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# Evaluation of the emissions and uncertainties of PM<sub>2.5</sub> originated from vehicular traffic and domestic wood combustion in Finland

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Primary fine particulate matter ( $PM_{2.5}$ ) emissions from low-altitude sources, such as traffic and domestic combustion, may cause immediate exposure near the source. In this paper we present emission estimate and uncertainty analysis of  $PM_{2.5}$  emissions from the vehicular traffic and domestic wood combustion sectors. Our estimate of national  $PM_{2.5}$  emissions in 2000 from domestic wood combustion was 7.6 Gg a<sup>-1</sup> and that from vehicular traffic, including non-exhaust emissions, 5.8 Gg a<sup>-1</sup>. These values correspond to 25% and 19% of the national total  $PM_{2.5}$  emissions, respectively. The uncertainties were high for nonexhaust traffic and domestic wood combustion emissions, 37% down, 53% up and 36% down, 50% up of the mean value (95% confidence interval limits), respectively. For traffic exhaust emissions, the uncertainties were lower, 11% down, 13% up. Uncertainties in the domestic combustion factors were the most important individual parameters accounting for total uncertainty.

# Introduction

Fine particulate matter ( $PM_{2.5}$ ) concentrations in ambient air have been shown to be severely detrimental to human health. Average  $PM_{2.5}$ concentrations in Finland are relatively low as compared with those in central and southern Europe. Concentrations in southern Finland are typically 12 and 8  $\mu$ g m<sup>-3</sup> at urban and regional background sites, respectively (Pakkanen *et al.*  2001). Although dominated by long range transported (LRT) secondary particles (Karppinen *et al.* 2005, Ojanen *et al.* 1998), local contributions can be high during peak pollution episodes, such as during extremely stable ground-based temperature inversions (Kukkonen *et al.* 2005).

Some studies have suggested that particles from mobile sources might be relatively more harmful to human health than others (Laden *et al.* 2000), and that primary combustion particles

might be more harmful than secondary particles (Tuomisto *et al.* 2007). There is thus a need for studies focusing on primary PM concentrations at high spatial resolution, especially due to low-altitude emissions (e.g. Forsberg *et al.* 2005, Greco *et al.* 2007).

Such emission sources include vehicular traffic and domestic wood combustion. They are the main sources of  $PM_{2.5}$  in many countries, and both are gaining importance due to increasing traffic volumes, promotion of renewable fuels and lack of established emission control technologies for domestic combustion and traffic-derived non-exhaust particle emissions. Although it is known that there is considerable uncertainty about these emission sources, detailed assessments of the uncertainty are few. The importance of knowledge about emission uncertainties in regional-scale integrated assessment modeling (IAM) has recently been highlighted (e.g. Whyatt *et al.* 2007).

The aim of this study was to systematically estimate PM<sub>25</sub> emissions from vehicular traffic and domestic wood combustion in Finland in 2000 and to quantify the associated uncertainties. The work was carried out within a regional IAM project KOPRA that includes, in addition to emissions at a resolution of  $1 \times 1$  km<sup>2</sup>, the modelling of atmospheric transport, chemistry and aerosol processes, and the modelling of population exposure and health risk at a resolution of  $10 \times$ 10 km<sup>2</sup> throughout Finland (www.environment. fi/syke/pm-modeling). The population exposure effects of vehicular traffic and domestic wood combustion in the vicinity of emission sources at  $1 \times 1$ -km<sup>2</sup> resolution will be assessed in an ongoing project PILTTI (www.ymparisto.fi/default. asp?contentid=202713&lan=fi&clan=en).

## Methodology

#### **Emission calculation**

We used the Finnish Regional Emission Scenario (FRES) model (Karvosenoja and Johansson 2003) to estimate  $PM_{2.5}$  emissions. FRES consists of a coherent bottom-up and top-down calculation of large point sources and area emissions, respectively. The pollutants include pri-

mary particles in several size fractions and precursor gases of secondary PM, and are spatially described at a resolution of  $1 \times 1 \text{ km}^2$  for the entire country (Karvosenoja *et al.* 2005). In use with dispersion models, the FRES annual emissions are temporally disaggregated, into monthly, daily and hourly emission patterns (will be documented later).

In this paper we present  $PM_{2.5}$  emission and uncertainty calculations for the vehicular traffic and domestic wood combustion sectors for annual emissions at the country level. Country level emissions (EM) were calculated from annual activity data (*A*) and emission factors (EF). For each source sector:

$$\mathrm{EM}_{\mathrm{p},t} = A_t \times \mathrm{EF}_{\mathrm{p}} \tag{1}$$

where p = pollutant and t = time.

Traffic activity data (i.e. fuel consumption in vehicles of different ages) in the year 2000 are based on fuel statistics (Statistics Finland 2006) that we disaggregated to vehicle classes of different age using vehicle fleet and use information that are compiled in the Finnish traffic model, LIISA (Mäkelä et al. 2002). Traffic exhaust emission factors were based on several measurement studies that are also used in the international RAINS model as country-specific data, and documented in Klimont et al. (2002). Emission factors represent the emission levels of vehicles of different ages defined by European legislation according to so called EURO standards (e.g., Directives 98/69/EC and 88/77/EC). Non-exhaust emission factors, i.e., tire and brake wear, and suspended dust from roads and the environment are based on a survey of international literature by Karvosenoja et al. (2002) that are comparable with more recent estimates (see e.g. Gehrig 2004). However, the effect of Finnish traction control methods (studded tires and traction sanding) that influence emission factors for road wear and suspension are currently not captured (see later discussion).

Estimates of domestic wood combustion activities (i.e., the amount of wood combustion in different types of combustion appliances in residential and recreational buildings) were based on questionnaire studies (Tuomi 1990, Sevola *et al.* 2003) and expert estimates (S. Tuomi, Finnish Work Efficiency Institute, pers. comm.). Emission factors for domestic wood combustion were based on Finnish (Tissari *et al.* 2007), Nordic (presented in Sternhufvud *et al.* 2004) and international measurements (Butscher and Sorenson 1979, McDonald *et al.* 2000, Environment Australia 2002).

#### **Uncertainty analysis**

Uncertainty distributions were assessed for all 69 input variables (Table 1). The uncertainties were estimated separately for variables affecting

activities and emission factors. The uncertainty of the activity was assumed to have a normal distribution and uncertainty of the emission factor lognormal distribution. The choice of the uncertainty distribution was based on the used data and authors' estimates. A lognormal distribution for emission factor uncertainties was also favored because emission factors are known to be strictly positive, and a large uncertainty together with a normal distribution assumption could result in negative confidence limits values. Domestic wood combustion sub-categories ("iron stoves", "other stoves and ovens", and "open fireplaces") represent similar group of combustion appli-

Table 1. Input parameters and their relative 95% confidence intervals (CI).

Sector	Uncertainty of input parameters		
	Total activity (normal distr.)	Activity division to sub-sectors (normal distr.)	Emission factors (log-normal distr.)
Gasoline consumption (PJ)	±1% <sup>a,1</sup>		
Four-stroke light-duty vehicles		±15% <sup>b</sup>	
4 EURO levels		±5% <sup>b,c</sup>	-20%, +24% <sup>d</sup>
Two-stroke light-duty vehicles		±15% <sup>b</sup>	
2 EURO levels		±5% <sup>b,c</sup>	-20%, +24% <sup>d</sup>
Diesel consumption (PJ)	±1% <sup>a,1</sup>		
Light-duty vehicles		±15% <sup>b</sup>	
4 EURO levels		±5% <sup>b,c</sup>	-20%, +24% <sup>d</sup>
Heavy duty vehicles		±15% <sup>b</sup>	
4 EURO levels		±5% <sup>b,c</sup>	-20%, +24% <sup>d</sup>
Non-exhaust activity (veh-km)			
Light-duty vehicles		Fuel eff.2: ±5%°	
road and tire wear and resuspension/brake wear			–54%, +88% <sup>e</sup>
Heavy duty vehicles		Fuel eff.2: ±5%°	
road and tire wear and resuspension/brake wear			–54%, +88% <sup>e</sup>
Wood combustion in residential buildings (PJ)	±10% <sup>f</sup>		
Primary heated		±15% <sup>g</sup>	
7 combustion appliance types		±25/15% <sup>f,g,3</sup>	–54%, +88% <sup>h</sup>
Supplementary heated		±15% <sup>g</sup>	
3 combustion appliance types		±25% <sup>f,g</sup>	–54%, +88% <sup>h</sup>
Wood combustion in recreational buildings (PJ)	±10% <sup>f</sup>		
3 combustion appliance types		±50% <sup>g</sup>	-54%, +88% <sup>h</sup>

References for uncertainty estimates: <sup>a</sup> Monni *et al.* 2004, <sup>b</sup> K. Mäkelä, VTT Technical Research Centre of Finland, pers. comm., <sup>c</sup> authors' expert judgement, <sup>d</sup> Laschober *et al.* 2004, Lough *et al.* 2005, <sup>e</sup> Ntziachristos *et al.* 2003, Abu-Allaban *et al.* 2003, <sup>f</sup> Sevola *et al.* 2003, <sup>g</sup> authors' and other experts' estimates (S. Tuomi, Finnish Work Efficiency Institute, pers. comm.), <sup>h</sup> Environment Australia 2002, Haakonson and Kvingedal 2001, McDonald *et al.* 2000, Butscher and Sorenson 1979.

<sup>1</sup> Fuels consumed in on-road vehicles in Finland are sold and compiled in statistics separately from fuels in off-road vehicles and machinery because of different fuel taxation.

<sup>2</sup> Average vehicle fuel efficiency for the conversion of fuel consumption into vehicle kilometers.

<sup>3</sup> ±25% uncertainty for the activity division of different appliances, ±15% for manual boilers with/without accumulator.



Fig. 1. Finnish total  $PM_{2.5}$  emissions in 2000. The sectors considered in this study are given in grey shades.

ances in different building and heating type subcategories (primary and secondary heated residential buildings and recreational buildings), and therefore their emission factors were assumed to be correlated.

The uncertainties were propagated through the model using a Monte Carlo simulation with 10 000 iterations. Analytica<sup>TM</sup> 3.1.1. (Lumina Decision Systems, Inc., CA) was used for this purpose. The effect of the uncertainties on FRES model emission results was evaluated using sensitivity analysis. The sensitivity analysis was performed on each individual input variable by calculating absolute rank-order correlations between the input variable and the model results. The model result is the total yearly national primary PM<sub>2.5</sub> emission from the domestic wood combustion and traffic sectors, unless otherwise mentioned.

# Results

#### Total PM<sub>2.5</sub> emissions

 $PM_{2.5}$  emissions from domestic wood combustion and vehicular traffic were respectively 7.6 and 5.8 Gg a<sup>-1</sup> (kilotons per year), which corresponds to 25% and 19%, respectively, of the national total emissions in 2000 (Fig. 1). The highest vehicular traffic emissions originated from the exhaust of light-duty diesel vehicles, contributing 42% of total vehicular traffic emissions. Heavy-duty diesel vehicles accounted for 23% and vehicular non-exhaust emissions for 27% of total vehicular emissions (Tables 2 and 3).

Wood combustion emissions were estimated for different heating and combustion appliance types in residential and recreational buildings. Primary wood heating in residential buildings accounted for 53% of total emissions of the sector. Supplementary wood heating in electricity-heated and oil-heated residential buildings accounted for 29% and wood heating in recreational buildings 17% of total emissions (Table 4).

Of the various domestic combustion appliance sub-categories, the high emissions were caused by "manual feed boilers operated without accumulator tank", mainly because of their high emission factor. Combustion in "other stoves and ovens", mainly comprising of masonry heaters, also had high emissions, but rather because of high levels of activity.

The FRES emissions are compared with the values officially reported to United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CLRTAP) and those of the RAINS model (Table 5). Officially reported emission of domestic wood combustion is based on one emission factor applied to the total sector activity. This emission factor do not include information from recent measurements of different combustion appliances and their substantially different emission characteristics (see e.g. Sternhufvud et al. 2004). A decision has been made to include the mean value estimates of this study in the next reporting to UNECE and the LRTAP Convention. Non-exhaust calculation of the RAINS model excludes resuspension emissions, and therefore it is not comparable with the FRES estimate.

#### Activity uncertainties

The uncertainties of activity values are the result of the uncertainties in the total activities and the divisions into the sub-categories of the vehicles of different ages and different types of combustion appliances (Table 1). For traffic (Tables 2 and 3), the resulting activity uncertainties of the sub-categories were relatively low,  $\pm 1\%$  for four-stroke gasoline and  $\pm 12\%$  for diesel vehicles, comprising of well known total activities from the fuel sale statistics ( $\pm 1\%$ , estimated by Monni *et al.* 2004) and more uncertain subdivisions ( $\pm 5\%$  to  $\pm 15\%$ , K. Mäkelä, VTT Technical Research Centre of Finland, pers. comm. and authors' estimates) that are based on the vehicle fleet age profile and usage data.

For the domestic wood combustion sector (Table 4), Sevola *et al.* (2003) reported  $\pm 10\%$  uncertainty for total wood use in residential and recreational buildings. The uncertainty due to different heating and combustion appliance types

Table 2. The mean values and 95% confidence intervals (CI) of the activities, PM<sub>2.5</sub> emission factors and emissions of vehicular traffic exhaust in 2000 in Finland.

Gasoline vehicles			
	72.2 (71.5–72.9)		357 (308–412)
4-stroke light-duty vehicles <sup>1</sup>	72.0 (71.3–72.7)		324 (276-378)
EURO0	35.3 (34.1–36.4)	6.00 (4.77-7.45)	212 (168-263)
EURO1	11.5 (10.9–12.1)	3.30 (2.62-4.10)	38.0 (30.0-47.3)
EURO2	22.3 (21.3–23.3)	3.30 (2.62-4.10)	73.7 (58.2–91.8)
EURO3	2.88 (2.72-3.05)	0.108 (0.0859-0.134)	0.311 (0.246-0.389)
2-stroke motorcycles & mopeds	0.201 (0.161-0.249)		33.3 (24.4-44.3)
EURO0	0.183 (0.147-0.226)	170 (135–211)	31.1 (22.4-41.7)
EURO1	0.0181 (0.0144–0.0225)	119 (94.6–148)	2.16 (1.56-2.90)
Diesel vehicles	77.0 (76.2–77.8)		3840 (3370–4370)
Light-duty vehicles <sup>1</sup>	32.0 (28.1–36.2)		2480 (1990-3030)
EURO0	16.6 (14.6–18.9)	111 (88.2—138)	1850 (1410–2360)
EURO1	4.48 (3.89-5.11)	72.2 (57.4-89.6)	323 (247-415)
EURO2	9.61 (8.38–10.9)	28.9 (22.9-35.8)	277 (212-356)
EURO3	1.28 (1.11–1.46)	21.1 (16.8–26.2)	27.0 (20.6-34.7)
Heavy-duty vehicles <sup>2</sup>	45.0 (40.8-48.8)		1360 (1160–1600)
EURO0	11.7 (10.5–12.9)	58.0 (46.1-72.0)	678 (528-858)
EURO1	9.00 (8.08-9.89)	37.1 (29.5–46.1)	334 (260-423)
EURO2	21.1 (19.1–23.1)	15.1 (12.0–18.7)	319 (248-401)
	3.15 (2.82-3.48)	10.4 (8.30-13.0)	32.9 (25.5–41.7)
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EURO3 Heavy-duty vehicles <sup>2</sup> EURO0 EURO1 EURO2	1.28 (1.11–1.46) 45.0 (40.8–48.8) 11.7 (10.5–12.9) 9.00 (8.08–9.89) 21.1 (19.1–23.1) 3.15 (2.82–3.48)	21.1 (16.8–26.2) 58.0 (46.1–72.0) 37.1 (29.5–46.1) 15.1 (12.0–18.7) 10.4 (8.30–13.0)	27.0 (20.6–34.7 1360 (1160–160) 678 (528–858) 334 (260–423) 319 (248–401) 32.9 (25.5–41.7

<sup>1</sup> Passenger cars, vans and motorcycles.

<sup>2</sup> Trucks, buses and other heavy duty.

**Table 3**. The mean values and 95% confidence intervals (CI) of the activities, PM<sub>2.5</sub> emission factors and emissions of vehicular traffic non-exhaust in 2000 in Finland.

Sector	Activity (10 <sup>6</sup> veh-km a⁻¹), mean (95% CI)	PM <sub>2.5</sub> emission factor (mg veh-km <sup>-1</sup> ), mean (95% CI)	PM <sub>2.5</sub> emission (Mg a <sup>-1</sup> ), mean (95% CI)
Light-duty vehicles <sup>1</sup> road and tire wear dust and resuspension brake wear dust	43.0 (40.9–45.1)	18.0 (8.46–33.8) 2 80 (1 32–5 26)	894 (472–1590) 774 (365–1450) 120 (56 5–225)
Heavy-duty vehicles <sup>2</sup> road and tire wear dust and resuspension brake wear dust Non-exhaust total	3.40 (3.23–3.57) 46.4 (44.2–48.5)	180 (84.6–338) 17.3 (8.13–32.5)	671 (341–1210) 612 (286–1150) 58.8 (27.6–110) 1570 (977–2390)

<sup>1</sup> Passenger cars, vans and motorcycles.

<sup>2</sup> Trucks, buses and other heavy duty.

in residential buildings, which was based on information from questionnaire studies (Tuomi 1990, Sevola *et al.* 2003), was estimated to be between  $\pm 15\%$  and  $\pm 25\%$  (S. Tuomi, Finnish Work Efficiency Institute, pers. comm. and authors' estimates). For recreational buildings, however, there were no literature information available and uncertainty associated with appliance types was estimated to be  $\pm 50\%$ . The resulting relative uncertainties associated with the activities of domestic combustion sub-categories were mainly between  $\pm 25\%$  and  $\pm 40\%$ .

#### **Emission factor uncertainties**

The uncertainties in emission factors were higher than for activities for nearly all categories. For domestic wood combustion emissions, the estimated uncertainties were particularly high; the lower limit of 95% CI was the mean value minus 54% and the upper limit the mean value plus 88% (54% down, 88% up). These values were estimated primarily based on several sets of measurement data on stoves (Butscher and Sorenson 1979, McDonald *et al.* 2000, Haakon-

Table 4. The mean values and 95% confidence intervals (CI) of the activities, PM<sub>2.5</sub> emission factors and emissions of domestic wood combustion in 2000 in Finland.

Sector	Activity (PJ a <sup>-1</sup> ), mean (95% CI)	PM <sub>2.5</sub> emission factor (mg MJ <sup>-1</sup> ), mean (95% CI)	PM <sub>2.5</sub> emission (Mg a <sup>-1</sup> ), mean (95% CI)
Residential buildings (RsB)	34.2 (30.8–37.6)		6270 (3900–9630)
Primary wood-heated RsB	20.2 (16.6–23.9)		4040 (2470–6350)
manual feed boilers with accumulator tank	5.42 (3.89-7.22)	80.0 (37.6–150)	434 (187–852)
manual feed boilers without accumulator tank	2.67 (1.67–3.87)	700 (329–1310)	1870 (762–3840)
automatic feed wood chip boilers	1.46 (1.01–2)	50.0 (23.5–93.9)	73.0 (31.4–148)
automatic feed pellet boilers	0.102 (0.0693-0.142)	30.0 (14.1–56.3)	3.05 (1.29-6.11)
iron stoves	0.142 (0.0976-0.196)	700 (329–1310)	99.5 (42.1–199)
other stoves and ovens <sup>1</sup>	10.2 (7.86–12.8)	140 (65.8–263)	1430 (634–2780)
open fireplaces	0.163 (0.111–0.224)	800 (376–1500)	130 (54.9–259)
Supplementary wood-heated RsB	14.0 (10.7–17.4)		2230 (1130–4080)
iron stoves	0.212 (0.135–0.316)	700 (329–1310)	148 (59.7–303)
other stoves and ovens <sup>1</sup>	13.6 (10.4–16.9)	140 (65.8–263)	1900 (849–3720)
open fireplaces	0.222 (0.14-0.332)	800 (376–1500)	178 (70.7–370)
Recreational buildings	5.00 (4.50-5.50)		1310 (758–2130)
iron stoves	0.780 (0.372-1.37)	700 (329–1310)	545 (186–1250)
other stoves and ovens <sup>1</sup>	3.96 (3.19-4.59)	140 (65.8–263)	554 (249–1060)
open fireplaces	0.262 (0.118-0.477)	800 (376–1500)	209 (67.6-488)
Total domestic wood combustion	39.2 (35.7–42.6)		7580 (4870–11400)

<sup>1</sup> Incl. masonry heaters, masonry ovens, kitchen ranges and sauna stoves.

**Table 5.** PM<sub>2.5</sub> emissions from vehicular traffic and domestic wood combustion in 2000 in Finland based on the FRES model, official inventory reported to UNECE LRTAP Convention and the RAINS model.

Sector	FRES model	Officially reported inventory	RAINS model
Traffic exhaust	4.2	4.1	4.2/3.71
Traffic non-exhaust	1.6	1.6	0.44 <sup>2</sup>
Domestic wood combustion	7.6	15	7.6

<sup>1</sup> Emissions based on different RAINS model versions' emission factors: from AutoOil programme documented in Klimont et al. (2002) / from June 2007 set of scenarios prepared for the EU within the National Emission Ceilings (NEC) Directive review (www.iiasa.ac.at/gains).

<sup>2</sup> Do not include resuspension.



**Fig. 2**. The sensitivity analysis of the input variables (rank correlation between each input variable and the output variable). Only 12 most important variables are shown. The bars referring to the parameters of domestic wood combustion and traffic are given grey and white, respectively. EF = emission factor, RsB = residential buildings.

son and Kvingedal 2001, Environment Australia 2002), and are comparable with the emission factor uncertainties presented by Bond *et al.* (2004) and Finnish expert consultations (J. Tissari, University of Kuopio, pers. comm.).

For non-exhaust vehicular traffic emissions, the estimated uncertainties were equally as high as those for domestic wood combustion, 54% down, 88% up. They are based on emission factor measurements and uncertainty reviews by Ntziachristos (2003) and Abu-Allaban *et al.* (2003).

For traffic exhaust of different vehicle age groups representing different EURO emission levels, the uncertainties were estimated considerably lower, being 20% down and 24% up. Tunnel measurement data sets (Laschober *et al.* 2004, Lough *et al.* 2005) with predominately light-duty vehicle fleet suggested that exhaust emission factor uncertainties are mainly around  $\pm 20\%$ .

#### Total emission uncertainties

The resulting emission uncertainties for domestic wood combustion were 2.7 Gg  $a^{-1}$  (lower 95% CI limit) and 3.8 Gg  $a^{-1}$  (upper 95% CI limit), corresponding to 36% down and 50% up of the total mean emission of the sector. For individual appliance type categories, the uncertainties were mainly around 58% down, 100% up. The highest uncertainties in relative terms were for "open fireplaces" and "iron stoves" in recreational buildings (approx. 67% down and 130% up), and in absolute terms for "manual feed boilers without accumulator tank" (1.1 Gg a<sup>-1</sup> down and 2.0 Gg a<sup>-1</sup> up). For traffic, uncertainties for exhaust emissions were 0.48 Gg a<sup>-1</sup> down, 0.53 Gg a<sup>-1</sup> up, or 11% down, 13% up of the mean emission value, and for non-exhaust 0.59 Gg a<sup>-1</sup> down, 0.83 Gg a<sup>-1</sup> up, or 37% down, 53% up.

#### Sensitivity analysis

The sensitivity analysis was done by calculating absolute rank-order correlations between all the input variables of the model and the model result (total annual PM<sub>2.5</sub> emissions from traffic and domestic combustion). The five highest ranked parameters were all associated with the domestic wood combustion sector. Correlations were the highest for the emission factor uncertainties of the domestic wood combustion categories with the highest emissions, i.e. "other stoves and ovens" and "manual boilers without accumulators", with correlations 0.77 and 0.39, respectively (Fig. 2). The high sensitivity of domestic wood combustion input variables compared to the traffic sector reflects both high emission volumes and high uncertainties in the input parameters.

For traffic, the highest sensitivities (correlations) were for non-exhaust emission factors (0.14 for both light and heavy duty vehicles). For traffic exhaust emissions, the highest correlation, 0.12, was for the emission factor of pre-1992 (EURO 0) light duty diesel vehicles.

# **Discussion and conclusions**

#### **Emission uncertainties**

Uncertainties in the PM25 emissions from the domestic wood combustion sector were considerable, with the most influential parameters related to emission factors and total activities. These parameters can be expected to be uncertain for a number of reasons. Firstly, domestic wood combustion appliances are mainly relatively simple and poorly controllable, and emissions are strongly dependent on individual combustion practises. It would be possible to diminish the uncertainties in the emission factors of different combustion appliances by conducting more detailed measurements. Secondly, since the majority of wood fuel in Finland is taken by the consumers from their own or relatives' forests. activity estimates can not be based on sale statistics, but have to rely on questionnaires instead. Existing questionnaires of total wood use are relatively comprehensive with uncertainties of ±10% (Sevola et al. 2003), however, the parameter of the total activity in residential buildings was found relatively sensitive (third highest rank-order correlation = 0.17). While combustion appliance use in residential buildings has been investigated (Tuomi 1990), there is a clear lack of knowledge about combustion appliance use in recreational buildings. The uncertainties of these appliance use parameters  $(\pm 50\%)$  have, however, relatively low sensitivity (correlations below 1.0).

Vehicular traffic exhaust emissions are less uncertain than domestic combustion emissions. In Finland, given the relatively small population and developed statistics on fuel sale and vehicle fleet, it is possible to make a relatively disaggregated calculation procedure in terms of vehicle classes with different emission characteristics.

The uncertainty is considerable in nonexhaust emissions; these are both temporally and spatially highly variable, and depend on various ambient conditions. If traffic volumes continue to increase, the importance of non-exhaust emissions will increase in the future. There is thus a clear need for a better understanding of nonexhaust emissions and means to abate emissions, especially of the fine size fractions.

### Features of non-exhaust traffic emissions specific to Finland

The non-exhaust emission factors that have been used in this study are probably underestimates as they are based on emissions from paved roads averaged from several countries. Emission factors for Finland are probably higher because of the increased suspension of particulates caused by street sanding and widespread use of studded tires in winter that causes road abrasion. Recent road dust measurements in Finland and Scandinavia (Gustafsson et al. 2005, Kupiainen et al. 2005a, 2005b, Omstedt et al. 2005) suggest that the re-suspension levels for  $PM_{10}$  and  $PM_{25}$ can be up to five to six times higher in the winter months (mid-January to mid-April) than during rest of the year. The effect of these features are not included in this study, but will be incorporated into future FRES modeling. Furthermore, the uncertainty analysis of this study on nonexhaust emissions does not attempt to cover the effect of the specific Finnish circumstances. Instead, it represents a general uncertainty of non-exhaust emissions.

# Heterogeneity in domestic wood combustion emissions

Our results show that domestic wood combustion emissions are heterogeneous and depend on the emission characteristics of stove and boiler types and usage in different countries. A comparative study for the Nordic countries showed that national average domestic wood combustion emission factors ranged from 200 mg MJ<sup>-1</sup> of PM<sub>2.5</sub> for Finland to 1800 mg MJ<sup>-1</sup> for Norway (Sternhufvud *et al.* 2004). In Finland, the most typical stove type is the masonry heater, which consists of a stone mass that stores the heat leading to relatively unified temperature profile on the inside surfaces of the stove and thus relatively low emissions. Conventional enclosed non-masonry stoves (referred as "iron stoves" in this study), which are common in many European countries and in the U.S., typically have higher emission factors. Other factors than stove type that strongly influence domestic wood combustion emissions include fuel quality and the combustion practices of the users. Therefore the national features of domestic wood combustion in emission inventories, e.g. substantially different emission characteristics of different combustion appliances, is important and should be included in international reporting processes.

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