variation of wood burning in Oslo for the winter of 2004. 38 days are shown corresponding to the filter samples taken during this period.

7.2 Comparison of wood burning source apportionment in Lycksele

For Lycksele three receptor modelling results are available for comparison with the inverse modelling. The receptor modelling results are similar (table 7.1) but cover a range form 2.5–3. 9 μgm^{-3} with the largest deviation being from the COPREM (User 3) results. The inverse modelling mean wood burning contribution is quite similar to the receptor model calculations.

Time series plots of these results are shown in figure 7.2. As in the Oslo case the dispersion model overestimates by a factor of 2 the contributions from wood burning at this site.

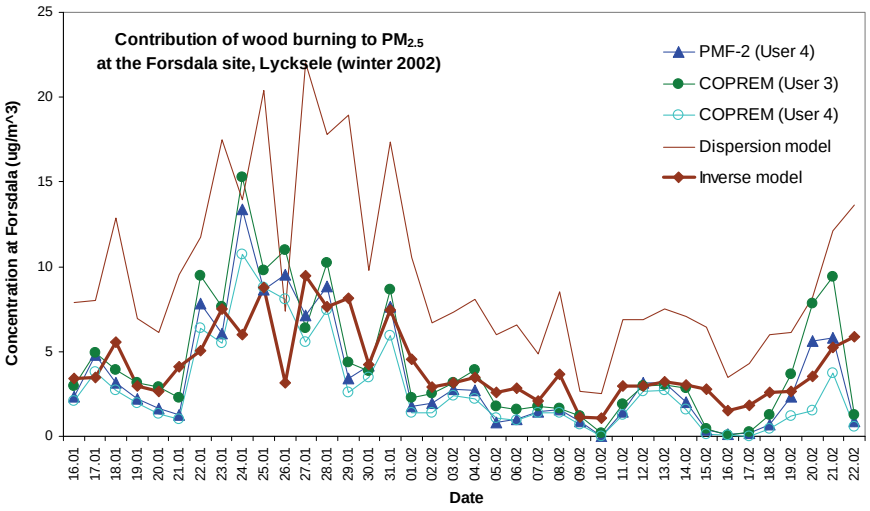


Figure 7.2. Plot showing the different estimates for the temporal variation of wood burning in Lycksele for the winter of 2002. 50 days are shown corresponding to the filter samples taken during this period.

7.3 Wood burning source apportionment in Helsinki

For Helsinki, without any receptor modelling and low model contributions from the wood burning emissions, there is very little that can be concluded in regard to the wood burning except that the results of the MLR confirm the expectations of the contribution of wood burning to the $PM_{2.5}$ concentrations in Helsinki.

7.4 Comparison of Oslo and Lycksele results

There are two important similarities between Lycksele and Oslo. In both cases the dispersion model calculations overestimate, in regard to the receptor modelling, the contribution of wood burning by a factor of 2. In both cases the inverse modelling captures this difference and provides improved modelling estimates of the wood burning contribution. This is despite the fact that two different models were applied to the two areas and that the scale and type of wood burning emissions are quite different. In Oslo home heating through individual ovens is prevalent whilst in Lycksele larger centralised boilers are employed.

In addition to the different characters of the wood burning emissions the traffic contribution in Oslo, at least at the sites studied, is much more significant than in Lycksele. The different mix of sources will tend to influence the results, dependent on the degree of correlation between these.

8. Discussion and conclusions

This study has implemented receptor, dispersion and inverse modelling techniques in order to derive source contributions and emissions strengths related to domestic wood burning. Four Nordic cities were included in the study, these being Oslo (Norway), Helsinki (Finland), and Lycksele and Gävle (Sweden). A range of receptor and dispersion models have been used in the study. The results show that dispersion models applied in Oslo and Helsinki overestimate the wood burning contribution compared to receptor modelling. The use of multiple users and receptor models shows significant variation between receptor modelling results, but the variation is less sensitive for the wood burning contributions than it is to other sources. The study has also shown that inverse modelling techniques, based on modelled source contributions and measured $PM_{2.5}$ concentrations, give similar results to the receptor models.

8.1 Uncertainty

A significant amount of attention was given to the question of uncertainty in this study. Model uncertainty was assessed through comparison with observations and through sensitivity studies. The models, particularly the Eulerian models, were seen to be sensitive to the vertical distribution of wood burning emissions. For Lycksele wood burning emissions are treated as point sources and plume rise models are employed. In Oslo the emissions are evenly dispersed in the lowest three layers of the model. In Helsinki emissions at a constant height of 7.5 m were used. Changing the manner in which the emissions are distributed could result in a factor of two difference in the long term concentrations. This source alone will lead to a significant uncertainty in the wood burning concentrations that is not stochastic in nature and so cannot be reduced with longer integration times. The traditional manner of assessing model uncertainty, through comparison with observations, does not allow us to disaggregate the emission/dispersion model uncertainty.

The receptor models employed show a large degree of consistency in their results for the wood burning source. Other sources, such as brakes and road dust, show more variability. Even so the range of values obtained with the different models, and not the least different users, indicates that there is still significant uncertainty when carrying out receptor modelling. Uncertainty may be reduced by the independent implementation of different receptor models by different users, or at the least by co-

operative iteration between users. There remains a subjective element to receptor modelling that cannot be easily reduced.

The uncertainty in the inverse modelling using multiple linear regression is more difficult to define. The boot strapping method employed provides some information on the uncertainty derived from the limited sampling size but does not provide more information. One important role of the inverse modelling in the form applied here is that it can remove model bias, such as that resulting from erroneous vertical distribution of the emissions, from the results. In this way it can reduce the uncertainty attached to the modelling but it will not decrease the uncertainty attached to the combined emission / dispersion modelling.

We summarise the estimated uncertainties for wood burning as follows:

- Estimated uncertainty of the dispersion models ~50–100%
- Estimated uncertainty in the source contribution form receptor models ~25%
- Estimated uncertainty in the inverse modelling method ~30%

8.2 What can we say about the emissions?

The main question asked at the beginning of this study is “Can we improve emission estimates of $PM_{2.5}$ form wood burning through indirect methods?” The indirect methods we considered were ambient air observations, chemical analysis, receptor modelling, dispersion modelling and inverse modelling.

Though we are able to determine the source contribution of wood burning at measurement sites to around 30%, using receptor modelling and/or inverse modelling, this does not tell us the actual emissions. To provide this link dispersion models are required and it is the dispersion modal that provides the highest level of uncertainty in the analysis. The inference from this study is that we cannot determine the average wood burning emission strength using dispersion models to anything better than a factor of two. For shorter time or spatial resolutions the uncertainty must be considered to be even higher.

If we assume that the dispersion models themselves are unbiased, then the results from this study indicate that emissions inventories overestimate wood burning emissions by a factor of 2 for Lycksele and by a factor of 1.5–2 for Oslo, depending on whether the inverse or receptor modelling is used. If this is the case then this has important consequences for a city such as Oslo which must find ways of reducing the PM levels in the city. If the emissions are significantly less than those currently estimated then the introduction of new technologies and changes in heating habits will not have as large an impact as expected.

8.3 What can be done to improve the methodology?

It should be possible to reduce the level of uncertainty associated with this methodology for determining emissions through indirect methods. The most important improvements must come in the dispersion modelling. A number of recommendations for improving the models are given below:

- Improved descriptions of the vertical distributions of areal emission sources for wood burning. This will necessarily involve observational campaigns that look at the vertical distribution of wood burning emissions.
- Improved meteorological descriptions in the dispersion models that parameterise buildings in the urban canopy in a better way.
- Other means, e.g. through the controlled release of known emission strengths, are required to determine the model uncertainty if the combined emission/modelling uncertainty is to be decoupled.

There will always remain a level of uncertainty attached to receptor modelling. From the study carried out here it is important to identify good "markers" for the sources being analysed. Levoglucosan has been found to be such a source though the study at Lycksele showed the results to be robust without the use of this marker. Another quite promising technique for improving the receptor modelling is the use of simultaneous measurements of both particles size distribution, particle mass and absorption in combination with PMF/COPREM, as shown for Lycksele by Krecl et al. (2008b).

The inverse modelling technique applied here, multiple linear regression, has been shown to be able to predict similar source contributions to those obtained through receptor modelling. The methodology has a number of limitations. These include the need for low correlation between the source categories and that the sources being considered are of a similar order of contributions. Even when this method is applied only a single scaling factor is available for each of the sources. A number of improvements in the method may be possible, including the use of hourly data instead of daily means. In previous applications hourly data has been seen to increase the noise associated with the regression but filter techniques may be applied to the data that will help improve the results. The use of hourly data provides the possibility of determining time varying scaling factors for sources, something that is also limiting in the methodology.

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