

THE SPATIAL ALLOCATION OF AIR POLLUTANTS IN FINNISH REGIONAL EMISSION MODEL

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ABSTRACT

Human health effects caused by fine particulate matter (PM) have raised a need for estimation tools, such as integrated assessment models (IAMs). The Finnish Regional Emission Scenario (FRES) model has been developed as part of the Finnish IAM of air pollution. FRES calculates the annual emissions of primary PM in different sizes and the main precursor gases of secondary PM from 1990 to 2020. Recently the model has been extended to include chemical speciation, i.e., climate and health relevant, black and organic carbon as well as sulphates. Emissions are calculated as point and area sources. Area sources are aggregated to 8 main sectors and more than 100 subsectors. The spatial resolution of area emissions has recently been refined to $1 \times 1 \text{ km}^2$ grid. This paper presents the spatial allocation of area emissions in FRES. The emission estimates of FRES were convergent compared to other emission inventories. The spatial allocation procedure in two steps: (i) first from country to municipality level in more sectoral detail, and (ii) second from municipality to $1 \times 1 \text{ km}^2$ level, was found useful. Municipality level weighting surrogates were available for the most important emission sources at adequate aggregation level. At $1 \times 1 \text{ km}^2$ level weighting surrogates were available for the two most important area emission sources, road traffic and domestic wood combustion.

Key Words: Emission, Area Sources, Fine Particles, GIS

1. INTRODUCTION

The health risk of fine particulate matter (PM) can be assessed with integrated assessment models (IAMs), which include spatially allocated emission estimates, atmospheric modeling and health risk modeling. Earlier Finnish regional IAM system has been used to assess acidification (Syri et al. 2002, Johansson et al. 2001). PM assessment has so far included emission scenario (Karvosenoja and Johansson 2003a, Karvosenoja et al. 2003) and qualitative health implication estimates (Johansson et al. 2003). The extension of IAM for PM assessment to include atmospheric dispersion and health risk modeling is in development in a KOPRA project (www.fmi.fi/research_air/air_47.html).

The Finnish Regional Emission Scenario (FRES) model has been used as an emission estimation tool in the Finnish IAM system. In acidification assessment relatively coarse spatial resolution of area emissions at municipality level has been found adequate. In order to better meet the requirements of PM health risk assessment, the spatial description of area source emissions in FRES has recently been refined to $1 \times 1 \text{ km}^2$ grid. There are two main arguments for the need of high resolution emission estimates. First, atmospheric models, into which FRES emission data are fed in the IAM system, requires more and more high resolution emission data processing capacity increases. The regional atmospheric model system that is used with FRES in the KOPRA project is SILAM (Sofiev & Siljamo, 2003) of Finnish Meteorological Institute with $30 \times 30 \text{ km}^2$ grid size for Europe and $5 \times 5 \text{ km}^2$ for Finland. Second, high spatial resolution allows the assessment of the relative importance of different low-height emission sources on PM concentrations in the vicinity of the sources.

This paper presents the spatial allocation of area emissions in FRES, including recent refinement to $1 \times 1 \text{ km}^2$ grid. In addition, sectoral country-total emissions of fine primary particles (PM2.5), sulphur dioxide (SO₂), nitrogen oxides (NO_x), ammonia (NH₃) and non-methane volatile organic compounds (NMVOCs), as well as black and organic carbon fractions in fine particles (BC and OC) are presented. The respective emissions at $1 \times 1 \text{ km}^2$ grid are graphically presented as maps.

2. METHODOLOGY

The Finnish Regional Emission Scenario (FRES) model works as a part of the integrated assessment model (IAM) system of PM. FRES consists of coherent emission calculation from all anthropogenic sources with spatial allocation of emissions. The pollutants include primary total suspended particles (TSP) and finer size fractions (PM10, PM2.5 and PM1), and the main precursor gases of secondary PM, *i.e.* sulphur dioxide (SO₂), nitrogen oxides (NO_x), ammonia (NH3) and nonmethane volatile organic compounds (NMVOCs). Primary particle estimate includes partition to different chemical compounds: black carbon (BC), organic carbon (OC) and sulphates. Large energy production and industrial plants are described as point sources (*i.e.* the plants utilizing boilers with thermal capacity exceeding 50 MW_{th}, and the plants with TSP, SO₂ or NO_x emissions higher than 20 Mg/a, respectively). Other sources are described as area sources. A more detailed model description can be found from Karvosenoja and Johansson (2003b). The spatial allocation of area emissions at two steps (municipality and 1 × 1km² level) is described in detail in the following.

Area emissions are described at three different spatial level: country, municipality and $1 \times 1 \text{km}^2$ level. Emission calculation takes place at country level. Area emissions EM^{cou} are calculated from country level annual activity data A^{cou}, unabated emission factors EF and removal efficiencies η of various emission control technologies which can be applied to each source sector (*i.e.* sector-fuel type combination) with certain utility rates X. The country level emission EM^a from an area source sector is:

$$EM^{cou}_{i,j,k,l}(t) = \sum_{l} A^{cou}_{j,k}(t) \cdot EF_{i,j,k} \cdot X_{i,j,k,l}(t) \cdot (1 - \eta_{i,j,k,l})$$
(1)

where t = time, i = pollutant, j = fuel, k = sector and l = control technology. The base year 2000 activity data is mainly based on national statistics (Statistics Finland 2003). The emission factors, control technology removal efficiencies and utilization rates are based on literature and other sources reported in Karvosenoja (2001), Karvosenoja and Johansson (2003b) and Karvosenoja *et al.* (2002). The emission factors for chemical species, black carbon, organic carbon, and sulphates, are calculated from source specific chemical emission profiles (as percentages in $PM_{2.5}$) that have been estimated based on national and international literature.

Country level area emissions are allocated to 448 municipalities using the respective relative fractions RF^{mun} of municipality level allocation data B in base year 2000 in 25 emission source sectors (see Table 1). Area source emission EM^{mun} in a municipality m is:

$$\mathrm{EM}^{\mathrm{mun}}_{i,j,k,l,m}(t) = \mathrm{EM}^{\mathrm{cou}}_{i,j,k,l}(t) \cdot \mathrm{RF}^{\mathrm{mun}}_{j,k,m}$$
⁽²⁾

where

$$RF^{mun}_{j,k,m} = \frac{B^{mun}_{j,k,m}}{B^{cou}_{j,k}}$$
(3)

Municipality level emissions are further allocated to $1 \times 1 \text{km}^2$ grid at four main emission sectors: road traffic, domestic wood combustion, agriculture and other sources. Because of high national interest in studying the importance of domestic wood combustion in health effects, its allocation takes place in 6 subsectors (see Table 1). Emission EM^{1×1} in an emission grid cell n is:

$$EM^{1x_{i,j,k,l,m,n}}(t) = EM^{mun}_{i,j,k,l,m}(t) \cdot RF^{1x_{l,k,m,n}}$$
(4)

where

$$RF^{1x1}_{j,k,m,n} = \frac{C^{1x1}_{j,k,m,n}}{C^{mun}_{j,k,m}}$$
(5)

where C is geographical allocation information at $1 \times 1 \text{km}^2$ level. Municipality and $1 \times 1 \text{km}^2$ level sectoral division and allocation data used in the determination of the relative fractions are presented in Table 1. For road traffic at municipality level fuel consumption values were based on the Finnish road traffic emission calculation system LIISA (Mäkelä *et al.* 2002). For the municipality allocation of animal-related PM and ammonia emissions from agriculture, municipality level animal numbers weighted with country-level PM and ammonia emissions, respectively, of different

animal types were used as weighting factors. The animal number data were taken from agricultural databases of Finnish Environment Institute (Grönroos et al. 1998). Agricultural field area information from national SLICES land use element (Mikkola et al. 1999) was used at municipality level as a basis for agricultural machinery and field-origin PM emissions. In addition, SLICES data at $1 \times 1 \text{km}^2$ level was used for all agricultural non-combustion emissions. SLICES data on road surface area was used in road traffic emission allocation to $1 \times 1 \text{km}^2$ level. SLICES is a combination of different national land use GIS databases, of which output is raster databases in 10×10 and $25 \times 25 \text{ m}^2$. For example, relative fraction RF^{1×1} for road traffic emission is road surface area in a grid cell divided by road surface area in respective municipality.

As a basis for the municipality allocation of domestic combustion of different fuels, degree-day weighted gross-floor areas of different types of buildings from national building and dwelling register were used. Building and dwelling register is a part of SLICES land use element, and includes all Finnish buildings with information on e.g. gross floor area, heating types and fuels, building date and resident. Degree-day weighting was assumed to represent the differences in room heating needs in different parts of the country. For the allocation of domestic peat and coal combustion, however, population was used as allocation basis instead of heating information. Domestic peat and coal combustion are rare in Finland, and building and dwelling register was noticed not to include adequate information on peat and coal heating. Population data used in this study was also based on building and dwelling register. The building and dwelling register gross-floor area data were used also in $1 \times 1 \text{km}^2$ level allocation of domestic wood combustion. Allocation to $1 \times 1 \text{km}^2$ 1km² level was not done with building and dwelling register on domestic oil combustion, which contributes to less than 5% of the total PM2.5 emissions of domestic combustion, and therefore was not considered relevant in terms of potential health risks.

Other data used in municipality emission allocation included managed forest area (8th nation-wide forest inventory data base), annual fuel combustion in energy production boilers below 50 MW_{th} (Korkia-Aho et al. 1995) and annual milled peat production values (VAPO Group data base).

Table 1. Spatial area source emission allocation in the FRES model

Main sectors at country	Sectors at municipality level;	Sectors at 1×1 km ² level;
level;	Data used as a basis for emission allocation	GIS data used as a basis for
Number of subsectors and		emission allocation from
tuels		municipality to 1x1km2 level
Road traffic;	Light-duty gasoline exhaust;	Road traffic;
TO Subsectors, 3 fuels	Annual LD gasoline consumption	Road surface area
	Annual I D diesel consumption ¹	
	Heavy-duty diesel exhaust	-
	Annual HD diesel consumption ¹	
	Light-duty dust emissions:	-
	Annual LD fuel consumption ¹	
	Heavy-duty dust emissions;	-
	Annual HD diesel consumption ¹	
Agricultural non-combustion	PM from animal husbandry;	Agriculture;
activities (primary PM and	Animal numbers weighted with PM emissions of	Agricultural field area ³
NH ₃);	animals ²	
6 subsectors	NH3 from animal husbandry;	
	Animal numbers weighted with NH ₃ emissions of	
	animals ²	_
	PM from field preparation and narvesting;	
Decidential and other small	Agricultural field area	Mood comp in wood bootod
Residential and other small-	(RsR).	ReB.
23 subsectors 6 fuels	(NSD), Degree-day weighted gross-floor area (DWGA) of	Gross-floor area (GA) of
	wood boiler -beated RsB ⁴	wood-beated RsR ⁴
	Wood comb in stove-heated RsB	
	DWGA of wood stove -heated RsB ⁴	
	Wood comb, in RsB heated primarily with other than	Wood comb. in RsB heated
	wood;	primarily with other than
	DWGA of detached RsB built after 1979 and heated	wood;
	primarily with other than wood ⁴	GA of detached RsB built
		after 1979 and heated
		primarily with other than
		wood*
	Wood comb. in recreational buildings (RcB);	Wood comb. in RcB;
	DWGA of all RCB	GA OF All RCB
	$DWGA$ of wood boated AB^4	CA of wood boated AB^4
	Wood comp in public buildings (PR):	Wood comb in PR:
	DWGA of wood-beated PB4	GA of wood-beated PB ⁴
	Wood comb, in industrial buildings (IB)	Wood comb. in IB
	DWGA of wood-heated IB ⁴	GA of wood-heated IB ⁴
	Oil combustion;	Other sources;
	DWGA of all oil-heated buildings ⁴	Population ⁴
	Peat and coal combustion;	
	Population ⁴	
Off-road traffic and	Forestry machinery;	
machinery;	Managed forest area [°]	_
14 subsectors, 4 fuels	Agricultural machinery;	
	Agricultural field area	
	Other off-road and machinery (incl. domestic	
	navigation and air traffic);	
Dower plants and industrial	Population Rewar planta and industrial comb :	4
Fower plants and industrial comb in boilers $<50 \text{ MW}_{\odot}$	Appual fuel consumption in boilers below 50 MW_{*}^{6}	
8 subsectors 10 fuels	Annual ruer consumption in boliers below 50 MWth	
Industrial non-comb		4
industrial non comb.	Industrial processes:	
processes, em. <20 Mg a ⁻¹	Industrial processes; Population ⁴	
processes, em. <20 Mg a ⁻¹ ; 14 subsectors	Industrial processes; Population ⁴	
processes, em. <20 Mg a ⁻¹ ; 14 subsectors Other primary PM related	Industrial processes; Population ⁴ Milled peat production;	_
processes, em. <20 Mg a ⁻¹ ; 14 subsectors Other primary PM related non-combustion sources;	Industrial processes; Population ⁴ Milled peat production; Annual milled peat production ⁷	-
processes, em. <20 Mg a ⁻¹ ; 14 subsectors Other primary PM related non-combustion sources; 19 subsectors	Industrial processes; Population ⁴ Milled peat production; Annual milled peat production ⁷ Other sources;	-
processes, em. <20 Mg a ⁻¹ ; 14 subsectors Other primary PM related non-combustion sources; 19 subsectors	Industrial processes; Population ⁴ Milled peat production; Annual milled peat production ⁷ Other sources; Population ⁴	
processes, em. <20 Mg a ⁻¹ ; 14 subsectors Other primary PM related non-combustion sources; 19 subsectors Other NMVOC related non-	Industrial processes; Population ⁴ Milled peat production; Annual milled peat production ⁷ Other sources; Population ⁴ Other NMVOC related sources;	-

1) Mäkelä et al. 2002, 2) Grönroos et al. 1998, 3) SLICES (Mikkola et al. 1999), 4) building and dwelling register (Mikkola et al. 1999), 5) 8th nation-wide forest inventory data base, 6) Korkia-Aho et al. 1995, 7) VAPO data base

3. RESULTS AND DISCUSSION

3.1 Country level

Emissions in main emission sources in the base year 2000 are presented in Table 2. The major contributors to fine primary PM emissions were small-scale wood combustion and traffic sources. These sources were predominant also in particulate black (BC) and organic carbon (OC) emissions. Stationary industrial activities, *i.e.* power plants, and combustion and processes in industry, were the main contributors to SO₂ emissions. Traffic sources caused the biggest NO_x and NMVOC emissions. Ammonia (NH₃) originated mainly from agriculture. Area sources contributed to 77, 37, 71, 97, 96, 98 and 98% of PM2.5, SO₂, NO_x, NH3, NMVOCs, BC and OC emissions, respectively, of the total emissions.

The results of the FRES model were convergent compared to other inventories. The biggest difference is in PM2.5, where FRES estimate for residential wood combustion is 8.2 Gg a⁻¹ lower than in other inventories. The PM2.5 emission factors of residential wood combustion have been lately revised in the light of recent Nordic (Sternhufvud et al. 2004) and Finnish (Raunemaa et al. in press) measurements. This review is reflected to FRES results, but not yet to other inventories. Residential wood combustion emissions are discussed in more detail in Karvosenoja et al. (this issue).

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Main area sources at country level	PM2.5	SO2	NOx	NH3	NMVOC	BC in PM2.5	OC in PM2.5
Road traffic	5.5	0.2	75		42	1.9	1.3
Agricultural non-combustion	0.4			32			
activities (primary PM and NH ₃)						0.0	0.2
Residential and other small-	8.0	3.2	8.0		28		
scale combustion						1.4	3.9
Off-road traffic and machinery	3.8	3.8	43		27	1.3	1.1
Power plants and industrial	2.4	17	14		1.9		
comb. in boilers below 50 MW _{th}						0.6	0.3
Industrial non-comb. processes	0.0	4.5	4.8		15		
with emissions below 20 Mg a ⁻¹						0.0	0.0
Other primary PM related non-	3.1						
combustion sources						0.0	1.6
Other NMVOC related non-					37		
combustion sources							
TOTAL area sources	23	29	145	32	151	5.3	8.3
Point sources	6.8	49	58	1.0	7.1	0.1	0.2
TOTAL	30	78	203	33	158	5.5	8.5
TOTAL in national inventories	38 ^a	74 ^a /76 ^b	210 ^a /206 ^b	33ª	160 ^a	-	-

Table 2. FRES emissions in 2000 and comparison to national inventories (Gg a^{-1})

a) Finnish Environment Institute 2005, b) Statistics Finland 2004

3.21×1 km² level

The total Finnish emissions of PM2.5, SO₂, NO_x, NMVOC, BC and OC spatially allocated to $1 \times 1 \text{km}^2$ are presented in $10 \times 10 \text{km}^2$ grid (Figure 1). Primary PM emissions, including BC and OC, are relatively evenly distributed to the whole southern and Central Finland, while NOx and NMVOC emissions are more weighted to population centers in southern and south-western Finland. Sulphur emissions take place mainly in large point sources, such as power plants and industrial processes.

The total number of grid cells in the Finnish land, lake and coastal sea area is 365 980, of which 102 393 are inhabited. Spatially allocated sectoral emissions are presented in 10×10 km² maps for the whole Finland and 1×1 km² for south-western Finland. The emissions of road traffic occur in population centers in south-western Finland (Figure 2a). Residential wood combustion emissions are relatively evenly distributed to the southern and central Finland (Figure 2b). Agricultural emissions take place in rural areas mainly in western Finland. (Figure 2c). Source sector other area emissions are presented in Figure 2d and point source emissions in Figure 2e.



Figure 1. Total emissions of (a) PM2.5, (b) SO₂, (c) NO_x, (d) NMVOC, (e) BC and (f) OC in 2000 presented in $10 \times 10 \text{km}^2$. Unit Mg a⁻¹.



Figure 2. The sectoral emissions of (a) road traffic, (b) residential wood combustion, (c) agriculture, (d) other area sources and (e) point sources in 2000 presented in $10 \times 10 \text{km}^2$ and (a) – (d) in $1 \times 1 \text{km}^2$. Unit Mg(PM2.5) a⁻¹, except (c) Mg(NH₃) a⁻¹ and (e) Mg(SO₂) a⁻¹.

4. CONCLUSIONS

This paper presents the emission calculation of Finnish Regional Emission Scenario (FRES) model a which combines a top-down approach of aggregated emission source sector description with more detailed bottom-up calculation of large point sources. The spatial allocation of area emissions at two steps to municipality and $1 \times 1 \text{km}^2$ level is presented.

The country total emissions of fine primary particles (PM2.5), sulphur dioxide (SO₂), nitrogen oxides (NO_x), ammonia (NH₃), non-methane volatile organic compounds (NMVOCs), particulate black carbon (BC) and organic carbon (OC) were presented and compared to official national emission inventories. The results were convergent, except for residential wood combustion where the latest emission factor estimates have not yet been implemented in official emission inventories.

The top-down spatial allocation from country level was carried out successfully using municipality and $1 \times 1 \text{km}^2$ level weighting surrogates. Municipality level weighting surrogates were available for the most important emission sources at adequate aggregation level. At $1 \times 1 \text{ km}^2$ level weighting surrogates were available for the two most important area emission sources, road traffic and domestic wood combustion.

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