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ESTIMATING DOMESTIC WOOD BURNING EMISSIONS OF PARTICULATE MATTER IN TWO NORDIC CITIES BY COMBINING AMBIENT AIR OBSERVATIONS WITH RECEPTOR AND DISPERSION MODELS*

The major emission source of primary PM_{2.5} in many Nordic countries is wood burning for domestic heating. Though direct measurements of wood burning emissions are possible under controlled conditions, emission inventories for urban scale domestic heating are difficult to calculate and remain uncertain. As an alternative method for estimating these emissions, this paper makes use of ambient air measurements, chemical analysis of filter samples, receptor models, dispersion models, and simple inverse modelling methods to infer the emission strengths. A comparison of dispersion models with receptor models indicates that the dispersion models tend to overestimate the contribution from wood burning. The inverse modelling results are found to agree with those from the receptor modelling. 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 a dispersion model error can be significant. It is recommended to improve plume rise and urban canopy meteorological descriptions in the dispersion models before these models are of sufficient quality to allow quantitative assessments of emission inventories.

Key words: particulate matter; urban air quality; receptor modeling; dispersion modeling; inverse modeling; wood burning.

Fine particles, defined as particles with diameters < 2.5 µm (PM_{2.5}), have been associated with numerous adverse health effects (*e.g.* [1]). The Clean Air for Europe (CAFE) program estimates that fine particles cause over 300,000 premature deaths annually in Europe and that exposure to fine particles lowers the life-expectancy by 8.6 months in average [2]. 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}.

Wood burning is known to be a significant source of PM_{2.5} 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 vary from city to city but Oslo has an estimated contribution of up to 80% from wood combustion [3]. In Denmark wood burning accounts for around 47% of all Danish PM_{2.5} emissions [4]. Cities such as Helsinki have less significant contributions with wood burning estimated to account for just 24% of the local emissions [5]. However, in smaller cities in Finland and even Denmark wood burning is estimated to be the dominating emissions source.

Current emission rates of PM_{2.5} from wood burning are based on estimates of consumption and emission factors. These factors are taken from measurements of direct emissions from a variety of wood burning ovens [6,7]. The consumption can be calculated from surveys or from total wood consumption distributed over the population. There is a large uncertainty in these estimates, for both the consumption of wood and the emission factors.

The actual emissions will depend greatly on local heating habits, the quality of stoves and wood as well as on meteorological conditions. Pragmatically, the

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only way to assess the integrated emissions of $PM_{2.5}$ from a large number of wood burning stoves is to measure the ambient atmospheric concentrations of $PM_{2.5}$ and then use some inversion modelling technique to infer the emissions. If only the relative source contribution is to be determined then source-receptor modelling methods may be applied. This involves identifying compounds that are indicative of the source, measuring these compounds at representative monitoring sites and then applying source-receptor modelling [8]. These models allow the identification of source chemical profiles and, by further analysis, their relative contribution to the measured concentrations.

However, recognition of the relative contributions does not provide the information on the emission strengths. To determine this dispersion models are required to link the ambient concentrations to the emissions. The aim of this study is to determine if the emission rates of $PM_{2.5}$ from domestic wood burning can be estimated through indirect means. 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 have been applied to two Nordic cities, where relevant data and modelling activities have been carried out. These are the cities of Oslo (Norway) and Lycksele (Sweden). Complete details concerning the study can be found in Denby *et al.* [9].

METHODOLOGY

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 enables 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. In this study four different receptor models, with different users, have been applied (UNMIX, PMF (ME-2), PMF-2 and COPREM). The filter samples have a time resolution of 12-24 h.

The second method uses dispersion models and $PM_{2.5}$ mass measurements at a number of sites and applies a multiple linear regression (MLR), as an inverse modelling technique, to the modelled source contributions [10]. This form of inverse modelling provides regression factors for the model source categories, which can be interpreted to be scaling factors

for the emission strengths. In reality these factors indicate the optimal least squares fit of the modelling data to the available measurements and can reflect bias in not just the emission rates but also in the dispersion model itself. The dispersion models used are the Eulerian model in AirQUIS [11] and the Gaussian model in Airviro [12] for Oslo and Lycksele, respectively. Daily mean concentrations have been used for the inverse modelling.

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 (up to five different receptor model runs are applied). In the case of dispersion modelling the uncertainty is based on sensitivity analysis, expert knowledge and direct comparison with observations. Uncertainty in the inverse modelling is quantified by the use of boot strapping techniques which involves the generation of an ensemble of randomly selected data from the dataset. 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.

STUDY SITES AND OBSERVATIONS

Two Nordic cities are selected for this paper. These are Oslo (Norway) and the small town of Lycksele (Sweden). Both of these cities have been found in previous studies [3,10,13] to have a significant proportion of their $PM_{2.5}$ contribution originating in domestic wood burning. In regard to the sources of wood burning emissions, domestic wood burning in individual dwellings in ovens and fireplaces is a major contributor to the total wood burning emissions in Oslo. In Lycksele wood burning is often centralised at boilers and wood burning in individual dwellings is less frequent than in Oslo. The meteorological conditions in these cities vary but both cities are subject to strong stagnant winter time conditions where low wind speeds and strong temperature inversions can lead to severe episodic pollution events.

In Oslo, a traffic station, known as RV4, collected around 80 daily mean filter samples of $PM_{2.5}$ for chemical analysis used for the receptor modelling in two winter periods January-May 2004 and December-April 2005. In addition, three other stations continuously measuring $PM_{2.5}$ were used for the inverse modelling applications. In Lycksele, a single station (Forsdala) in a residential area was used to collect 103 12-hour filter

samples for chemical analysis and for continuous monitoring in the period January–March 2002. Chemical analysis of the filters varied at the two sites but elemental and ionic analyses were carried out at both. In Oslo Levoglucosan was also analysed as a wood burning tracer. Up to 22 elements and compounds were used in the receptor modelling.

RESULTS

Receptor models were applied to the available data in both Oslo and Lycksele. Figure 1 shows the results of 5 different receptor model runs and calculations for the Oslo dataset. All models and users are reasonably consistent in predicting the biomass contribution to PM_{2.5}. The exhaust contribution is also consistently predicted. The different receptor models identify different numbers of factors (sources) reflecting the need for user interpretation.

Dispersion and inverse model calculations were carried out for both cities. The results of the inverse modelling are summarised in Table 1. 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 uncertainty associated with these estimates is around 20–30 %. For the case of Lycksele the model appears to be overestimating the wood burning contribution by a factor of two (regression slope of 0.43) with an associated uncertainty of 25%.

A comparison of all the results for Oslo and Lycksele is shown in Figure 2 where the mean contribution of wood burning to PM_{2.5}, determined by the different methods is given. There is significant variability between the receptor models in Oslo, less in Lycksele, and in both cases the dispersion model seems to be overestimating the wood burning contribution. The inverse modelling confirms, independently of the receptor modelling, this overestimation by the dispersion models.

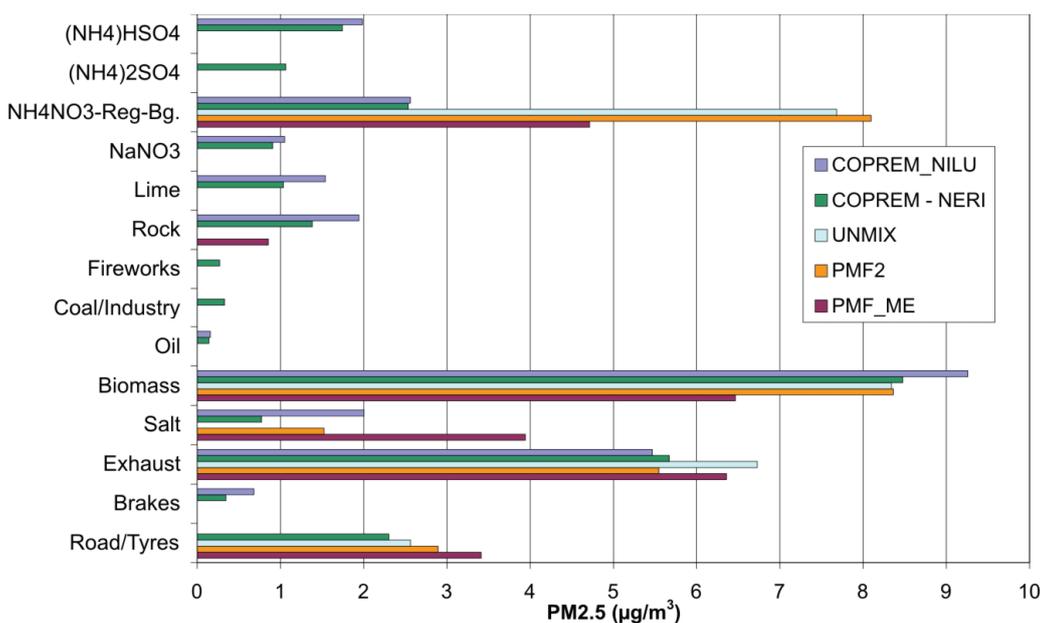


Figure 1. Source contributions for the 5 receptor model applications in the Oslo study. Note the different number of sources in the models and therefore the different subdivisions of the source contributions from “Regional background”(Reg-Bg) and “Exhaust”.

Table 1. Multiple linear regression slopes (scaling factors) determined for the various model sources of PM_{2.5} for Oslo and Lycksele. Uncertainty estimates show standard deviations of the slope parameters using boot strapping methods

Emission sources	Oslo (RV4)	Lycksele (Forsdala)
	4 Sources	2 Sources
Regional background	1.22±0.07	1.41±0.19
Traffic induced suspension	7.6±1.0	-
Wood burning	0.30±0.06	0.43±0.11
Other area sources	0.75±0.42	-

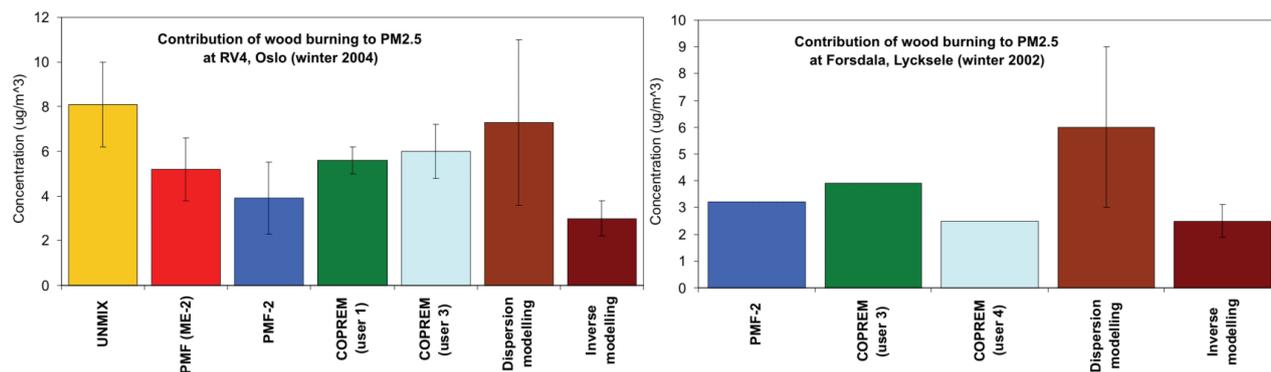


Figure 2. Mean contribution of wood burning to PM_{2.5} concentrations for Oslo (left) and Lycksele (right) as determined by the different receptor models, the dispersion model and the inverse modelling. Error bars indicate uncertainty estimates based on various methods.

CONCLUSIONS

In this study we have applied alternative methods to provide estimates of wood burning emissions in Nordic cities. These methods include the chemical analysis of daily filter samples, the application of receptor models to these analyses to determine source contributions, the use of dispersion models to provide source contributions and the application of inverse modelling to PM_{2.5} concentrations as an independent assessment of the source contribution to wood burning.

The results indicate that there is significant variability in receptor model calculations (25% for the Oslo application) that is indicative of the uncertainty in determining the source contributions using these methods. In both the cases of Oslo and Lycksele the dispersion model overestimates the contribution of wood burning when compared to the receptor modelling. The independent application of a simple inverse modelling technique, a multiple linear regression, confirms this overestimation.

Though we are able to determine the source contribution of wood burning at measurement sites to around 25% using receptor modelling, this does not tell us the actual emissions. To provide this link dispersion models are required and it is the dispersion model that provides the highest level of uncertainty in the analysis. The inference from this study, given the uncertainty in the dispersion models, is that we cannot determine the average wood burning emission strength to anything better than a factor of two using the current tools. Significant improvement in the description of the vertical emissions profiles used in the dispersion models is recommended, as the models have been found to be sensitive to this parameterisation.

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NAUČNI RAD

PROCENA LOKALNE EMISIJE ČESTICA SAGOREVANJEM DRVETA U DVA NORDIJSKA GRADA KOMBINOVANJEM POSMATRANJA VAZDUHA SA MODELIMA PRIJEMA I RASPROSTIRANJA

U Skandinavskim zemljama najveći izvor emisije primarnih čestica manjih od 2.5 µm (PM_{2.5}) je sagorevanje drveta koje se koristi za grejanje domaćinstava. Iako su direktna merenja emisije pri sagorevanju drveta moguća pod kontrolisanim uslovima, inventar emisije na nivou grada za zagrevanje domaćinstava se izračunava teško uz veliku nesigurnost. U ovom radu je za donošenje zaključka o jačini emisije sagorevanja drveta za zagrevanje domaćinstava korišćena alternativna metoda koja uključuje uzorkovanje ambijetnog vazduha, hemijske analize uzoraka filtera, receptorsko modelovanje, modelovanja rasprostiranja i primenu metode inverznog modelovanja. Rezultati dobijeni na osnovu modela rasprostiranja imaju tendenciju da preценjuju doprinos usled sagorevanja drveta u odnosu na rezultate dobijene receptorskim modelovanjem. Rezultati inverznog modelovanja se slažu sa rezultatima receptorskog modelovanja. Iako oba, receptorsko i inverzno modelovanje, preценjuju emisiju PM_{2.5} nije moguće to pripisati samo kao grešku inventara emisija, jer i greška modela rasprostiranja može biti značajna. Da bi modeli rasprostiranja bili dovoljno kvalitetni za kvantitativnu procenu inventara emisija, preporučuje se usavršavanje opisa vertikalnih profila emisija.

Ključne reči: čestice; kvalitet vazduha urbanih sredina; receptorsko modelovanje; modelovanje rasprostiranja; inverzno modelovanje; sagorevanje drveta.