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Emission scenario model for regional air pollution

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Emission scenario model for regional air pollution

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Contents

List of original publications and author's contribution

I Syri S., Karvosenoja N., Lehtilä A., Laurila T., Lindfors V. and Tuovinen J.-P. 2002. Modeling the impacts of the Finnish climate strategy on air pollution. *Atmospheric Environ.* 36: 3059-3069.

The author was responsible for the management and calculation of air pollution emission data. The Results and Discussion and conclusions sections for the part of emissions were carried out jointly with the corresponding author.

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IV Karvosenoja N., Klimont Z., Tohka A., Johansson M. 2007. Fine primary particle emission reduction potential and cost efficiency in Finland. *Environ. Res. Lett.* 2 (2007) 044002.

The author was responsible for manuscript coordination, the calculation of emissions, reduction potentials and costs (except for the traffic sector) and the presentation of the results. The discussion and conclusions, as well as the assessment of emission reduction potentials in industrial process sector, were done jointly.

V Karvosenoja N., Tainio M., Kupiainen K., Tuomisto J. T., Kukkonen J. and Johansson M. 2008. Evaluation of the emissions and uncertainties of $PM_{2.5}$ originated from vehicular traffic and domestic wood combustion in Finland. *Boreal Environ. Res.* 13:465-474.

The author was responsible for the manuscript coordination and the calculation and presentation of emissions. The input parameter uncertainty values were estimated and the calculated uncertainties presented jointly.

VI Rypdal K., Rive N., Åström S., Karvosenoja N., Kupiainen K., Bak J. and Aunan K. 2007. Nordic Air Quality Co-Benefits from European Post-2012 Climate Polices. *Energy Policy*, 35: 6309-6322.

The author was responsible for the coordination of the Finnish PM exposure case study, the conversion of RAINS scenarios into FRES model, the calculation and presentation of Finnish $PM_{2.5}$ emissions and exposures (except for the application of PM source-receptor matrices). The discussion and conclusions of the exposure case were performed jointly.

Symbols and abbreviations

Symbols

NH_3	ammonia
NH_4^+	ammonium
NMVOCs	non-methane volatile organic compounds
NO_x	nitrogen oxides
NO_3^-	nitrates
O_3	ozone
PM	particulate matter
PM_{10}	particles with diameter below 10 μm
$PM_{2.5}$	particles with diameter below 2.5 μm
PM_1	particles with diameter below 1 μm
$PM_{0.1}$	particles with diameter below 0.1 μm
SO_2	sulfur dioxide
SO_4^{2-}	sulfates

Abbreviations

ASAM	Abatement Strategy Assessment Model of the Imperial College
BAU	Business-As-Usual activity pathway
BAT	Best Available Technologies defined in IPPC
CAFE	Clean Air for Europe programme of EU
CASM	Coordinated Abatement Strategy Model of the Stockholm Environment Institute
CEPMEIP	Co-ordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance
CLE	Current Legislation emission reduction scenario
CLRTAP	Convention on Long-Range Transboundary Air Pollution
EMEP	Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air pollutants in Europe of CLRTAP
ESP	electrostatic precipitator
EU	European Union
FGD	flue gas desulfurization
FRES	Finnish Regional Emission Scenario model
GHG	greenhouse gases
IAM	integrated assessment model(ing)
IIASA	International Institute for Applied Systems Analysis
IPPC	Integrated Pollution Prevention and Control directive of EU
K-Nu	Kyoto-Nuclear activity pathway
LCP	Large Combustion Plants directive of EU
LRT	long-range transport
MERLIN	Multi-pollutant, Multi-Effect Assessment of European Air Pollution Control Strategies: an Integrated Approach of the University of Stuttgart
NEC	National Emissions Ceilings directive of EU
RAINS	Regional Air Pollution INformation and Simulation model of IIASA
SCR	selective catalytic reduction of NO_x
SNAP	Selected Nomenclature for Air Pollution
SNCR	selective non-catalytic reduction of NO_x
UNECE	United Nations Economic Commission for Europe
WAM	With-Additional-Measures activity pathway

Emission scenario model for regional air pollution

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Air pollution emissions are produced in a wide variety of sources. They often result in detrimental impacts on both environments and human populations. To assess the emissions and impacts of air pollution, mathematical models have been developed. This study presents results from the application of an air pollution emission model, the Finnish Regional Emission Scenario (FRES) model, that covers the emissions of sulfur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3), non-methane volatile organic compounds (NMVOCs) and primary particulate matter (TSP, PM_{10}, $PM_{2.5}$ and PM_1) in high 1×1 km^2 spatial resolution over the area of Finland. The aims of the study were to identify key emission sources in Finland at present and in the future, to assess the effects of climate policies on air pollution, and to estimate emission reduction potentials and costs. Uncertainties in emission estimates were analyzed. Finally, emission model characteristics for use in different air pollution impact applications were discussed.

The main emission sources in Finland are large industrial and energy production plants for SO_2 (64% of 76 Gg a^{-1} total in the year 2000). Traffic vehicles are the main contributors for NO_x (58% of 206 Gg a^{-1}), NMVOCs (54% of 152 Gg a^{-1}) and primary $PM_{2.5}$ (26% of 31 Gg a^{-1}) emissions. Agriculture is the key source for NH_3 (97% of 33 Gg a^{-1}). Other important sources are domestic wood combustion for primary $PM_{2.5}$ (25%) and NMVOCs (12%), and fugitive dust emissions from traffic and other activities for primary PM_{10} (30% of 46 Gg a^{-1}).

In the future, the emissions of traffic vehicle exhaust will decrease considerably, by 76% (NMVOCs), 74% (primary $PM_{2.5}$) and 60% (NO_x), from 2000 to 2020, because of tightening emission legislations. Rather smaller decrease is anticipated in the emissions of large combustion plants, depending on future primary energy choices. Sources that are not subject to tight emission standards, e.g. domestic combustion and traffic-induced fugitive dust (i.e. non-exhaust), pose a risk for increasing emissions.

The majority of measures to abate climate change, e.g. energy saving and non-combustion based energy production, lead to co-benefits as reduced air pollution emissions, especially of SO_2 (20% to 28% reduction). However, promotion of domestic wood combustion poses a risk for increase in $PM_{2.5}$ and NMVOCs emissions. Further emission reductions with feasible control costs are possible mainly for $PM_{2.5}$ in small energy production plants and domestic combustion sources. Highest emission uncertainties were estimated for primary PM emission factors of domestic wood combustion, traffic non-exhaust sources and small energy production plants.

The most important characteristics of emission models are correct location information of flue gas stacks of large plants for the assessment of acidification, and description of small polluters with high spatial resolution when assessing impacts on populations. Especially primary $PM_{2.5}$ emissions originate to a considerable degree from small low-altitude sources in urban areas, and therefore it is important to be able to assess the impacts that take place near the emission sources. Detailed descriptions of large plants and 1×1 km^2 spatial resolution for small emission sources applied in the FRES model enable its use in the assessment of various national environmental impacts and their reduction possibilities.

The main contribution of this work was the development of a unique modeling framework to assess emission scenarios of multiple air pollutants in high sectoral and spatial resolution in Finland. The developed FRES model provides support for Finnish air pollution polices and a tool to assess the co-benefits and trade-offs of climate change strategies on air pollution.

Keywords: air pollution, emission, modeling, emission reduction, reduction costs, Finland

1. Introduction

Air pollution is defined as the presence of substances in the atmosphere above their natural levels, causing measurable undesired impacts on the environment (Seinfeld and Pandis 1998). The emissions of air pollutants can be either of natural origin (e.g. Simpson et al. 1999) or produced by human activity, i.e. anthropogenic air pollution. Impacts of major concern in recent decades include acidification, eutrophication, effects of tropospheric ozone (O_3) formed in the atmosphere and human health deterioration. These impacts are caused by emissions of the following air pollutants: sulfur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3), non-methane volatile organic compounds (NMVOCs) and primary particulate matter (PM) that are treated collectively in the air pollution emission reduction strategies of United Nations Economic Commission for Europe (UNECE) and European Union (EU).

Emission models are representations of the real systems that produce emissions (Winiwarter and Schimak 2005). They describe quantities, location and temporal variation of emissions from multiple sources in a coherent framework. The use of emission models enables the identification of key emission sources and assessment of emission reduction possibilities in the future. In the assessment frameworks of air pollution impacts, emission models play an important role as the provider of emission fields to atmospheric models. These modeling frameworks support the planning of effective emission reduction strategies to abate the environmental impacts of air pollution.

Correct emission estimates in the assessment of air quality and air pollution impacts are important (e.g. Harrison et al. 2008). Especially the importance of spatial emission source description and understanding of emission uncertainties have been highlighted (e.g. Miller et al. 2006). Furthermore, emission data requirements in terms of e.g. spatial resolution vary depending on the studied impacts.

This study focuses on the assessment of anthropogenic air pollution emissions of SO_2, NO_x, NH_3, NMVOCs and primary PM with a mathematical model, the Finnish Regional Emission Scenario (FRES) model. The FRES model is the first coherent framework that describes emission scenarios of multiple air pollutants in high spatial resolution over the area of Finland. Emissions from different emission sources and their future development are explored with the FRES model. In addition, FRES model emission uncertainties are estimated. Finally, recommendations on the characteristics of emission data in different modeling applications are presented.

1.1 Air pollution cycle

Anthropogenic air pollutants are released to the atmosphere from various emission sources. High-temperature processes, e.g. combustion and industrial production processes, cause emissions of SO_2, NO_x and primary PM. Airborne particles may also be formed in low temperatures as a result of mechanical wear of materials. Other important low-temperature sources of air pollution include agriculture (NH_3) and solvents use (NMVOCs).

After their release the emissions disperse and dilute in the atmosphere. The dispersion and dilution are strongly influenced by many external factors, e.g. meteorology and geographical landform. The distance that pollution travels from the emission source may vary from a few meters to thousands of kilometers. High flue gas stacks are employed in industrial plants in order to ensure efficient dispersion and dilution, and thus decrease the magnitude of pollution concentrations in the locations where the pollution will eventually cause the impacts. In contrast to high-stack emissions, emissions from low altitudes and near impact receptors, e.g. from vehicle tail pipes near large human populations, may cause exposure in relatively high concentrations in the immediate vicinity of the source.

The pollutants, once emitted to the atmosphere, go through various chemical reactions; some main processes are shortly presented here. Emitted sulfur gases oxidize to sulfates (SO_4^{2-}), which tend to condensate to form sulfate aerosol via gas-to-particle conversion. SO_2 is also oxidized to H_2SO_4 in cloud water. NO_x oxidize to nitrates (NO_3^-),

that can condensate to nitrate aerosols. NO_x also contribute to tropospheric O_3 photochemical formation together with carbon monoxide (CO), methane (CH_4) and NMVOCs. NMVOCs can also be converted to organic aerosols, partly as by-products of photochemical reactions with NO_x. NH_3 in the atmosphere may form ammonium (NH_4^+) aerosols by neutralizing SO_4^{2-} and NO_3^- acids. (Wallace and Hobbs 2006)

As described above, aerosols, i.e. PM, can be formed in the atmosphere from gases. These particles are called secondary PM. Gases may condensate onto existing particle surfaces, or they may form new particles by nucleation. In addition, part of the atmospheric PM is the result of emissions that are already in particle form when released to the atmosphere; they are called primary PM. Once airborne, particles go through chemical and physical processes; they can change their size and chemical composition by condensation, evaporation or coagulation with other particles. The size of atmospheric particles ranges from a few nanometers to tens of micrometers. The most commonly used particle size categories are PM_{10} (particle diameter below 10 μm), $PM_{2.5}$ (<2.5 μm), PM_1 (<1 μm) and $PM_{0.1}$ (<0.1 μm). The $PM_{2.5}$ size category is often called fine particles and particles larger than 2.5 μm coarse particles. (Seinfeld and Pandis 1998)

Gaseous and particulate air pollutants are present in the atmosphere in various concentrations and they interact with each other. Eventually they are removed from the atmosphere by wet and dry deposition. Environmental and health impacts are caused by deposition onto ecosystems or materials, or by concentrations present in ambient air. Different pollutants contribute to different impacts, and many of the pollutants contribute to multiple effects. Deposition of acid compounds (predominately SO_2, SO_4^{2-} and NO_3^-) causes acidification of ecosystems and corrosion and soiling of built environments. Eutrophication is caused by NH_3, NH_4^+ and NO_3^-deposition. Elevated ozone concentrations in the troposphere damage vegetation and human health. Human exposure to PM causes health deterioration, i.e. respiratory and cardiovascular diseases. Figure 1 summarizes the main sources and

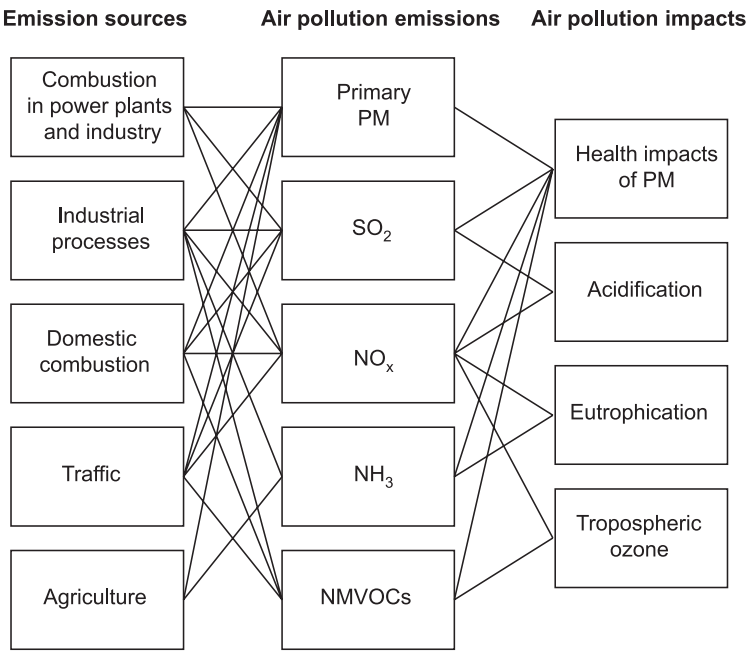

Figure 1. The main emission sources, air pollutants and impacts considered in this study (after Grennfelt and Pleijel 2007)

environmental impacts caused by the pollutants considered in this study.

In addition to the pollutants discussed above, there are several other air pollutants and impacts that are not dealt with in this study. Carbon monoxide (CO) is a toxic substance that also contributes e.g. to tropospheric ozone formation. Other toxic air pollutants include various heavy metals and persistent organic pollutants (POPs). Stratospheric ozone depletion is caused by emissions of industrially manufactured chlorofluorocarbons (CFCs). The emissions of greenhouse gases, i.e. carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O) and halocarbons (HCFs, PFCs and SF_6) increase retention of the earth's heat radiation and thus cause global warming. O_3 and airborne PM (i.e. aerosols) also affect the earth's radiation balance, although they are not included in the protocols to abate global warming. (Wallace and Hobbs 2006)

1.2 Emissions

The following sections present the main formation mechanisms and sources of primary PM, SO_2, NO_x, NH_3 and NMVOCs emissions. The emission characteristics and sources of primary PM are more diverse than those of gaseous pollutants, and therefore they are given relatively more attention.

1.2.1 Primary particulate matter

Primary particulate matter (PM) denotes emissions that are in particle form, i.e. solid or liquid, when they are released to the atmosphere, in contrast to secondary PM that are formed via gas-to-particle conversion in the atmosphere. Primary PM may be comprised of a variety of chemical compounds, e.g. organic and elemental carbon, sulfates and various minerals and trace elements (Lighty et al. 2000). Anthropogenic primary PM originates predominately from combustion and other high-temperature processes, and from activities causing suspension of particles produced by the mechanical wear of solid materials, i.e. fugitive dust. High-temperature processes produce predominately fine PM below 2.5 μm, whereas fugitive dust emissions are mainly coarse particles.

Combustion is a major source of anthropogenic air pollution. Primary PM emissions from combustion processes can be divided into two categories based on their origin: (1) ash, i.e. a combustion product formed from non-combustible mineral constituents in fuel, and (2) carbonaceous particles, e.g. char, coke, soot and condensed hydrocarbons, which are formed by pyrolysis of unburned fuel molecules (Flagan and Seinfeld 1988). In the case of solid fuel combustion in controlled processes, e.g. power plants, fuel molecules are combusted almost completely, and ash-forming species are the main source of particles. Carbonaceous particles are the product of incomplete combustion processes e.g. in small domestic heating appliances.

Ash-forming minerals form particles of different sizes depending on e.g. mineral matter composition and combustion conditions. The mineral matter, occurring as mineral inclusions or heteroatoms, consists of refractory and more volatile species. The refractory compounds are not directly volatilized at the temperatures prevailing in normal combustion processes, and they form mainly relatively large sized particles (1 - 50 μm). The mineral compounds that volatilize in high temperatures form fine PM (<1 μm) by nucleation, condensation and coagulation. (Flagan and Seinfeld 1988)

Combustion processes in small domestic appliances are not usually as well controlled as in large boilers. Especially domestic wood combustion has been shown often to entail incomplete combustion and thus relatively high PM emissions (McDonald et al. 2000, Boman 2005, Tissari et al. 2007). Part of the volatile hydrocarbons of wood remain unburned and entrain the flue gases, forming fine particles of soot and organic compounds adsorbed or condensed on the surfaces of the soot particles.

Internal combustion engines in traffic vehicles and machinery are important sources of fine PM. Diesel engines cause particles consisting mainly of soot, organic PM and residues from fuel and lubricant additives. Gasoline-fuelled four-stroke vehicles produce

relatively smaller masses of exhaust particles than diesel vehicles with high fractions of organic compounds (Lighty et al. 2000). Two-stroke technology, typical in small engines such as mopeds and machinery, produces relatively high emissions of unburned fuel and lubricants (Etissa et al. 2008, Volckens et al. 2008). International ship traffic also produces significant PM emissions (Fridell et al. 2008). However, ship emissions are included in this work only for the part of domestic shipping, which is a relatively minor source of PM.

Liquid and gaseous fuel combustion in stationary sources causes mainly carbonaceous PM emissions in relatively small amounts. Particles from heavy fuel oil boilers are mainly unburned carbonaceous coke particles relatively large in size (1 - 50 μm), whereas light fuel oil combustion causes predominately fine (0.01 - 0.5 μm) soot and organic particles (Flagan and Seinfeld 1988). Combustion of gaseous fuels typically produces very low emissions of fine carbonaceous particles with high fractions of organic compounds (Hildemann et al. 1991).

In addition to combustion, many industrial high-temperature processes cause particle emissions originating from volatilized raw or process materials. These often contain high fractions of fine PM. Examples of such emissions include metallic fumes from various metal industry smelters (Passant et al. 2000) or alkali salt ash particles from black liquor combustion in the pulp and paper industry (Mikkanen et al. 1999). In addition to industrial scale, there are also some very small high-temperature sources, e.g. meat frying, tobacco smoking and fireworks, producing mainly organic particles finer than 1 μm (Kleeman et al. 1999, Vecchi et al. 2008).

Many human activities cause particle emissions in the form of fugitive dust. Fugitive dust emissions are the product of mechanical abrasion of materials and their suspension caused by various activities. Dust particles are predominately larger than 2.5 μm (Chow et al. 1994). Road traffic is a considerable source of fugitive dust emissions, originating from road abrasion, tyre and brake wear, and suspension of dust deposited on road surfaces (e.g. Kupiainen 2007). Agricultural activities cause fugitive dust associated with animal husbandry, field preparation and crop harvesting activities. (Takai et al. 1998, Clausnitzer and Singer 1996). Considerable fugitive dust emissions have also been estimated for peat extraction activities (Tissari et al. 2006), material storage and handling, and construction activities (USEPA 1998).

1.2.2 Sulfur dioxide

Sulfur dioxide (SO_2) is formed in high temperature processes by the oxidation of sulfur contained in fuels or process materials. The most important anthropogenic source of SO_2 is fuel combustion. Coals and heavy fuel oils contain considerable percentages of sulfur, typically from 0.5 to 3.0% (Flagan and Seinfeld 1988). Peat, which is a commonly used fuel in Finnish heat and power plants, also contains sulfur, although in slightly lower shares at around 0.2% (Alakangas 2000). Wood fuels and natural gas typically contain negligible amounts of sulfur, and cause thus practically no SO_2 emissions. On-road vehicles in Europe nowadays use sulfur-free diesel oils and gasoline.

In addition to combustion, SO_2 can be formed in industrial processing of sulfur-rich process materials. Some industrial activities, e.g. pulping processes in paper production, also cause sulfur emissions in reduced form, often called total reduced sulfur (TRS) (Bordado and Gomes 2003). TRS emissions oxidize in the atmosphere to SO_4^{2-}, and are therefore from the point of view of acidification equivalent to SO_2 emissions. In this study, all anthropogenic sulfur emissions to air are referred as SO_2.

1.2.3 Nitrogen oxides

Nitrogen oxides emissions (NO_x, i.e. the sum of NO and NO_2) are formed in high temperature processes in the oxidation of nitrogen contained in fuels or process air. Fuel-NO_x formation depends on the fuel composition, being important for fuels with high nitrogen content, e.g. coals. Thermal-NO_x is the main formation mechanism of NO_x from atmospheric N_2. The formation of thermal-NO_x is strongly

dependent on process temperature: the higher the temperature, the higher the formation rate. (Flagan and Seinfeld 1988)

All combustion processes cause NO_x emissions. Since the rate of NO_x formation depends strongly on temperature, processes with high combustion temperatures tend to produce high emissions. Such processes include large combustion boilers and internal combustion engines. Therefore combustion processes in large power plants are often modified to conditions that favor low NO_x emissions (for more about emission control see Section 1.3). In smaller combustion devices, e.g. for domestic heating, temperatures and thus NO_x emissions are lower.

1.2.4 Ammonia

In contrast to the formation of SO_2, NO_x and the majority of fine PM emissions, ammonia (NH_3) emissions are primarily caused by other processes than combustion. The main anthropogenic emission source is agriculture, i.e. manure of domestic animals and nitrogen-containing fertilizers. Some industrial processes, e.g. fertilizer production, also cause considerable ammonia emissions. Recently gasoline-fuelled vehicles have been detected to produce considerable NH_3 emissions (e.g. Heeb et al. 2006). In Finland, for example, traffic is estimated to cause 2.7 $Gg(NH_3)\ a^{-1}$, or 7% of the national total in 2006 (Finnish environment institute 2008, see Figure 2). The FRES model versions considered in this study do not include NH_3 emission calculation for traffic vehicles.

1.2.5 Non-methane volatile organic compounds

The term non-methane volatile organic compounds (NMVOCs) is used to denote vapor phase atmospheric organics, e.g. alkanes, alkenes and aromatics, excluding CH_4 (Theloke and Friedrich 2007). Anthropogenic NMVOCs emissions originate from unburned fuels in combustion and from low temperature evaporation of organics. The main contributing sources are traffic vehicles (in the form of both fuel evaporation and tailpipe exhaust), incomplete combustion processes (e.g. domestic wood combustion), solvents and industrial processes.

1.3 Emission control technologies

Reduction of air pollution emissions by technical measures has been an important element in the decrease of emissions during recent decades. The emissions of primary PM, SO_2, NO_x and NMVOCs from large power plants and industrial units, and lately also from traffic vehicles, have declined considerably thanks to environmental legislations that require the utilization of emission control technologies (Figure 2). For NH_3 there are no emission control technologies in use in Finland, and the emissions depend mainly on the volumes of animal husbandry and fertilizer use.

The emissions can be reduced by abating pollutants in flue gases after the process before release to the atmosphere (end-of-pipe technologies) or by performing modifications in the process or fuels in order to achieve conditions in which the formation of pollutants is decreased (primary measures). This section introduces the most important control technologies for combustion and industrial emission sources. This study does not cover the abatement of agricultural NH_3 or non-combustion NMVOCs emissions. Papers II and IV (primary PM) and Paper III (SO_2 and NO_x) present the main emission control technologies of the FRES model; therefore only a summarizing presentation is given here.

Air pollution emissions from large combustion and industrial sources have been abated for several decades. High PM concentrations in flue gases are reduced with end-of-pipe equipment such as electrostatic precipitators (ESPs), fabric filters and wet scrubbers, with removal efficiencies often higher than 99% (Ohlström et al. 2000). Cyclones and multi-cyclones are less efficient, and are used mainly in smaller combustion plants of a few megawatts in capacity and for the reduction of coarse PM.

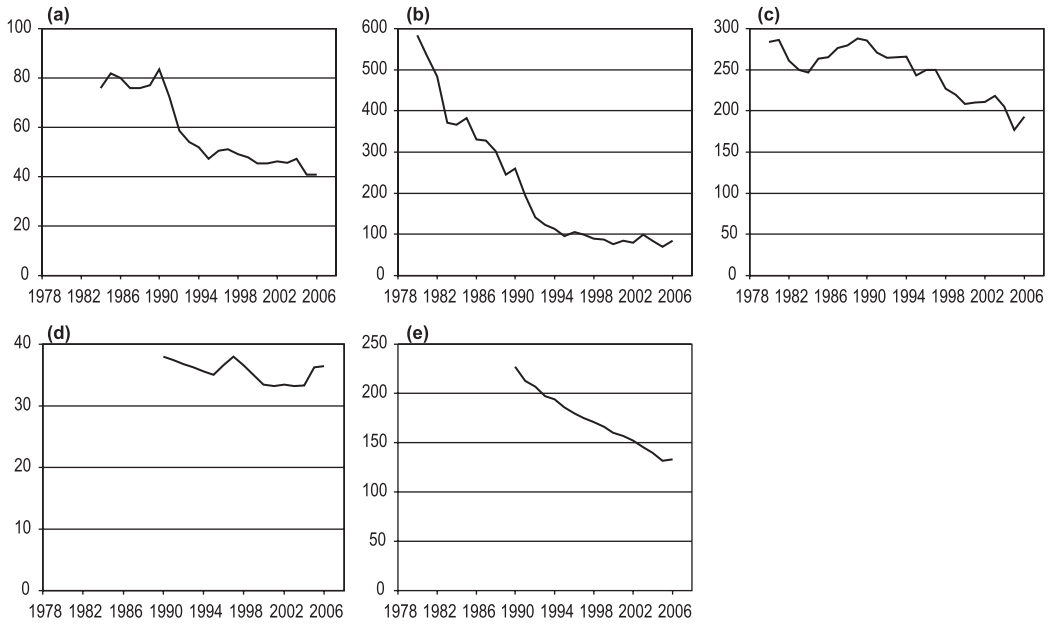

Figure 2. The emissions of (a) primary PM (TSP), (b) SO_2, (c) NO_x, (d) NH_3 and (e) NMVOCs in Finland. For TSP, only combustion and industrial process based emissions are included. For NH_3, traffic emissions are included only for 2005-2006. Sources: 1990-2006: Finnish environment institute (2008), except TSP: Statistics Finland (2008); 1980-1989 (SO_2 and NO_x) and 1984-1989 (TSP): Statistics Finland (2005). Unit Gg a^{-1}.

NO_x emissions in industrial and power plants are reduced by both primary measures and end-of-pipe technologies. Primary reduction measures refer to in-process modifications of combustion conditions that reduce NO_x formation. Common technologies include e.g. low-NO_x burners, staging of combustion air and flue gas recirculation. Reduction efficiencies are often relatively low, around 20 to 70% (Graus and Worrell 2007). Higher efficiencies can be achieved by end-of-pipe measures, or the combination of primary and end-of-pipe measures. The most commonly used are selective catalytic (SCR) and non-catalytic reduction (SNCR).

The emissions of SO_2 from large plants are reduced with flue gas desulfurization (FGD) appliances. Flue gases are led into contact with sorbent material (typically calcium) that reacts with SO_2. The reduced material is taken out as wet slurry (wet FGD) or collected by particle removal devices (spray dry FGD and limestone injection into a boiler). Removal efficiency in coal power plants can be above 90% for wet FGD and lower for dry and limestone injection methods (Graus and Worrell 2007).

In addition to FGD, sulfur emissions can be reduced by reducing the sulfur content of fuel or process material. The reduction of fuel sulfur content is most widely applied for fuel oils. Low-sulfur oils used in power plants and domestic boilers typically have sulfur contents at around 0.1% and 1.0% for light and heavy fuel oils, respectively. For traffic vehicles the use of ultra-low-sulfur (<0.001%S) gasoline and diesel oils has become a standard in recent years. The use of low-S oils typically leads to additional reduction in PM emissions (Goldstein and Siegmund 1976).

Traffic vehicle emissions have not been controlled until recent years. Three-way catalysts in gasoline-driven vehicles, reducing the emissions of NO_x, NMVOCs, CO and PM, became prevalent in the 1990s. Diesel vehicle emissions can be controlled by optimized engine design and flue gas end-of-pipe treatment (e.g. oxidation catalysts, particle filters and SCR). The use of end-of-pipe measures in new diesel vehicles will be required by about 2010

because of increasingly stringent European emission standards (e.g. EC 1998).

For small domestic combustion sources the use of emission reduction technologies is not common at the moment. Practically, emission legislations require only the application of low-sulfur oils (EC 1999a). However, the emissions of especially PM and NMVOCs from domestic solid fuel combustion can be relatively high, and interest in their reduction has increased. Abatement technologies for domestic scale, e.g. ESPs, have been developed in recent years (see Paper IV of this study).

In addition to technical end-of-pipe or process modification measures, emission reductions are possible by various measures that involve changes in the structure or inputs of production processes or consumer behavior leading to decreased pollution. Examples of these include fuel switch or efficiency improvement of energy production, traffic infrastructure planning or public information campaigns directed to behavioral changes. Such measures can be assembled under the term non-technical measures (NTM) (Sternhufvud and Åström 2005). However, emission abatement by NTM is not included in this study.

1.4 International emission reduction agreements

Air pollution emissions increased in late 1900s along with the strong increase in industrial activities in developed countries. The negative impacts on the environment and the transboundary nature of air pollution became slowly evident. Acidification of lakes, streams and forests in the 1960s and 70s triggered political actions to reduce the impacts at both national and international levels. Later the reduction of eutrophication, tropospheric ozone and PM health impacts were also addressed. Air pollution assessment by computer models has been an important factor in the support of the policy processes. This section introduces the history of the multilateral emission reduction agreements of transboundary air pollution in Europe.

The first multilateral instrument to reduce cross-border emissions, the Convention on Long-range Transboundary Air Pollution (CLRTAP), was signed in 1979 by 32 UNECE member States in Europe and North America. Currently 51 Parties have ratified it. The first action was to address sulfur related impacts, in particular acidification. The first protocol on pollutants was signed in 1985. It obligated each signatory to reduce its sulfur emissions by 30% by 1990 from the base year 1980 level. Later protocols addressed the emission reductions of NO_x (1988), NMVOCs (1991), sulfur (1994), heavy metals (1998), POPs (1998) and multiple pollutants (1999).

The first protocols comprised flat percentage reductions and technical measures for the Parties. However, because of national differences in economic structure and the degree of emission reductions already adopted in the base year, flat rates were not equally feasible or costly to achieve in different countries. For more stringent reduction requirements, the flat rate approach was not deemed to treat countries equally justly. Furthermore, sensitivity of receptors, e.g. ecosystem tolerance against acidifying deposition expressed by the critical load concept, varies largely in different parts of Europe. More sophisticated reduction strategies were called for that would take these country- and area-specific circumstances into account.

The 1994 Sulphur Protocol was the first CLRTAP protocol to base its reduction obligations on ecosystem sensitivities (critical loads). The attainment of environmental targets was reflected in needs to reduce depositions at specified locations and allocation of cost-efficient emission reductions among the Parties. As a result, different emission reductions were set for each country as national total emission caps.

The 1999 Gothenburg Protocol (UNECE 1999), was the first protocol to address multiple pollutants and multiple effects simultaneously. It comprised the same effects-based approach as the 1994 Sulphur Protocol and included interconnections between different pollutants and impacts at the same time (see Figure 1). The Gothenburg Protocol lays down country-

specific emission caps for SO_2, NO_x, NH_3 and NMVOCs that are based on the reduction of exceedance of critical loads of acidification, eutrophication and ground-level ozone impacts on vegetation and health (the gap closure approach). Integrated assessment models (IAM), the RAINS model in particular, played a prominent role in the determination of the emission reduction strategies in both protocols (see Section 1.5.3).

In parallel to the work of UNECE/CLRTAP, the European Commission prepared its reduction strategies of air pollution in Europe. An approach similar to the multi-pollutant multi-effect Gothenburg Protocol was adopted: It addressed same pollutants and used the same modeling framework (the RAINS model). As a result, the National Emissions Ceilings (NEC) directive (EC 2001a) was adopted. The NEC directive and CLRTAP protocol obligations for Finland are given later in Table 5 (Section 4.2).

After the adoption of the 1999 Gothenburg Protocol and the NEC directive, health effects caused by PM gained increasing attention as a new element in the analysis framework. In 2001 the European Commission launched a thematic strategy on air pollution, which was prepared under the Clean Air for Europe (CAFE) programme (EC 2005). The CAFE programme was set up to take a broad view on air pollution in Europe, comprising air quality directives and the thematic strategy, with a strong emphasis on PM health effects. As a result of the CAFE programme, the proposed revision of the NEC directive sets national emission ceilings for PM in 2020, in addition to revised ceilings on pollutants included in the original directive. CLRTAP decided in 2007 that the first review of the 1999 Gothenburg Protocol was completed (UNECE 2007). It then initiated its revision, which should include new scientific knowledge about primary PM and PM precursors to evaluate further cost-effective abatement measures.

In addition to the multilateral environmental agreements presented above, there are also other types of European wide legal instruments, which aim to reduce emissions at sector or industrial plant type level. These include i.a. limits for allowable emission levels per activity for combustion plants (e.g. Large Combustion Plants (LCP) directive EC 2001b), limits for vehicles (EURO standards, e.g. EC 1998), limits for sulfur contents of fuels (e.g. EC 1999a), practices for NMVOCs reduction (EC 1999b, 1994) and recommendations for best available technologies (BAT) in the Integrated Pollution Prevention and Control (IPPC) directive (EC 2008). Air quality limit values (e.g. EC 1999c) do not directly regulate emissions, although in the case of exceedance, action plans to reduce emissions at local level might be required.

1.5 Tools for emission and impact assessment

The assessment of air pollution emissions to produce information for the reduction of their negative impacts is not trivial. Emission estimates of multiple pollutants from a wide variety of sources is required. In order to link the emissions with their environmental impacts, the transport of pollutants and the location and sensitivity of different impact receptors must be estimated. In order to manage the assessment of these extensive and multidimensional emission - impact relationships, complex mathematical models have been developed.

Emission inventories and emission models are employed in order to estimate emissions originating from various sources. The fate of the emissions, i.e. their contribution to atmospheric concentrations, can be estimated with atmospheric models that utilize emission dispersion and meteorology representations. In order to estimate the impacts of air pollution, emission models together with dispersion estimates can be combined with corresponding environmental and health impact receptors, e.g. the tolerance of ecosystems to acidification or human populations subject to exposure. The descriptions of applied and potential emission reduction technologies and reduction costs in the emission models enable the assessment of cost-effective emission abatement alternatives.

Model systems that combine information from the above mentioned components

(emissions and their reduction potentials and costs, dispersion and impacts) are often known as integrated assessment models (IAMs). Figure 3 shows a schematic view of the Finnish IAM framework. IAMs describe emission – impact relationships over wide geographical areas, typically on the level of countries or continents, i.e. on a regional level. They can be used in environmental policy planning and assessment; different emission control scenarios can be evaluated from the perspective of both reduction costs and environmental benefits. The following sections introduce different aspects of emission models and IAMs applied at European and national levels.

1.5.1 Emission inventories

Air pollution emission inventories are formal compilations of emissions reporting to conventions, e.g. Convention on Long-range Transboundary Air Pollution (CLRTAP). The emission estimates are collected into inventories or databases in the format that follows the requirements within respective reporting guidelines (e.g. UNECE 2003). The required format may include simply the reporting of annual country total emissions (e.g. for the NEC directive, EC 2001a), following certain source sector nomenclature (e.g. Selected Nomenclature for Air Pollution (SNAP) of the CORINAIR (2007) inventory) and regular grid definition (e.g. EMEP 50 × 50 km^2 grid, Vestreng 2003).

Procedures to compile emission inventories vary in different countries (e.g. Johansson et al. 2001). They often contain calculation systems that have similar characteristics to emission models, especially in the estimation of area source emissions, but they may also involve simply a collection of measured emissions of e.g. power plants into databases. Furthermore, emission inventory systems are often not designed for the estimation of future emissions, which is an important feature of emission models. Formal emission inventories complemented with spatial and temporal emission processing routines can be also used as input to atmospheric models (e.g. Borge et al. 2008, see Table 1). The characteristics and application of emission models are introduced in the next section.

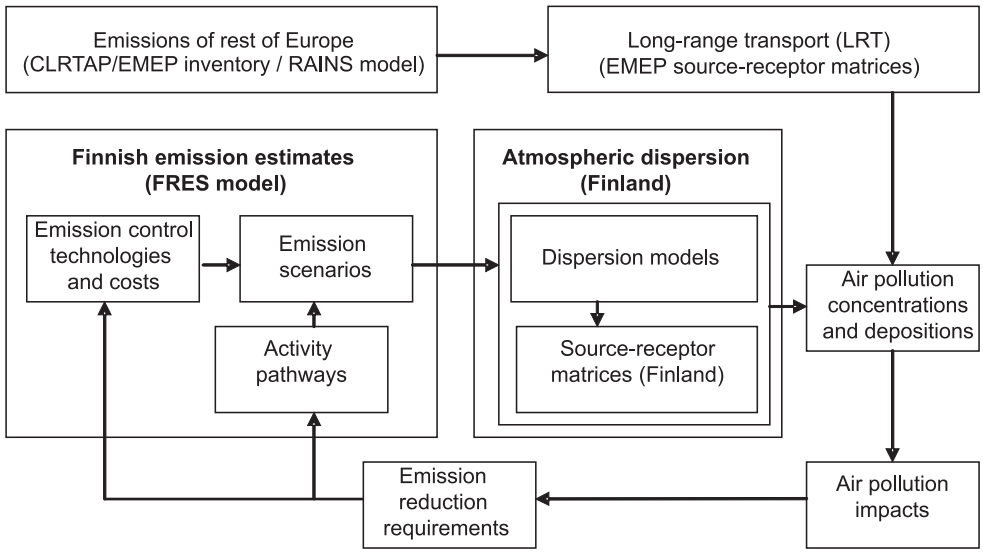

Figure 3. Flowchart of the Finnish integrated assessment modeling (IAM) framework (after Johansson 1999 and Johansson et al. 2003). The FRES model provides estimates of Finnish emissions. Their dispersion is estimated with atmospheric models or source-receptor matrices for Finland (see Section 3.2). Long-range transport is assessed with the emissions of other European countries (EMEP or RAINS) and their dispersion estimates by EMEP source-receptor matrices.

1.5.2 Regional emission models

Emission models enable the assessment of emissions and reduction possibilities of multiple pollutants and sources in a coherent framework. Their geographical scale may vary from local to regional to global. This section, and this study, concentrates on regional scale emission models that can be used as part of IAMs of air pollution. Regional scale in this context refers to a domain of hundreds to thousands of kilometers, typically of a continent, country or a part of a country (Pleijel and Grennfelt 2007).

In technical terms emission models are representations of input (economic activity) – output (emissions) relationships of certain economic sectors (e.g. industry - traffic - domestic sector) in a certain geographical area (e.g. a country) and time frame (e.g. one year). As a basis for emission calculation, emission models typically employ the description of activity units (e.g. energy consumption) and emission factors (i.e. emission quantities per activity unit) in a certain sector categorization. Utilized and potential emission control technologies are typically described since they often have significant effects on emission factors. The estimates of future activities and factors affecting changes in emission factor, i.e. primarily the use of emission control technologies, enable the assessment of alternative future emission developments. When used as a part of IAM, the emissions are spatially and temporally disaggregated within the geographical and temporal domain of the model.

Spatial and temporal resolutions

The practical temporal and spatial resolutions of emission fields depend on the extent of the modeling domain and the need of the specific model application to describe the studied environmental impacts accurately. When the emission results are to be used as input in atmospheric models, the temporal resolution depends primarily on the compatibility with the applied meteorology data within the atmospheric model. In regional scale applications typically 1 hour temporal resolution is used (Cuvelier et al. 2007).

The spatial resolution of emissions is typically the coarser the wider is the domain area of the model. For example in the European-wide assessments, the emission inventory of EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air pollutants in Europe) of CLRTAP exploits emission data at 50×50 km^2 grid resolution (Vestreng 2003), and the RAINS model of IIASA (International Institute for Applied System Analysis) (Schöpp et al. 1999) utilizes country total emissions as input to its scenario analysis. Figure 4 presents the Finnish submission to CLRTAP/EMEP of $PM_{2.5}$, SO_2 and NO_x emissions in 2000 (Finnish environment institute 2008).

National or sub-national level models, in contrast, typically employ emissions in higher resolution than is used in European level assessments. Resolutions down to 1×1 km^2 are typical in national applications (e.g. King et al. 2006 for the United Kingdom, this study for Finland). The higher resolution is currently possible due to improved availability and practical management of representative high resolution disaggregation data from national databases. Table 1 summarizes examples of national emission models and inventories that have been applied in regional air pollution impact studies in different countries.

The spatial resolution in emission and atmospheric models affects the precision of modeling system performance when used for the assessment of different environmental impacts. Spatial resolutions of tens of kilometers are adequate to describe emissions and impacts that are primarily caused by long-range transport (LRT) of air pollution, e.g. acidification and eutrophication. The concentrations and health impacts of PM and ozone are strongly influenced by LRT, but considerably also by local emission sources, especially in urban areas. In order to assess these more local scale aspects, e.g. on the level of cities or parts of cities, higher spatial resolution than that used in European level models is beneficial. Thunis et al. (2007) reported that models for PM and ozone at 5 km resolution perform better than the RAINS model in urban areas.

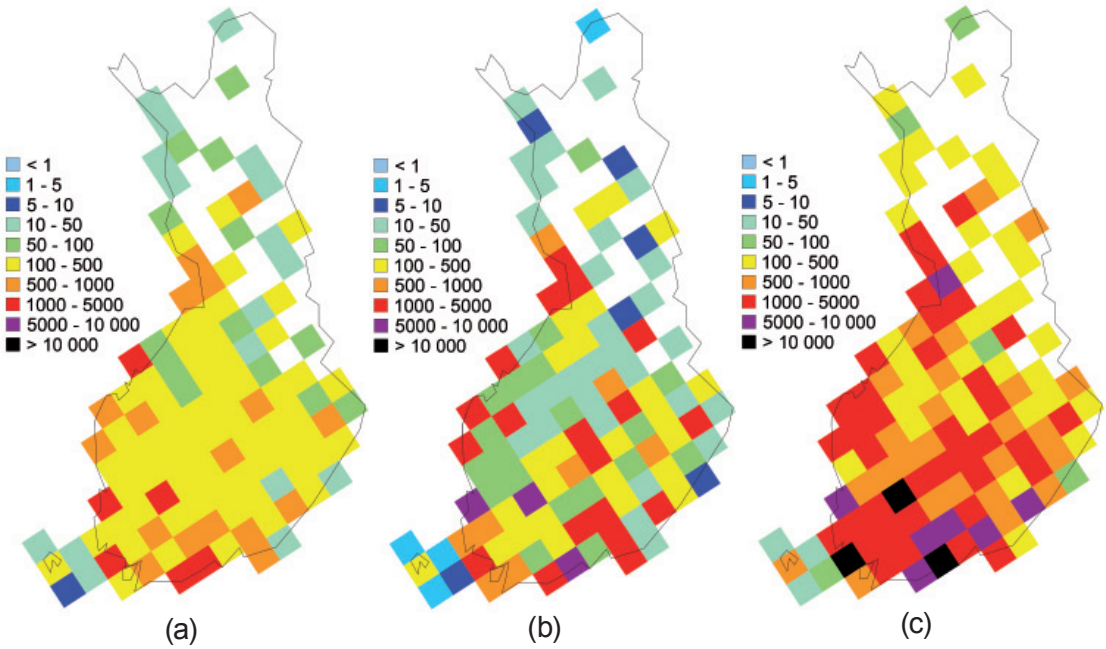

Figure 4. Total emissions of (a) $PM_{2.5}$, (b) SO_2 and (c) NO_x of the Finnish CLRTAP inventory in 2000 in the EMEP 50 × 50 km² grid (Finnish environment institute 2008). Unit Mg a⁻¹.

Table 1. Examples of national emission models and inventories, their basic characteristics and regional air pollution impact applications.

Country	Emission model / inventory	Modeled pollutants	Description of emission sources	Spatial emission resolution	Types of application
Finland	FRES[1]	Primary PM, SO_2, NO_x, NH_3 and NMVOCs	Combined bottom-up, top-down	1 km	Acidification, trop. ozone[2]; PM exposure[3]
The U.K.	NAEI[4]	Primary PM, SO_2, NO_x, NH_3, NMVOCs, CO, GHGs, heavy metals etc.	Combined bottom-up, top-down	1 km	Acidification, eutrophication, PM exposure[5]
Spain	Spanish CORINAIR inventory + SMOKE emission processor[6]	Primary PM, SO_2, NO_x, NH_3, NMVOCs and CO	Combined bottom-up, top-down	15 km (Madrid area: 5 km)	Trop. ozone[7]
Italy	RAINS emissions for Italy + emission processing[8]	Primary PM, SO_2, NO_x, NH_3 and NMVOCs	Combined bottom-up, top-down	20 km (northern Italy 4 km)	Acidification, eutrophication, trop. ozone, PM exposure[8]

[1)] This study; [2)] Paper I of this study; [3)] Paper VI of this study; [4)] NAEI = National Atmospheric Emissions Invetory, King et al. 2006; [5)] Oxley et al. 2003; [6)] Borge et al. 2008; [7)] Jiménez et al. 2007; [8)] Zanini et al. 2005

Top-down and bottom-up approaches

The number and nature of emission sources vary widely between different emission source sectors. This influences the way in which they can most advantageously be treated in emission models on different scales. The emissions of large polluters, e.g. industrial or power plants, can be calculated as individual point sources. Different characteristics of the plant that are relevant to emissions can be described, e.g. plant capacity, applied combustion and emission control technologies, and stack locations and heights. This type of individual source basis calculation is called the bottom-up approach (Winiwarter and Schimak 2005).

Contrary to large point polluters, many emission sector categories consist of a high number of relatively small individual polluters, e.g. traffic vehicles or domestic heating boilers. In these cases the emission calculation of each individual source would make the emission model laborious to manage and heavy to use. Furthermore, the availability of information from such small emitters on an individual basis is often limited. Therefore, such sectors are typically calculated as a group of polluters with similar characteristics, i.e. with the top-down approach.

Emission models with different spatial domains exploit top-down and bottom-up approaches variably. The European wide RAINS is a top-down model. From the whole European perspective individual energy production and industrial plants appear with such a high number and relatively low emissions that it is justified to describe them as area sources. Smaller domain models, by contrast, often utilize a combined approach describing the largest plants as point sources and the rest as area sources (e.g. King et al. 2006, Zanini et al. 2005, this study).

The different approaches influence how precisely emissions from different sources can be described, both in terms of emissions quantities and spatial distribution. For example, urban area emission estimates derived from the EMEP inventory have been shown to differ considerably (up to 7-fold) from corresponding local estimates (Cuvelier et al. 2007). The differences were estimated to derive to a large extent from the top-down and bottom-up emission approaches of the EMEP and local inventories, respectively.

1.5.3 Integrated assessment models

The development of integrated assessment models (IAMs) of air pollution in Europe started in the 1980s. They have been used to support the preparation of several emission reduction strategies of the UNECE and EU (see Section 1.4). Results from several frameworks have been used in the strategy evaluations, e.g. the RAINS model developed at the International Institute for Applied Systems Analysis (IIASA) (Schöpp et al. 1998), ASAM (Abatement Strategy Assessment Model) of the Imperial College (ApSimon et al. 1994), CASM (Coordinated Abatement Strategy Model) of the Stockholm Environment Institute (Gough et al. 1995) and MERLIN (Multi-pollutant, Multi-Effect Assessment of European Air Pollution Control Strategies: an Integrated Approach) of the EU project led by the University of Stuttgart (Reis et al. 2005). Especially in the 2000s the RAINS model has been the central model in the strategy development.

The European RAINS model incorporates the description of country-level emissions of multiple pollutants, emission dispersion and impacts, and the least-cost optimization of future emission reductions to reduce environmental stress. Emissions and their future scenarios are based on the description of activity levels, emission factors and emission control technology use in different economic sectors in each country. As a basis for the analysis, future activity levels in so-called national activity pathways are determined by the means of economy and energy modeling both by each country individually (e.g. Lehtilä et al. (2005) for Finland) and collectively for all countries in Europe (e.g. Mantzos and Zeka-Paschou 2004). Pollution dispersion and transformation, i.e. contributions of emissions from each country to pollutant concentrations and depositions, are described by so-called source-receptor matrices that are based on European scale atmospheric modeling by EMEP (Tarrason et al. 2003).

Impact receptors include various natural and cultivated ecosystems with their sensitivities to acidification, eutrophication and ozone, and human populations susceptible to exposure. Emission reduction potentials and costs in each country are assessed by the means of feasible emission reduction measures and their marginal reduction costs. The optimization determines required emission reductions in each country in order to achieve preset environmental targets with minimum costs.

In addition to the continental scale, IAMs have been developed at the national level in many countries (Table 1). National IAMs can utilize emissions from national emission models and operate at higher spatial resolution than continental scale models. Johansson et al. (2001) assembled experiences from Denmark, Finland, Spain and Sweden with IAMs at different stages of progress. A more developed national application is the UKIAM model that addresses acidification, eutrophication and PM exposure at 1 and 5 km spatial resolutions for major cities and other areas, respectively, in the United Kingdom (Oxley et al. 2003, Oxley and ApSimon 2007). IAM in Spain has concentrated primarily on tropospheric ozone (Jiménez et al. 2007, Borge et al. 2008). Some of the national IAMs have been designed as national versions of the RAINS model with 5 to 20 km resolutions (e.g. Zanini et al. 2005 for Italy). To support the development of the models and encourage collaboration between national activities and with IIASA, the Network for National Integrated Assessment Modelling Activities has been established under the UNECE CLRTAP (www.niam.scarp.se). The Finnish IAM framework with the FRES model (Figure 3, Papers I and VI of this study) is also a part of the network.

The benefits of higher spatial resolution have also emerged in European level assessment, especially with the recent focus on human health impacts. Local scale effects in urban areas are an important factor to be addressed in the assessment of exposure of populations to air pollution. The EU project CityDelta has been launched in the context of the EU CAFE programme in order to integrate urban air quality characteristics into the European IAM framework at 50 km spatial resolution (Cuvelier et al. 2007). CityDelta applies results from several participating modeling exercises at 5 to 50 km resolution, and derives "ensemble" model predictions from the average of different models. The "ensemble" results are to be used to estimate the magnitude of urban effects on air quality in European cities.

2. Objectives and structure of this study

The objective of this study was to develop and apply a national emission model, the Finnish Regional Emission Scenario (FRES) model. The FRES model is the first mathematical framework that describes multiple air pollution emissions and emission scenarios comprehensively in high spatial resolution over the area of Finland. It has been developed to flexibly estimate the emissions resulting from different activity pathways and assess emission reduction possibilities. Other Finnish country-level air pollution emission estimates include formal inventories that present sector emissions for the whole country (Statistics Finland 2008) or for EMEP 50 × 50 km^2 grid resolution (Finnish environment institute 2008) and do not contain future emission estimates. The Finnish MARKAL/TIMES model (Lehtilä et al. 2005) that is primarily used for energy system optimization and greenhouse gas assessment also includes air pollution emissions from combustion sources at country-level.

This study presents results from different types of FRES model applications at different stages of the model development. On the basis of the results the following specific research questions can now be answered:

- What emission model characteristics support impact assessment of air pollution?
- What are the key emission sources in Finland today and in the future?
- How will different climate policies affect emissions of air pollution in the future?
- Which emission sources hold further cost-efficient emission reduction potential in the future?

- What are the main sources of emission uncertainty?

The structure of the study is as follows: Section 3 presents the FRES model structure and other methods and data used in the study. The general emission calculation approach, main calculation parameters and their data sources, as well as the procedure of emission disaggregation in space and time of the FRES model are presented. The FRES model does not describe emission dispersion or impact receptors, and their assessment is not the aim of this study. Therefore, methods to assess emission dispersion and impacts in Papers I and VI are presented only in descriptive manner.

The results of model applications are presented and discussed in Section 4. The results are structured to the following sub-sections:

- Finnish emissions in the model base year 2000 are presented in Section 4.1. The characteristics of key emission sources for different pollutants are analyzed: contributions to total emissions, spatial emission distributions and applied emission control technologies (Papers I, II, IV and V).
- In order to assess air quality changes in the future, it is important to know how the emissions from different sources will develop. Section 4.2 presents estimates of future emissions and discusses influential factors behind them. Both general air pollution emission development from the year 2000 to 2020 and the effect of different climate change mitigation measures on air pollution are analyzed (Papers I, II, IV and VI).
- Environmental legislations define the level of required emission controls in the future. Further emission reduction potentials and related costs in 2020 are estimated in Section 4.3. Cost curves are presented as illustrations of cost-efficiencies of the emission reductions (Papers III and IV).
- Section 4.4 presents FRES model applications in the assessment of environmental impacts: acidification and tropospheric ozone (Paper I), and PM exposure (Paper VI). The performances of applied model set-ups are discussed from the emission modeling perspective.
- Verification of emission modeling is important to assess the quality of emission estimates and to identify focuses for further model development. Section 4.5 presents comparisons of FRES results with other emission estimates (Papers II and V), and an analysis of emission uncertainties (Paper V). Furthermore, the role of verifications as a contributor to FRES model development is analyzed.

Section 5 draws conclusions from the model application results and answers the specific research questions set above. Finally, Section 6 presents outlines for future model development.

3. Methods and data

3.1 Finnish Regional Emission Scenario (FRES) model

The Finnish Regional Emission Scenario (FRES) model has been developed to provide information for the assessment of air pollution emissions and emission abatement possibilities in Finland. For emission impact assessment, it can be used as a part of IAM systems of air pollution. The results are aimed at the promotion of policy-making to reduce negative impacts of air pollution.

The model structure and calculation approach has been documented in the Papers I – VI for various versions and parts of the model (Table 2). In the following, the structure and main calculation parameters of the model version 2.1 are presented in a summarizing manner with reference to the relevant papers and other data sources. The emission results presented in Section 4 are based on FRES version 2.1 unless otherwise indicated.

3.1.1 Emission calculation and source aggregation

The FRES model consists of coherent emission calculation of several air pollutants from anthropogenic sources: primary PM in different size categories (TSP (total suspended particles), PM_{10}, $PM_{2.5}$, PM_1 and $PM_{0.1}$), sulfur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3) and non-methane volatile organic compounds (NMVOCs). In addition, the calculation of primary PM includes the fractionation to main chemical species (black and organic carbon, sulfate, main heavy metals and mineral matter)[1)] The emissions are calculated from the parameters of activity levels, emission factors and emission control technology removal efficiencies and utilization rates (Figure 5). The basic spatial and temporal domains of the model are the country of Finland and one year, respectively, which are then disaggregated to 1 km and 1 hour resolutions, respectively (Section 3.1.5).

The emission sources are aggregated into source sector categories. The FRES aggregation is convergent with the RAINS model categories, with more refined structure for some sectors with specific national characteristics that are not described in RAINS with adequate disaggregation (e.g. domestic wood combustion). The source sectors include combustion-related activities (centralized and industrial energy production plants, domestic combustion, road traffic, off-road and machinery), industrial non-combustion process plants, and various sources associated with NH_3 (agriculture), primary PM (several fugitive dust and other small non-combustion sources) and NMVOCs (solvents use, fuel evaporation). Combustion-related source sectors are described as sector-fuel combinations (e.g. industrial boilers – coal), the numbers of sectors and fuels being 101 and 15, respectively. The number of non-combustion source sectors is 53. The source sectors and fuels are presented in Appendix 1, Tables A1 and A2, respectively.

The emission sources are described with a combined bottom-up and top-down approach for large point sources and area sources, respectively. Emissions of most significant individual polluters are calculated as point sources, i.e. on an individual plant basis (bottom-up). For the model base year 2000, point sources include 117 large energy production plants (i.e. plants utilizing boilers with thermal capacity exceeding 50 MW_{th} with annual operating hours above 2000 h a^{-1}), and 88 industrial process plants (i.e. plants with emissions >20 Mg(TSP, SO_2 or NO_x) a^{-1}). Plant-specific characteristics, e.g. plant capacity, emission control equipment, emission factors and physical location information are applied on an individual basis. This enables emission estimates with good precision both spatially

Table 2. The documentation of different FRES model versions in the papers of this study

Paper	FRES version	Extent of model documentation
I	1.0 [a]	Descriptive model documentation
II	1.0 [a]	PM emission calculation and data sources of stationary combustion sources
III	1.0 [a]	Cost calculation and data sources of emission control technologies for SO_2 and NO_x
IV	2.1 [c]	Emission and emission control cost calculation and data sources for primary $PM_{2.5}$; General emission and cost calculation formulae
V	2.1 [c]	Primary $PM_{2.5}$ emission calculation and data sources of road traffic and domestic wood combustion
VI	2.0 [b]	Descriptive model documentation

[a)] SO_2, NO_x, NH_3 and NMVOCs calculation for point and area sources, primary PM (TSP, PM_{10}, $PM_{2.5}$) for area sources (area emissions spatially at municipality level)
[b)] SO_2, NO_x, NH_3, NMVOCs and primary PM (TSP, PM_{10}, $PM_{2.5}$, PM_1, $PM_{0.1}$) calculation for point and area sources (area emissions spatially in 1 × 1 km^2 grid)
[c)] As version 2.0 with updated emission calculation for domestic wood combustion

[1)] The data for $PM_{0.1}$ and PM chemical species have been used hitherto mainly for atmospheric modeling input purposes; therefore they are not presented in this study.

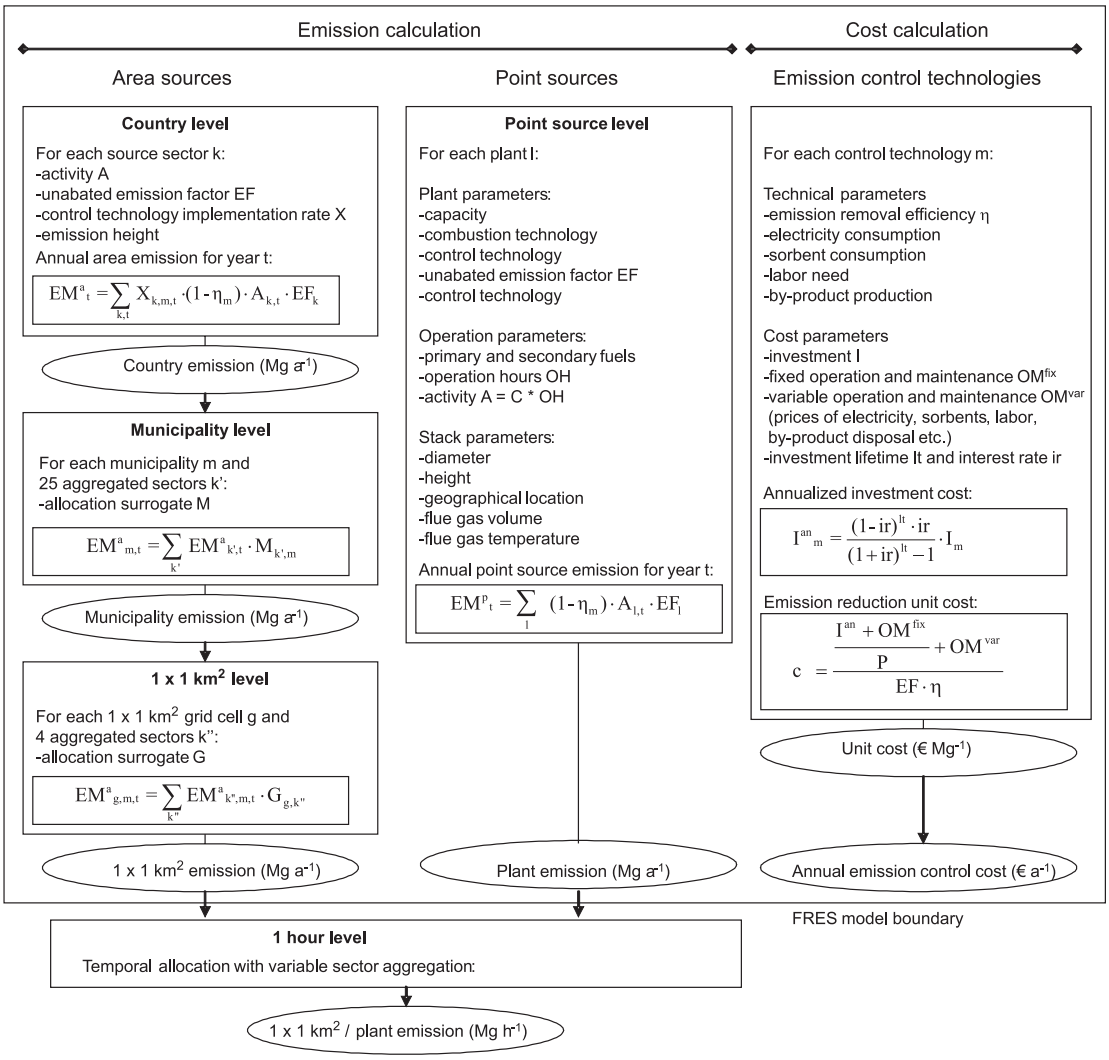

Figure 5. Flowchart of the FRES model emission and emission control cost calculation. Temporal emission allocation is performed outside the FRES model.

and in the terms of emission quantities, which is especially important in the applications of air pollution impact assessment.

Area sources have such high numbers of emitting units or plants and relatively low emissions per one unit that they are practical to describe, rather than as individual emitting units, as a group of units within an area source sector (top-down). The emission factors of an area source sector represent the averages of the corresponding individual emitting units. Emission locations are described as spatial distributions (see Section 3.1.5).

3.1.2 Activity data

Activity data describes the magnitude of activity of an emission source or source sector in a given time frame. In the FRES model the activity unit for combustion processes is annual primary energy use (e.g. PJ a^{-1}) and for industrial non-combustion processes annual production or raw material use (e.g. Mg a^{-1}). Other activity units include e.g. animal numbers and manure application for NH_3 emissions from agriculture and driven vehicle km for non-exhaust primary PM from road traffic.

The model base year 2000 activity rates of source sectors are primarily based on statistics information (e.g. Statistics Finland 2008). For some sources the sector division in FRES is more refined than that which is available in the statistics, and the activity disaggregation is carried out using other information (e.g. questionnaire studies for domestic combustion and vehicle fleet information for traffic, see paper V). Point source specific activities are determined using the parameters plant capacity and annual operation hours.

For emission scenario assessment, the estimates of future activities are needed at FRES sector aggregation. In practice, this means estimates of the use of different fuels in different sectors, the levels of industrial production etc., resulting from forecast economic developments and other influential factors, e.g. climate policies. Such future activity level estimates, denoted as activity pathways, can be produced e.g. using economy and energy system models. The FRES model studies have utilized modeling results from the Finnish MARKAL/TIMES modeling framework (Lehtilä et al. 2005) used in the planning of Finnish Climate Strategy in 2001 (Hildén et al.2001) and the Climate Strategy revision of 2005 (Hildén et al.2005). Two activity pathways of the 2001 Climate Strategy are considered in this study (Papers I, II and IV): (1) Business-as-usual (BAU), assuming no climate actions; and (2) Kyoto-Nuclear (K-Nu), a pathway compliant with the agreed EU-burden sharing including the introduction of one new nuclear power unit. The 2005 Climate Strategy pathway With-Additional-Measures (WAM) is a national activity pathway in the RAINS model, and thus a basis for the Finnish S2 pathway in Paper VI. Key assumptions of these Climate Strategy pathways are summarized in Table 3. The other activity pathways in Paper VI have been developed with the computable general equilibrium (CGE) model for Global Responses to Anthropogenic Changes in the Environment (GRACE) (Aaheim and Rive 2005).

Table 3. Key assumptions and GHG mitigation instruments in the Finnish Climate Strategy activity pathways considered in this study

Activity pathway	Activity background assumptions		GHG emission target	Main primary energy carriers for meeting increasing energy demand	GHG mitigation instruments			
	Economic growth (% a^{-1} (GDP))	Agriculture, growth in cattle numbers (% a^{-1})			Energy production and industry	Traffic sector	Domestic sector	Agriculture and forestry
Business-as-usual (2001 Strategy)[1)]	2.4	-1	No reductions	Coal, natural gas	-	-	-	-
Kyoto-Nuclear (2001 Strategy)[1)]	2.4	-1	Kyoto target for Finland[3)]	Nuclear, wood, natural gas	Energy saving, renewables	Economic instruments, logistics planning, information	Building regulations, urban planning	Emphasis on biofuel production
With-Additional-Measures (2005 Strategy)[2)]	2.3	-1	Kyoto target for Finland[3)]	Nuclear, wood, natural gas	Energy saving, renewables	Economic instruments, logistics planning, information, biofuels development	Building regulations, urban planning	Emphasis on biofuel production

[1)] Hildén et al. 2001; [2)] Hildén et al. 2005; [3)] Stabilization of GHG emissions at 1990 level

3.1.3 Emission factors

The FRES model applies the concept of unabated emission factors, which describes the emissions per activity unit without the influence of emission control technologies. The unabated emission factors represent the average within a source sector or plant determined for the model base year. Emission model base year is the period of time into which the model calculation is "anchored". This means in practice that the emission factors must be determined so that they represent the average situation of that time. It is practical if base year is a relatively recent past year, so that it represents the current or state-of-the-art situation as well as possible. Furthermore, years that are used as bases in other models are useful e.g. because of emission comparisons. The year 2000 has been selected as the FRES model base year because it is also applied as a base year in the RAINS model.

In the FRES model the base year emission factors of energy production and industrial processes for point and area sources are determined based on the analysis of 1995 – 2001 data sets from a plant-basis register of air pollution permits of the Finnish environment administration VAHTI (Korkia-Aho et al. 1995). For traffic exhaust emissions, country-specific information of the RAINS model (Klimont et al. 2002) have been used. Other area source emission factors are mainly based on Finnish and international literature. The emission factor data sources are reported in detail in papers II to V and in other literature (Karvosenoja and Johansson 2003, Karvosenoja et al. 2002).

Emission factors in the FRES model are assumed to be constant over time. Changes in emission factors are thus to be described by changes in the use of emission control technologies. Emission factor changes due to e.g. modernization of combustion appliance stock can be described by corresponding source sector disaggregation and relative changes in activity levels.

3.1.4 Emission control technologies and costs

The FRES model describes removal efficiencies and costs of emission control technologies. The technologies include e.g. end-of-pipe and process modification measures of energy production and industry sources, technologies applied in traffic vehicles and manipulations of fuel qualities. A general presentation of various control technologies is given in Chapter 1.3. The parameters and data sources are presented in Papers II and IV for primary PM and in Paper III for SO_2 and NO_x.

The current and future use of emission control technologies is to a large extent defined by the requirements of the environmental legislation. Nowadays different EU directives define emission limit values for different emission sources, either directly or by defining boundary conditions for national legislation. Major emission legislations include the Large Combustion Plants directive (LCP, EC 2001b) that sets limit values for SO_2, NO_x and primary PM (TSP) emission factors for combustion plants larger than 50 MW_{th} (thermal capacity), so-called EURO standards (e.g. EC 1998) that give increasingly tightening emission limits for new traffic vehicles, and NMVOCs directives (EC 1999b, 1994) for solvents and fuel handling practices to reduce NMVOCs emissions.

The use of emission reduction technologies may have technical or economical limitations in certain sectors. It is of great importance to be able to identify these limitations. The estimates of control technology applicability are needed e.g. in order to assess the feasibility of adopted or proposed emission legislations, or to estimate the magnitude of future emission reduction potentials. Technical applicability is to be assessed by technical experts. Economical applicability can be assessed by the means of calculated emission control costs.

Emission control costs are calculated from investment and operational costs of control equipment or process changes (Figure 5). In order to rank the technologies in terms of cost-efficiencies, unit costs (cost of the reduction of one unit of emission) and marginal costs (cost of the reduction of the last unit of emission) are calculated. For a more detailed presentation of cost calculation, see Papers III and IV.

3.1.5 Spatial and temporal emission allocation

In order to assess the impacts of air pollution and to provide information in an appropriate format to atmospheric models, it is important to know not only the quantity but also the physical location and temporal variation of emission release. Therefore, the emissions must be resolved in space and time, i.e. allocated to certain grid and temporal patterns. The spatial aspect is particularly important for emissions that may cause considerable impacts relatively near the source, e.g. impacts on human populations from sources with low emission altitude.

The spatial allocation for point sources implies simply the association of the geographical location and height of the stack with the corresponding grid cell and vertical layer of the atmospheric model, respectively. Area emissions, by contrast, must be spatially allocated using weighting factors, i.e. surrogates (Winiwarter and Schimak 2005). The choice of surrogate parameters for different source sectors depends on the availability of data that would represent the emission distribution in a given sector at the desired spatial resolution as well as possible. Typical surrogates include e.g. agricultural field areas or animal numbers for agricultural emissions, or vehicle counts or road networks for traffic emissions (Monforti and Pederzoli 2005). For anthropogenic combustion-related sources the most commonly used surrogate is population distribution (Wilson et al. 2006), which is often accurately available in statistics and Geographic Information Systems (GIS) at high resolution. However, for some combustion sources, e.g. domestic combustion, population distribution might not represent the emission distribution because of the different prevalence of the activity e.g. between urban and rural areas (e.g. Butler et al. 2008).

In the FRES model, point source emissions are given with stack parameters information from the VAHTI register. Area emissions are spatially allocated at two different steps: First, country level emissions are allocated to 448 municipalities. Second, these municipality level emissions are further allocated to a 1 × 1 km^2 grid resolution (Figure 5). This two-step approach is applied because municipality level statistical data is available relatively widely, whereas representative 1 × 1 km^2 mapped GIS data can be used only for a limited number of source sectors. A similar two-step allocation procedure was used by Dalvi et al. (2006) for a CO emission inventory in India.

The municipality and 1 km level allocations in FRES take place with aggregation to 25 and 4 source sectors, respectively. The sectors and GIS data of the 1 km allocation include (1) domestic wood combustion (floor areas of wood-heated buildings), (2) road traffic (vehicle counts), (3) agriculture (agricultural field areas) and (4) other area sources (population). Spatial distributions of different pollutants and sectors in the year 2000 are presented as maps in Figures 8 and 9. More complete descriptions of the spatial allocation and data sources are available in Karvosenoja et al. (2005).

The disaggregation of emissions in time is carried out using typical temporal patterns for different source sectors. These might include different patterns for monthly, daily and hourly variations (see the example in Figure 6). For FRES this is not an integral part of the model, but is done externally, i.e. FRES model output (i.e. spatially resolved emission fields) are temporally disaggregated externally to the FRES model before using as atmospheric model input (see Figure 5). The level of temporal disaggregation depends on the need for specific atmospheric modeling. Typically it is made down to 1 hour resolution (Cuvelier et al. 2007).

3.2 Emission dispersion and deposition estimates

The emissions of the FRES model have been applied together with atmospheric dispersion and deposition estimates in order to assess environmental impacts caused by air pollution emissions: (1) acidification and tropospheric ozone impacts (Paper I) and (2) population exposure to primary $PM_{2.5}$ (Paper VI). The assessment of methods to estimate dispersion, deposition or impacts is not the aim of this

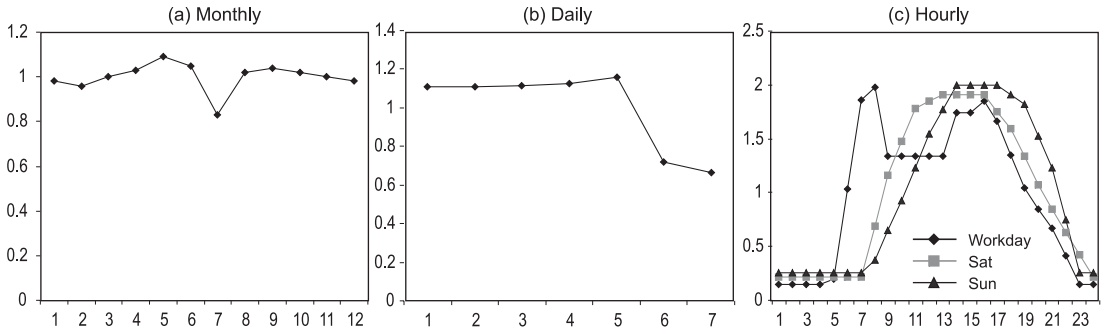

Figure 6. Temporal patterns of traffic emissions for (a) monthly, (b) daily and (c) hourly relative variation. Data from Karasmaa et al. (2003).

study. Therefore, the methods are presented only shortly in the following.

In 2000-2001 the FRES model was used in the environmental impact assessment (EIA) of the Finnish Climate Strategy (Hildén et al.2001, Paper I). The focus of the study was primarily on acidification of forest soils and lakes and ground-level ozone effects on vegetation and human health. The FRES emissions of SO_2, NO_x, NH_3 and NMVOCs described at municipality and point source level were used as input to regional acidifying deposition (Kangas and Syri 2002) and ozone formation models (Schöpp et al. 1999, Lindfors et al. 1999). LRT acidic deposition was estimated on the basis of the source-receptor matrices of the EMEP model (EMEP/MSC-W 1998). The resulting acidic depositions and ozone concentrations were combined with the data of ecosystem critical loads and critical ozone level guidelines, respectively.

The climate policy analysis in Paper VI applied a computable general equilibrium (CGE) model GRACE (Aaheim and Rive 2005) to study different policy options and to calculate activity pathways in different European countries. As a part of the analysis, a case study on Finnish primary $PM_{2.5}$ emissions and their effects on population exposure was carried out with FRES emissions at municipality and point source levels and with source-receptor matrices for primary PM in different size fractions ($PM_{0.1}$-PM_1-$PM_{2.5}$) that have been developed based on the Lagrangian SILAM dispersion modeling (Sofiev et al. 2006).

4. FRES model applications

The results of the use of different types of FRES model data are presented in this section. These are classified to the following sub-sections: (1) emissions and emission sources in the model base year 2000, (2) emission estimates of future activity pathways, (3) assessment of emission abatement potential and costs in the future, (4) the use of emissions with dispersion and impact assessment, and (5) verification and uncertainty analysis of the model.

4.1 Main emission sources in Finland - Base year emissions

Analysis of base year emissions brings valuable information about the characteristics of the anthropogenic system that produces the emissions; what are the main contributing sectors for different pollutants, what is the status of emission controls and how are the emissions geographically located. Furthermore, base year emissions can be compared with other, preferably more detailed, emission estimates, and thus used as model verification (see Section 4.5.1).

Different emission sources have different characteristics in many terms. Many of the sources, especially combustion, cause multiple emissions in various contributions. The size and number of emission sources vary greatly in different source categories, from individual large power plants and industrial processes to high numbers of small domestic heating

installations or personal vehicles. Large sources are often characterized by, on the one hand, relatively large emission quantities released from one point and, on the other hand, high emission altitudes of flue gas stacks. By contrast, diverse sources typically cause less emissions per one installation, and emit from low altitudes. Furthermore, emissions are released to the atmosphere in different geographical locations in urban, rural and industrial environments. These characteristics are important particularly when assessing the impacts of emissions; how the emissions disperse and how the emissions and consequent concentrations locate in relation to impact receptors. Therefore it is important to know, not only the emission quantities from different sources, but also the physical location or distribution of emission release points.

Country total emissions in the main emission sources in the base year 2000 are presented in Figure 7 and Table 4. Comparison to emissions of the Finnish CLRTAP inventory and RAINS model (Table 3) is discussed in Section 4.5.1. Figure 8 shows the spatial distribution of the total emissions of $PM_{2.5}$, SO_2, NO_x, NH_3 and NMVOCs presented in a 10 × 10 km^2 grid. Figure 9 presents area emissions of $PM_{2.5}$ (for agriculture NH_3) from different sectors in a 1 × 1 km^2 grid for south-western Finland. For comparison, the total emissions of $PM_{2.5}$, SO_2 and NO_x of the Finnish CLRTAP inventory in the EMEP 50 × 50 km^2 grid are presented in Figure 4 (Section 1.5).

Large point sources were the main producers of SO_2 emissions, with 64% contribution of the total SO_2 emissions. NO_x emissions from point sources were also considerable, although with lower relative contribution (28% of the total NO_x emissions). The predominance of point sources (or the minor importance of area sources) for SO_2 can also be seen from the spatial emission distribution. Sulfur emissions (Figure 7b) occur only to a minor extent outside individual emissions points, whereas primary $PM_{2.5}$ (Figure 8a), NO_x (8c) and NMVOCs (8e) emissions are more evenly distributed throughout southern and central Finland, weighted to population centers and along main highways.

Of the different point source types, coal power plants are the biggest contributor, 22 and 8.5% of the total SO_2 and NO_x, respectively. The coal power plant stock is relatively old, commissioned mainly before 1987, and therefore the emission limit values applied by LCP directive are not as stringent as for newer plants (Paper I). Therefore, the use of end-of-pipe equipment is not required for NO_x controls. Of the industrial processes black liquor recovery and other processes in pulp and paper industries cause the greatest emissions because of extensive activity levels (7.1 million tons chemical pulp in 2000). Black liquor recovery boilers are also a considerable source for $PM_{2.5}$ emissions, although high standard emissions controls are utilized (Paper IV).

Domestic wood combustion causes considerable fine primary PM emissions, with 25 and 34% contribution to the total $PM_{2.5}$ and PM_1, respectively. Domestic wood combustion also contributes considerably to NMVOCs emissions (12% of total). Wood is used in the domestic sector for both primary and supplementary heating purposes. Primary heating occurs mainly in kW size-range heating boilers. Some of the boilers have relatively high emission factors, especially when inadequately operated because of e.g. lack of a proper heat storage tank or poor fuel quality (Paper V). Therefore, even individual high-emitting boilers may affect local air quality considerably.

Wood is also combusted in different types of domestic stoves, e.g. masonry heaters and sauna stoves, often as supplementary heating in electrically heated houses. Supplementary heating is typically used only occasionally. However, it takes place in a high number of devices and, to a considerable extent, also in relatively densely populated areas, thus having air quality effects that concern large populations. The domestic wood combustion emissions occuring in the vicinity of urban areas presented in Figure 9b are predominately those caused by supplementary heating. For more details about domestic wood combustion emissions, see Papers IV and V.

Traffic sources are the main contributors of NO_x and NMVOCs emissions, with 58 and 54% contribution to the Finnish totals, respectively. Traffic also causes large primary PM emissions as both exhaust and non-exhaust emissions. Traffic vehicle exhaust, originating from on- and off-road vehicles and machinery, causes 26% of the total primary $PM_{2.5}$. Non-exhaust emissions of on-road vehicles, comprising wear products of tyres, brakes and road surface, as well as the suspension of dust deposited on roads, are a minor contributor to $PM_{2.5}$ (5.1% of the total), but a more important contributor to coarse particles (17% contribution to the total PM_{10}). Spatially, road traffic emissions occur mainly near population clusters and along main highways in south-western Finland (Figure 9a).

Non-combustion based area emissions are important contributors for NH_3, NMVOCs and coarse PM. Agriculture, i.e. emissions from livestock manure and fertilizers, is the predominant source of NH_3. NMVOCs emissions originate from a variety of different sources, e.g. the use of paints and solvents, fuel distribution and small industrial sources. Fugitive dust sources, e.g. agriculture, peat production, construction activities and material handling and storage, cause considerable PM emissions in the form of coarse particles larger than $PM_{2.5}$, but are of minor importance in finer size fractions.

Table 4. Base year 2000 emissions from main source sectors in Finland and comparison to Finnish emission inventory reported to CLRTAP and the RAINS model. Unit Gg a^{-1}.

Source sector	TSP	PM_{10}	$PM_{2.5}$	PM_1	SO_2	NO_x	NH_3	NMVOCs	Fuel primary energy (PJ a^{-1})
Point sources									
Power plants and industrial combustion (>50 MW_{th})	2.6	2.2	0.9	0.4	26.6	36.2	0	0	318
Industrial processes	10.5	7.7	5.8	2.9	22.2	22.4	1.0	7.1	-
TOTAL point sources	***13.1***	***9.9***	***6.7***	***3.3***	***48.8***	***58.6***	***1.0***	***7.1***	***318***
Area sources									
Power plants and industrial combustion (<50 MW_{th})	4.8	3.7	2.5	1.8	19.2	18.7	0	1.9	176
Domestic combustion	9.4	9.0	8.6	8.4	4.2	8.0	0	20.0	107
Road traffic	20.2	12.0	5.7	4.5	0.2	79.0	0	47.2	148
Off-road and machinery	4.2	4.0	3.8	3.4	3.8	41.5	0	34.4	47
Other non-combustion sources[1]	16.8	7.3	3.5	1.5	0.1	0.2	32.3	41.0	-
TOTAL area sources	***55.4***	***36.0***	***24.1***	***19.6***	***27.6***	***147.3***	***32.3***	***144.6***	***478***
TOTAL	**68.3**	**45.8**	**30.8**	**23.0**	**76.4**	**206.0**	**33.3**	**151.7**	**797**
Inventory to CLRTAP	72[a]	47[a]	37[a]	-	76	210	33	160	
RAINS[b]	61[c]	41[c]	31[c]	31[c,d]	76	212	35	160	

[1)] Small industrial processes, various fugitive dust and other small primary PM-related sources, agricultural activities (NH_3 and PM) and solvents use and other NMVOCs-related sources

[a)] CLRTAP inventory (Finnish environment institute 2008) emissions calculated with old emission factor estimates on domestic wood combustion (see Table 5 in Paper V)

[b)] Emissions extracted from RAINS online Nov. 2007 (www.iiasa.ac.at/gains)

[c)] do not include traffic resuspension emissions

[d)] PM_1 emissions extracted from Kupiainen and Klimont (2004), not directly comparable with other RAINS PM emissions

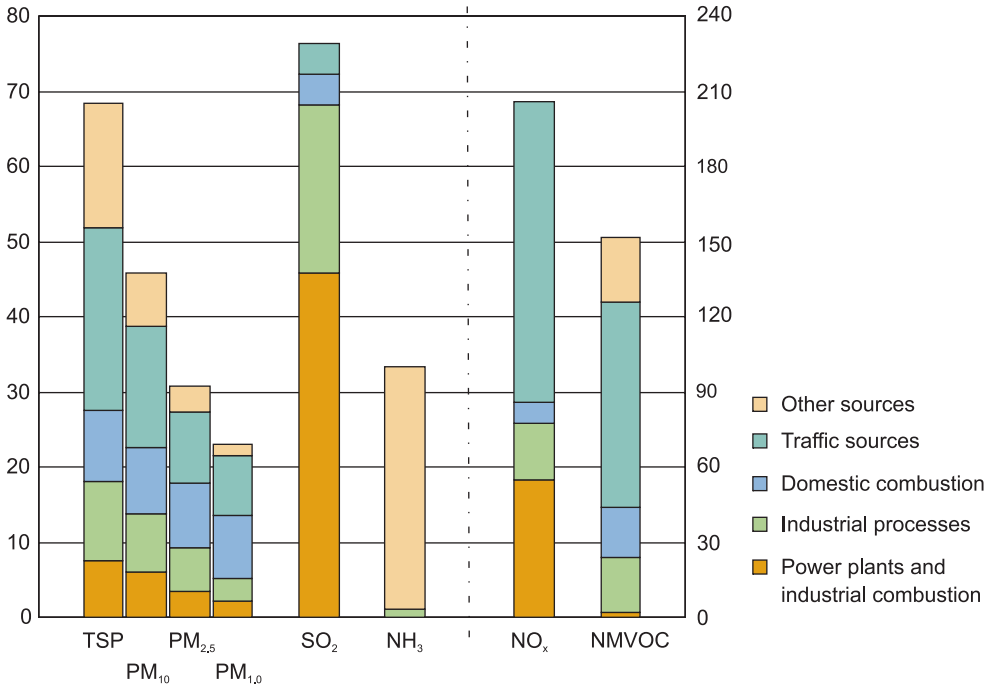

Figure 7. Base year 2000 emissions from main source sectors in Finland. The left-hand axis refers to six columns on the left (TSP, PM_{10}, $PM_{2.5}$, PM_{1}, SO_{2}, NH_{3}) and the right-hand axis refers to two columns on the right (NO_{x}, NMVOCs). Unit Gg a^{-1}.

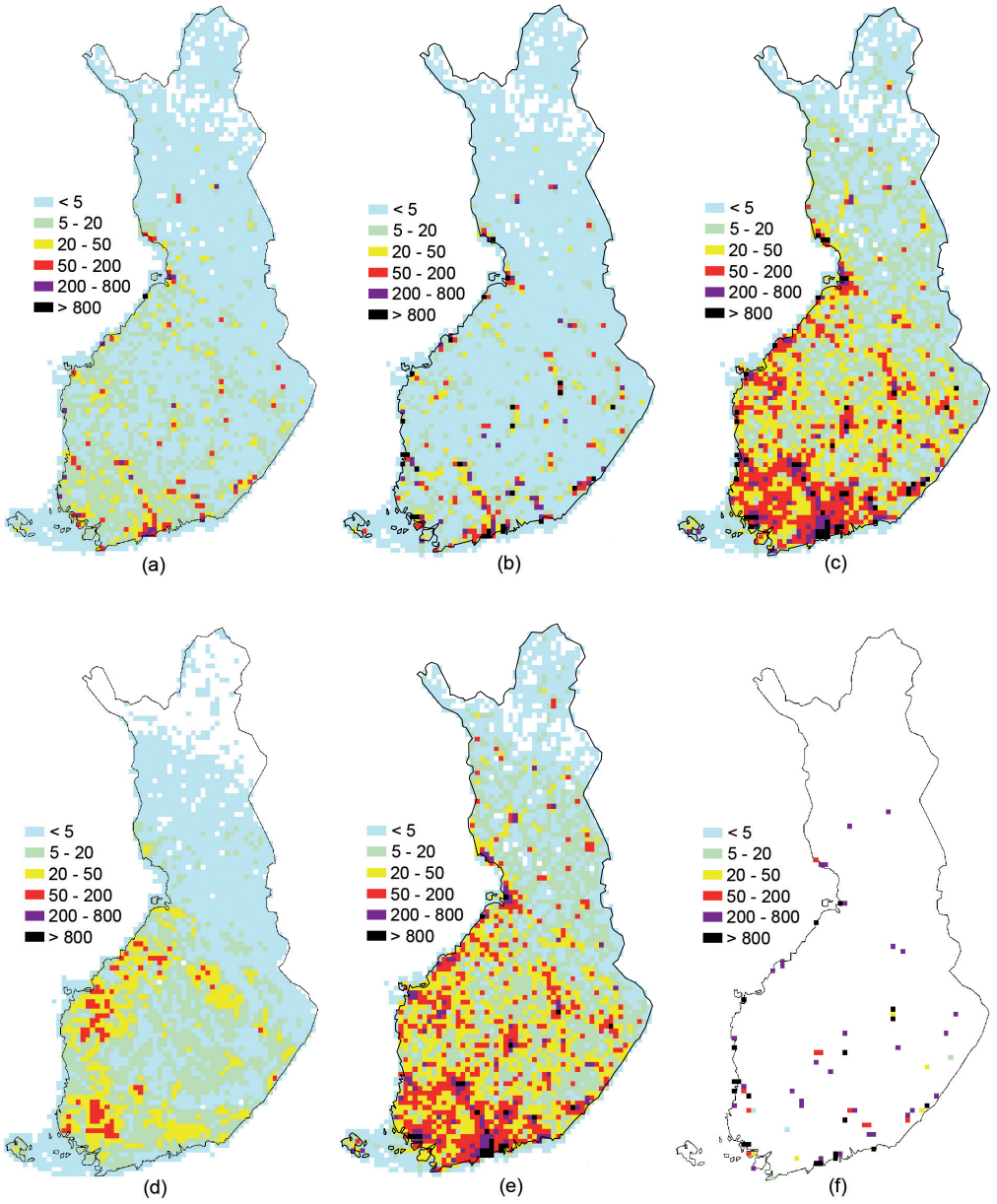

Figure 8. Total emissions of (a) $PM_{2.5}$, (b) SO_2, (c) NO_x, (d) NH_3 and (e) NMVOCs in 2000 presented in 10 × 10 km² grid. Point source emissions of SO_2 are given in (f). Unit Mg a⁻¹.

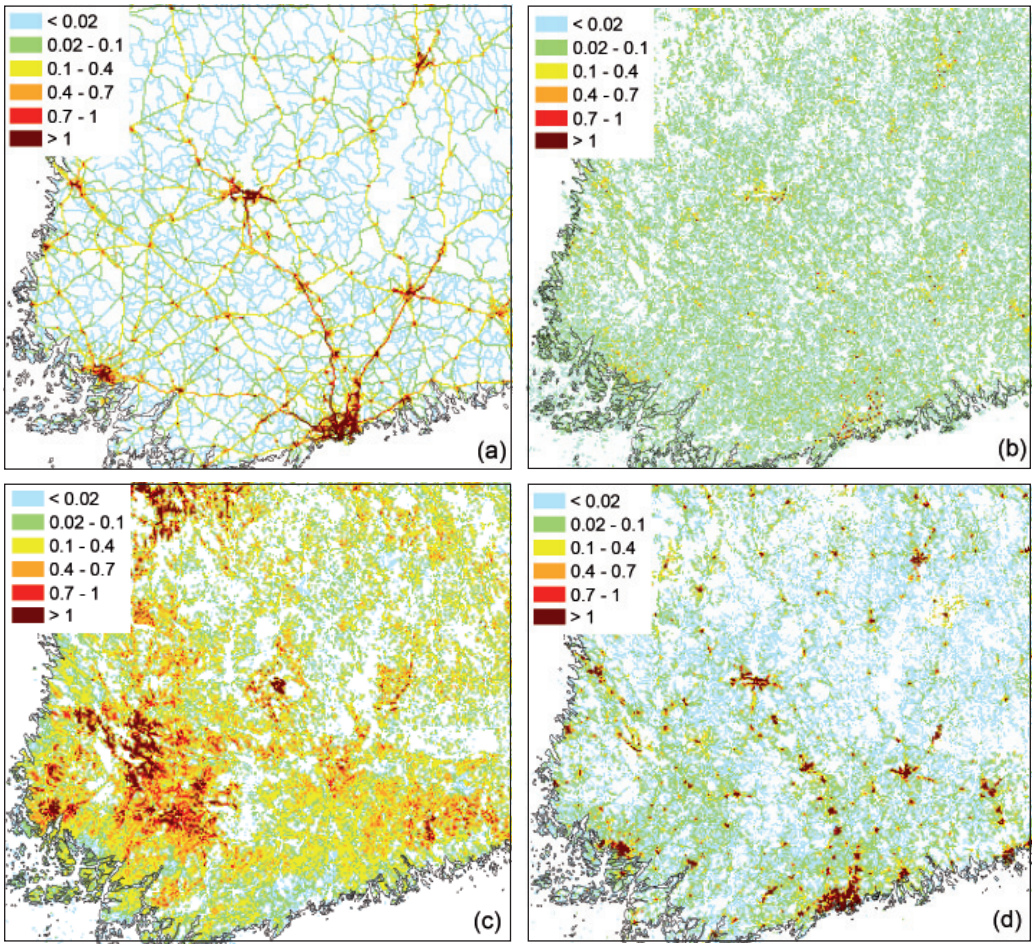

Figure 9. The sector emissions of (a) road traffic, (b) domestic wood combustion, (c) agriculture, and (d) other area sources in 2000 presented in a 1 × 1 km² grid for south-western Finland. Unit Mg($PM_{2.5}$) a^{-1}, except (c), Mg(NH_3) a^{-1}.

4.2 Emissions of future activity pathways

4.2.1 Emission development to 2020

Future air pollution emissions (Table 5, Figure 10) are based on different activity pathways and the assumptions of the use of emission controls following the requirements of current EU and national legislation, i.e. the so-called current legislation (CLE) scenario. In general, it can be seen that air pollution emissions will decrease in the future, although the main activity levels, i.e. primary energy use and industrial production (not presented), will increase. The emission decrease is influenced primarily by tightening emission standards in CLE.

The emission decrease is strongest in the traffic sector. The traffic exhaust emissions of primary $PM_{2.5}$, NO_x and NMVOCs will be reduced (by 74%, 60% and 76% from 2000 to 2020, respectively) due to EURO standards for new vehicles (e.g. EC 1998), and SO_2 (59%) because of increasing use of sulfur-free oils in machinery and off-road vehicles. In addition, NMVOCs emissions from fuel distribution will be considerably reduced (50%) due to the effect of NMVOCs directives (e.g. EC 1994). By contrast, primary PM traffic non-exhaust emissions will not be regulated, and they will increase along with the increase in traffic volume, i.e. by 35%. This is considerable especially for coarse PM, but in the future increasingly also for fine PM; road traffic non-exhaust based primary $PM_{2.5}$ emissions will exceed those of road traffic exhaust at around 2010, and will be 5-fold in 2020.

Emission developments in power plants and in the industrial combustion sector will be relatively stable, with slight decreases mainly between 2010 and 2020. LCP directive (EC 2001b) for new plants affect mostly the emissions of SO_2 and NO_x, although the long life time of power plants leads to emission decline in the relatively long course of time along with the renewal of the plant population. For SO_2, another important factor in addition to emission controls is the fuel used (see next section). For primary PM, in contrast to SO_2 and NO_x, LCP directive will not bring significant requirements compared to current standards of emission reductions (Paper II).

The exception in terms of emission reduction and activity level developments is ammonia; although agriculture will not be subject to significant air pollution emission reduction requirements in the future (Ministry of the Environment 2002), NH_3 emissions will decrease by 10% along with the projected decline in agricultural activities (see Table 3). The review of the IPPC directive (EC 2008) currently in progress might recommend further technical emission controls in agriculture; however, these have not been analyzed for Finland. Other studies have estimated feasible ammonia emission reduction potential at around 20 to 30% of the total emissions in Finland (Kallioniemi 2002, Cowell and ApSimon 1998).

Table 5. The emissions of primary $PM_{2.5}$, SO_2, NO_x, NH_3 and NMVOCs in 2000, 2010 and 2020 in Business-As-Usual (BAU), Kyoto-Nuclear (K-Nu) and With-Additional-Measures (WAM) activity pathways. The emissions of the official national pathway in the RAINS model, i.e. With-Additional-Measures (WAM/RAINS) are also presented. The last column on the right gives the obligations of National emission ceilings (NEC) directive of EU and Gothenburg Protocol of UNECE/CLRTAP for Finland. Unit Gg a^{-1}.

		BAU (I, II)		K-Nu (I, II, IV)		WAM (VI)		WAM/RAINS [a]		NEC/CLRTAP
	2000	2010	2020	2010	2020	2010	2020	2010	2020	2010
$PM_{2.5}$	30.8	28.7	26.1	28.2	25.8	27.4	25.7	25.9	24.0	- [b]
SO_2	76.4	92.9	76.5	74.6	59.8	66.9	59.8	66.4	58.8	110 / 116
NO_x	206.0	177.7	137.4	163.3	126.3	162.0	129.2	168.8	129.0	170 / 170
NH_3	33.3	31.1	30.4	31.1	30.4	31.1	30.4	30.5	29.9	31 / 31
NMVOCs	151.7	105.4	81.5	105.0	81.5	105.2	84.1	111.4	90.7	130 /130
Total primary energy (PJ)	1321	1513	1583	1502	1559	1544	1616	1544	1616	-

[a] Emissions extracted from RAINS online Nov. 2007 (www.iiasa.ac.at/gains)
[b] Current protocols do not include emission caps for $PM_{2.5}$

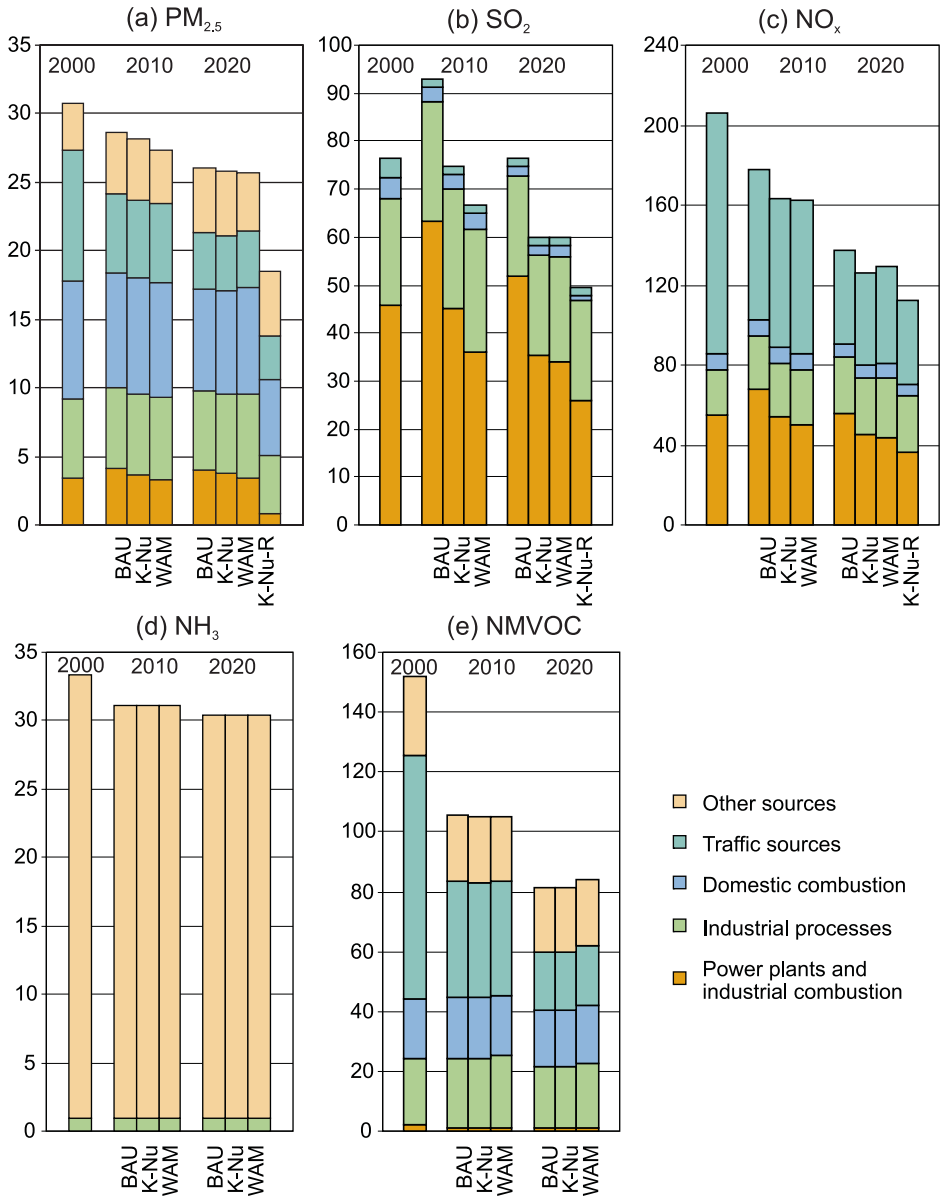

Figure 10. The emissions of (a) primary $PM_{2.5}$, (b) SO_2, (c) NO_x, (d) NH_3 and (e) NMVOCs in 2000, 2010 and 2020 in the Business-As-Usual (BAU), Kyoto-Nuclear (K-Nu) and With-Additional-Measures (WAM) activity pathways. Additional emission reduction potentials in the Kyoto-Nuclear pathway (K-Nu-R) are also presented for primary $PM_{2.5}$, SO_2 and NO_x emissions in 2020.

4.2.2 Effects of climate measures on air pollution

When comparing air pollution emissions in different activity pathways of the Climate Strategies, it can be concluded, in general, that measures to reduce GHG emissions result in reduced air pollution emissions. The emissions of SO_2 and NO_x in the climate action pathways (Kyoto-Nuclear, With-Additional-Measures) are considerably lower than in the no-action pathway (Business-as-usual), by 20 to 28% and 8 to 9%, respectively, in 2020. For primary PM the difference is less prominent, 2 to 5% lower emissions, respectively. This is because the Climate Strategies concentrated primarily on alternatives in industrial scale energy production, and PM emissions from large plants are controlled by efficient end-of-pipe reduction measures and thus are relatively low regardless of the chosen primary energy carrier (Paper II). The measures in the studied Climate Strategies have negligible effect on NH_3 and NMVOCs emissions.

The main GHG emission reduction measures that influence air pollution are fuel changes in the energy production system and energy saving. Energy saving has a moderate decreasing effect on air pollution because of decrease in activity levels. More important, however, are fuel changes, i.e. the switch from carbon-intensive fuels (e.g. coals and oils) to low-emission (e.g. natural gas) or CO_2-neutral energy carriers (e.g. renewable and nuclear energy). The most important co-benefit mechanisms behind these are:

- Energy production by nuclear, wind and hydro power does not cause direct air pollution emissions
- Natural gas and wood fuels contain no or very low fractions of sulfur, whereas coals and oils have relatively high sulfur contents
- PM emissions from gaseous fuels are negligible, in contrast to solid fuels and heavy fuel oils
- The shift away from coal combustion requires accelerated removal of the relatively old population of existing large coal plants, which means tighter emission limits for substituting capacity. This mechanism affects especially 2010 emissions (Paper I). The effects of accelerated renewal are smaller in 2020 because the old coal boiler population is estimated to be mainly removed by 2020.

In addition to the co-benefits described above, there are also some potential trade-offs between climate measures and air pollution. The most important such trade-off for Finland is the potential increase in domestic wood combustion as a substitute for oil and electricity heating. Wood is a renewable energy source and thus a climate friendly fuel, although many domestic wood combustion technologies entail high primary PM emissions. When compared to oil or electricity heating, wood heating would lead to approximately 10- (pellet boiler) to 20-fold (modern masonry heater) increases in primary $PM_{2.5}$ emissions. The Climate Strategies pathways did not concentrate on domestic heating options, and therefore this trade-off cannot be seen directly from the results in Table 5. Instead, the importance of the domestic wood combustion sector for PM emissions has been studied and the potential effects of climate measures qualitatively discussed in papers II, IV, V and VI.

4.3 Future emissions reduction

The FRES model describes applied and potential emission control technologies along with their removal efficiencies and costs for different sectors. If there are technically and economically feasible control technologies for a given sector with higher efficiency than that which is already in use, an additional emission reduction potential exists. By means of the emission reduction and cost estimates, cost-efficiencies of control technologies can be assessed. Cost-efficiencies of a large group of reduction technologies can be illustrated and compared in cost curves. This section presents emission reduction potentials and cost estimates, as well as cost curve compilations carried out with the FRES model data.

4.3.1 Emission reduction potentials and costs

As stated in the previous section, air pollution emissions will decrease in the future, mainly as a result of the emission control requirements set by CLE. Further reductions in addition to the controls defined by CLE have been estimated with the FRES model. The following presents reduction potentials and costs in 2020: (1) for primary $PM_{2.5}$ based on Paper IV, and (2) for SO_2 and NO_x estimated approximately here on the basis of control technology data from Paper III. The reduction potentials are also presented in Figures 10a-c as K-Nu-R.

Paper IV presents an estimate of primary $PM_{2.5}$ emission reduction potential and associated costs in 2020 in the Kyoto-Nuclear activity pathway. Figure 11 shows the reduction potentials for different sectors grouped by marginal costs of reduction measures. The largest reduction potential, 3.0 Gg a^{-1}, is estimated in power plants and industrial combustion, both in large sources by fabric filter technologies and in smaller <50 MW_{th} boilers by ESPs and multicyclones. The marginal costs of these measures vary between 2000 and 11 000 € Mg^{-1}. Considerable cost-efficient reduction potential, 1.7 Gg a^{-1}, with marginal cost below 5000 € Mg^{-1} can be found in domestic wood combustion heating boilers by small ESPs, although the estimates carry high uncertainties in both costs and performance of the technology. Domestic combustion emissions from wood stoves could be potentially reduced by the application of low-emission appliances and operation practices, although their effect could not be quantified in this study. For industrial processes some reduction potential was identified in few individual plants by fabric filter utilization, with variable marginal costs from 600 to 13 000 € Mg^{-1}. In the traffic sector, a reduction of 0.9 Gg a^{-1} with high marginal cost (around 70 000 € Mg^{-1}) can be achieved in diesel machinery by full implementation of the most efficient EURO-standards. In total, the reduction potential in 2020 was estimated at 7.4 Gg a^{-1}, or 29% of the total primary $PM_{2.5}$ emissions.

Future emission reduction potentials of SO_2 or NO_x were not estimated in the papers of this study. Instead, approximate estimates of SO_2 and NO_x reduction potentials in 2020 will be given in the following based on emission controls data of Paper III and the activity levels of the Kyoto-nuclear pathway. Based on Karvosenoja et al. (2001), the majority of the control technologies presented in Paper III were already in use in 1995 (see Figure 12).

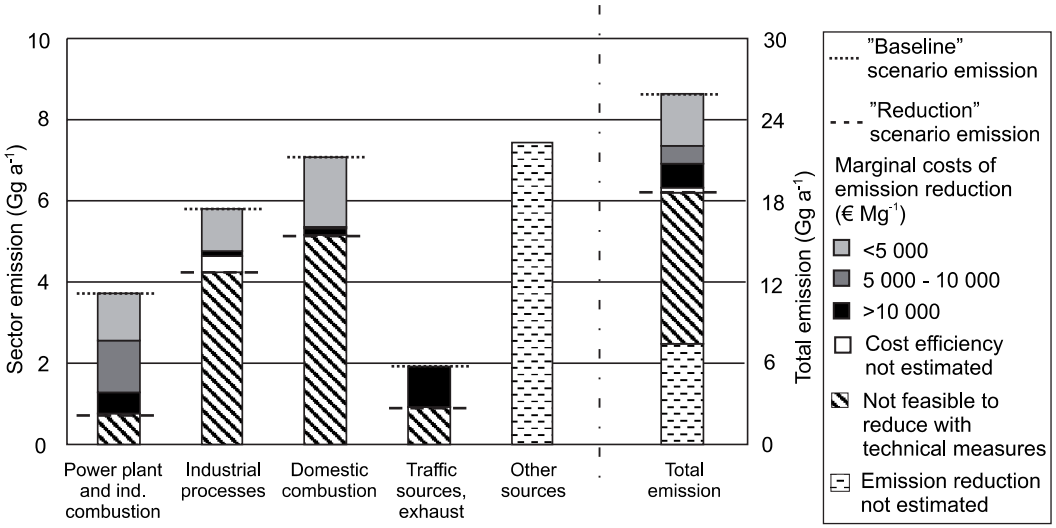

Figure 11. Primary $PM_{2.5}$ from main source sectors (the left-hand axis and first five columns) and total emissions (right-hand axis and column) in Finland in 2020. Additional emission reduction potentials and cost-efficiencies are presented in grey shades. (Paper IV)

By 2020, only a few remaining control options exist in addition to the CLE requirements. The main remaining emission reduction potentials (rp) in addition to CLE and their marginal costs (mc) include:

For SO_2:

- **Spray dry scrubbers in peat/wood co-combustion boilers.** Peat is mainly combusted in fluidized bed (FB) boilers mixed with wood. Further SO_2 reduction (rp approx. 5 Gg a^{-1}) could be achieved by the introduction of spray dry FGD equipment with relatively low mc (3000 € $Mg(SO_2)^{-1}$).
- **Spray dry scrubbers in heavy fuel oil boilers.** Heavy fuel oil is combusted mainly as a peak supply fuel with low operation hours. Therefore the introduction of end-of-pipe measures would lead to high mc (above 20 000 € $Mg(SO_2)^{-1}$). Rp less than 5 Gg a^{-1}.
- **Ultra low-sulfur fuel oil (0.001%S) in the domestic combustion sector.** The gained rp would be minor (less than 1 Gg a^{-1}), with relatively high mc (around 10 000 € $Mg(SO_2)^{-1}$).
- **Total SO_2 reduction potential** is estimated at approximately 10 Gg a^{-1}, or 17% of the total emission in 2020

For NO_x:

- **Selective non-catalytic reduction (SNCR) technologies in peat/wood co-combustion boilers.** Relatively low NO_x emission levels of FB boilers could be further reduced by SNCR with relatively low costs (rp approx. 7 Gg a^{-1}, mc approx. 1000 € $Mg(NO_x)^{-1}$).
- **Combustion modification technologies in small (<50 MW_{th}) gas, peat and wood boilers.** Small energy production units are not subject to strict emission standards. NO_x emissions could be reduced cost-efficiently by combustion modification technologies (rp 2 Gg a^{-1}, mc approx. 1000 € $Mg(NO_x)^{-1}$).
- **Full implementation of the most efficient EURO-standards in diesel machinery.** This refers to the same set of technologies as assumed with primary $PM_{2.5}$ reduction (rp approx. 5 Gg a^{-1}, mc > 10 000 € $Mg(NO_x)^{-1}$)
- **Total NO_x reduction potential** is approximately 14 Gg a^{-1}, or 11% of the total emission in 2020

In summary, air pollution emissions in 2020 will be mainly relatively efficiently controlled in large power plants, industry and in the traffic sector. The emission standards of CLE are less demanding for smaller energy production plants below 50 MW_{th} and for domestic combustion sources. These sectors have further cost-efficient potentials especially for the reduction of primary $PM_{2.5}$. Domestic wood combustion causes air quality deterioration locally in Finland, that can be detected e.g. in the form of complaints to authorities (Salonen 2004). Given the severity of human health impacts of fine PM (e.g. Pope and Dockery 2006), it would be important to further develop measures to reduce domestic wood combustion emissions.

New national emission ceilings for 2020 are being negotiated in the revision processes of the UNECE/CLRTAP Gothenburg Protocol and of the EU's NEC directive. Proposed ceilings will be derived by attaining agreed environmental targets with reduction measures which are based on European-wide least-cost optimization of the RAINS model (see Section 1.5.3). In order to ensure realistic assumptions on the emission reductions in the RAINS model, it is crucial to be able to assess these European-level model results with national estimates, e.g. the FRES model (see also Section 4.5.1).

4.3.2 Cost curves

A cost curve is an illustrative tool to compare cost-efficiencies of emission reductions in different sectors. Emission reductions achieved by different control technologies in different sectors are ranked along their marginal cost to a piece-wise linear steepening curve. Each segment of the curve represents one control technology – sector combination and x- and y-axis shifts additional emission reductions

and cost, respectively. Cost curves can be compiled in different ways depending on the definition of the group of technologies that are included in the curve:

- A "no-control" cost curve illustrates cost-efficiencies of all applicable emission controls, including both in-use technologies and remaining potential. The curve starts from an unabated emission level of certain year, i.e. a hypothetical situation in which no controls are used. In Paper III SO_2 and NO_x "no-control" cost curves for the year 1990 are presented.
- A cost curve can represent remaining emission reduction potential. In this case the starting point is the actual emission of a target year and the curve includes technologies with higher removal efficiencies than those that are already in use. Such a cost curve for primary $PM_{2.5}$ in 2020 is shown in paper IV.
- A cost curve can also illustrate cost-efficiencies of already adopted control measures. Figure 12 shows Finnish SO_2 and NO_x cost curves in 1995 for both adopted controls and remaining potential.

4.4 Use of emission models in air pollution impact assessment

FRES emission estimates have been used in the environmental impacts assessment of (1) acidification and tropospheric ozone (Paper I) and (2) population exposure to primary $PM_{2.5}$ (Paper VI). The following sections introduce the main findings of these studies from the emission modeling perspective. The main aim is to discuss the suitability of emission model characteristics in the applied study set-ups.

4.4.1 Acidification and tropospheric ozone

The impact assessment of Paper I predicted declining acidic deposition and tropospheric ozone levels in the future in all the studied activity pathways. Although acidification in Finland is mainly caused by LRT, the effects of Finnish emission reductions could also be detected. Lower emissions of SO_2 and NO_x by 20 – 32% and 8 – 12%, respectively, in climate action pathways compared with the Business-as-usual (BAU) pathway, resulted in 6 – 8% and 2 – 3% reductions in ecosystems threatened by acidification and harmful ozone levels, respectively, in southern Finland.

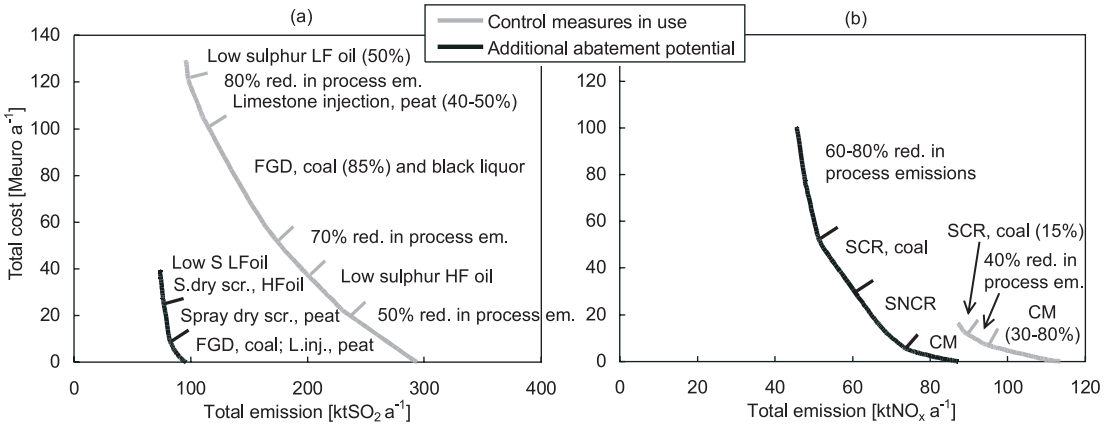

Figure 12. Finnish 1995 cost curves for (a) SO_2 and (b) NO_x for adopted controls (grey curve) and remaining potential (black curve) (from Karvosenoja et al. 2001) (FGD = flue gas desulfurization, L.inj. = limestone injection, LF oil = light fuel oil, HF oil = heavy fuel oil, CM = combustion modifications, SCR = selective catalytic reduction, SNCR = selective non-catalytic reduction)

In the case of acidification, the impact receptors (i.e. forest soils and lakes) are situated relatively far from the highest emission volumes. Finnish emission sources contributing to acidification are, to considerable degree, those of energy production and industry described as point sources with actual stack location and height parameters in the FRES model (approx. 70 and 43% of SO_2 and NO_x emissions in 2020, respectively). Therefore, although area emissions in the study were spatially relatively coarsely described (at the municipality level, i.e. mostly with 10 – 30 km resolution), the emission description in FRES could be deemed reasonable for the assessment of acidification, considering the 14 km grid resolution of the applied deposition model.

For the human exposure to ozone, in contrast to acidification, the impact receptors are situated to a large extent in the vicinity of ozone precursor emission sources (43% of NO_x and 73% of NMVOCs are low-altitude area emissions in 2020). Locally, ozone formation (and destruction) time scales can range within minutes to hours (destruction mechanisms being more rapid) (Seinfeld and Pandis1998). Therefore, spatial scales within kilometers to tens of kilometers are important in ozone formation assessment. Syri et al. (2001) proposed that urban NO_x levels are a significant factor explaining differences between urban and background ozone concentrations, and that the use of spatially refined emission data with corresponding ozone formation estimates would enhance the 50 km resolution European-scale estimates. Therefore, the urban ozone exposure assessment of Paper I would have benefited from a higher emission resolution (e.g. of 1 to 10 km), assuming corresponding ozone model resolution.

4.4.2 Population exposure to primary particulate matter

Paper VI presents a Finnish case study concerning population exposure to Finnish primary $PM_{2.5}$ emissions in different climate policy activity pathways. The effect of the studied climate policy options is not analyzed here because of the relatively coarse energy system description in the applied GRACE model and the resultant imperfect representativity of the effects of policy measures on sector level activity data in a single country. However, the results from the use of PM source-receptor matrices demonstrate the variable emission – population exposure relationships of different emission sources. As an example, decrease of primary $PM_{2.5}$ emissions from 2000 to 2020 (16%) leads to relatively higher benefit as decreased population exposure, 22% (Figure 4a in Paper VI). This is because of the fact that emissions decrease occurs mainly in traffic emissions that have a considerably (48%) higher population exposure effect than average emissions (Figure 13). Figure 14 shows corresponding $PM_{2.5}$ concentrations caused by primary emissions in Finland in 2000 and 2020.

The results highlight the importance of both geographical and vertical representation of emissions in the assessment of human PM exposure. The exposure effect of the same amount of predominately urban emissions is considerably higher than that of predominately non-urban emissions (e.g. 63% higher exposure from traffic than from domestic wood combustion). Correspondingly, the effects of low-altitude emissions are higher than those of high-stack sources, i.e. large power plants and industrial combustion (PP&IN (>50MW)) and industrial processes (PROC).

The FRES emission resolution in Paper VI (i.e. municipality level, approx. 10 – 30 km) can be deemed reasonable for use with the applied source-receptor matrices at 12 km grid resolution. However, in order to assess urban population exposure effects, e.g. in different parts of a city, or the differences between central and suburban areas, the 1 km gridded emissions of the FRES model should be utilized with corresponding high resolution dispersion estimates.

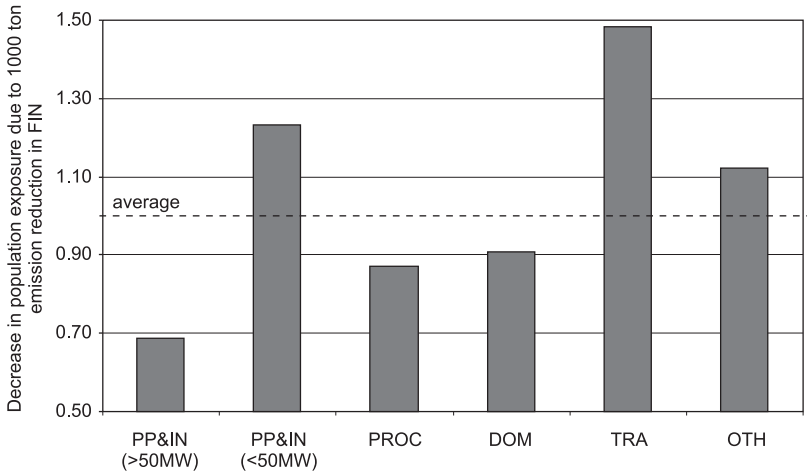

Figure 13. The relative emission – population exposure relationships for one unit of $PM_{2.5}$ emission reduction (here as an example 1000 tons) performed in different sectors (PP&IN = power plants and industrial combustion, PROC = industrial processes, DOM = domestic wood combustion, TRA = road traffic, OTH = other emission sources) and as an average, i.e. if the emission reduction were to take place evenly in all the sectors (=1.0, dotted line) (Paper VI)

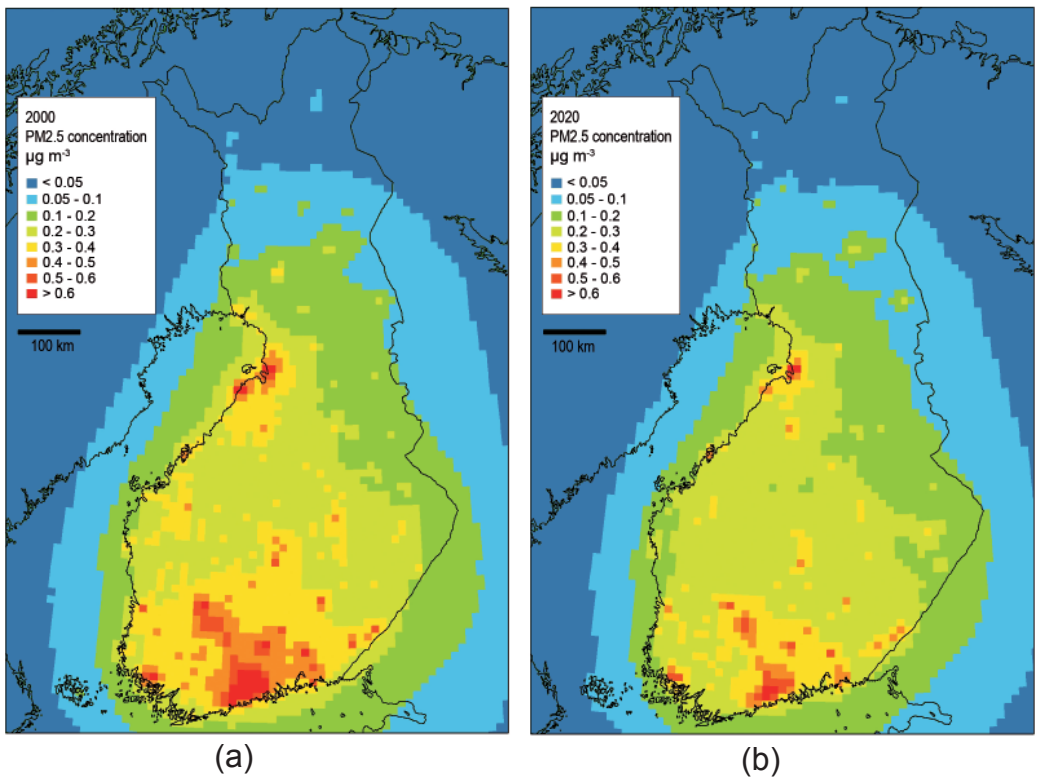

Figure 14. $PM_{2.5}$ concentrations caused by primary $PM_{2.5}$ emissions in Finland in (a) 2000 and (b) 2020. The estimates are based on the FRES model emission data and source-receptor dispersion matrices applied in Paper VI.

4.5 Model verification

Emission model estimates should always be verified. The importance of the correctness of emission estimates has been highlighted especially in the context of air quality modeling (e.g. Butler et al. 2008, Harrison et al. 2008, Miller et al. 2006, Whyatt et al. 2007). Special emphasis has been placed on the comprehensiveness of emission sources, accuracy of emission factors and spatial allocation. Lindley et al. (2000) listed different techniques for emission model verification:

- Model and data documentation
- Quality checks by the application of the model
- Comparison of alternative estimates
- Uncertainty estimates
- Ground truth verification

Model and data documentation ensures the transparency of the applied methods and results. Basic model use shows how the model output is applicable in different types of end applications, e.g. as part of IAM. Comparisons between different emission estimates at the sector level provide information about the correctness of performed source aggregation and reveals possible errors (Miller et al. 2006). Uncertainty assessment quantifies the degree of confidence in input parameters in different source sectors and pinpoints the parameters that would benefit from further investigation. Ground truth verification refers to techniques that make comparisons between the emissions and some other independent but closely related data, e.g. ground-based observations (Mellios et al. 2006, Fagerli et al. 2003) or satellite instruments (Richter et al. 2005).

In this study, the first two of these techniques were covered in previous sections: the FRES model is documented in Section 3 and different model applications are presented and discussed in this section. In the following, the third and fourth techniques, i.e. comparisons of the FRES model against other emission estimates and uncertainty assessment are presented based on the papers of this study and other literature.

The fifth point, i.e. ground truth verification, is not directly covered in this thesis. In the case of the FRES model, ground truth verification could involve e.g. a comparison of the FRES – atmospheric model system with ambient measurements. Comparison of modeled concentrations or depositions using several past years' emissions and meteorology against trends in measurements can bring valuable information on the performance of the modeling system. However, a problem of such a comparison to be used solely as an emission model verification is that applied atmospheric and meteorology models, as well as the estimates of LRT contributions, have their own additional uncertainties and their isolation from national emission uncertainties would be very difficult (Miller et al. 2006).

4.5.1 Emission comparisons

Comparisons between different emission estimates are important as model verification. In the course of model development, discussion between model developers and comparisons of models operating on different sectoral and spatial scales play an important role in the harmonization of different models. In addition, these act as a learning process about both the emission characteristics of different sources and practical model aggregation levels (Winiwarter and Schimak 2005). As a top-down model evaluation, comparison of sector emissions to concurrent bottom-up inventory data provides information about the correctness of performed source aggregation and reveals possible errors (Miller et al. 2006).

In Paper II the FRES model PM emissions are compared to national statistics and the European-wide CEPMEIP (2001) inventory. At the time of Paper II the FRES model was in an early stage of its PM calculation development (see Table 2). However, important conclusions based on the comparisons can be drawn:

- Correct estimates of emission control technology utilization rates are important especially in the case of PM. This is because of the fact that end-of-pipe controls of PM have typically very high

removal efficiencies and thus have a strong influence on the realized emission factors.
- The use of common European default emission factors, such as used in the CEPMEIP inventory, may lead to large errors in emission estimates at the national level. Instead, emission factors of an emission model should be derived, if possible, from sectorally more detailed or spatially smaller domain emission calculation systems, or directly from representative measurements of e.g. power plants. For example, country emission calculations in European-wide models should exploit information from the relevant national models, or they should be reviewed by national experts. In turn, national models should preferably base their emission factor estimates on e.g. plant-by-plant basis data aggregated to practical sector division.
- The emission source aggregation in national models should reflect specific national features. This might require disaggregation or other tailoring of the source aggregation provided in European models (e.g. as in the case of domestic wood combustion in Finland).
- Domestic wood combustion is a remarkable contributor to total PM emissions. In addition, the lack of representative national measurements and the inadequacy of the emission estimation procedure for the domestic wood combustion sector in the FRES model of that time was highlighted.

Since the time of the Paper II study, considerable FRES model development has taken place, actuated to large extent on the basis of the conclusions above:
- A point source specific bottom-up approach for large sources of PM has been incorporated into the model. It enables more flexible and transparent management of emission factor and control technology utilization data, as well as more accurate spatial presentation of emissions.
- The domestic wood combustion emission calculation procedure has been renewed to take combustion appliance specific emission characteristics into account (see Paper V). Recent Finnish measurement activities (e.g. Tissari et al. 2007) have been an essential source of emission factors. The new estimates will also be reflected in national statistics and inventory reports (e.g. Statistics Finland 2008, Finnish environment institute 2008) in the future.

The emissions of the current FRES model version are compared with the inventory emissions reported to UNECE CLRTAP (Table 4) and with the RAINS model (Tables 4 and 5). In general, the emissions show a good agreement, mainly within 5%. The few exceptions can be explained by different emissions factors, e.g. in domestic wood combustion PM emissions in the FRES and CLRTAP inventory (see above), or different emission source definition, e.g. in traffic non-exhaust PM emissions in FRES and RAINS (for both, see also Table 5 in Paper V).

The good comparability between these different models and registers is a result of the emission harmonization process taking place during the years of model developments. The harmonization has been in the form of both direct data transfer between the models and personal communication between the modelers.

An annually compiled national inventory report to CLRTAP is based on plant specific and other emission information compiled on a level as detailed as possible. The base year emission factor data of the FRES model are largely based on the same detailed data sources, only averaged over several years and, for area sources, aggregated over several plants.

The RAINS model emission calculation for Finland is also influenced, if only indirectly, by the same national data sources as the FRES model. The correctness of the RAINS emission model relies to a large extent on the bilateral consultation process between RAINS model administrators and experts from each country. In the consultations country experts assess and correct different RAINS model parameters, e.g. emission factors and control technology use, for the part of their country.

The FRES model has been the main reference in the Finnish consultation, and many RAINS emission calculation parameters have been tuned on the basis of FRES data. As the Finnish consultation process takes place as a "dialogue" between the two models, it also acts as a model comparison in a fine sectoral detail.

4.5.2 Uncertainty assessment

Emission estimates are always subject to uncertainties. Input parameters, i.e. activity and emission factor values used in emission models, are always imperfectly known. The importance of systematic uncertainty analysis for air pollution emission estimates and, on the other hand, the lack of such analyses, have been discussed (Placet et al. 2000, Miller et al. 2006, Whyatt et al. 2007). In uncertainty analysis the degree of confidence in the input parameters in different source sectors is quantified and the resulting uncertainties are defined by the implementation of e.g. Monte Carlo simulation (Romano et al. 2004).

The input parameter uncertainties contain both random uncertainty, i.e. natural heterogeneity of data within a certain source sector, and systematic uncertainty, i.e. deviation of the parameter from its real value. The former includes random uncertainty of measurements and other methods, and natural variability of a parameter. Random uncertainty is thus affected by parameter estimation methods, the nature of a source sector and the level of source aggregation. The latter is related to deficiency or lack of knowledge about parameters or emission sources. It is not exceptional that, for certain emission sources, systematic uncertainty dominates over random uncertainty. (Winiwarter and Rypdal 2001)

Systematic uncertainty can be reduced or eliminated by improving emission estimation procedures and scientific understanding about the source, e.g. by performing focused emission measurements. A case of the reduction of systematic uncertainty can be identified in the development of primary PM emission estimates of domestic wood combustion from the time of Paper II to Paper V (see discussion in the previous section). The earlier mean emission estimate, 14 Gg($PM_{2.5}$) a^{-1}, was clearly out of the 95% confidence interval limits estimated for the current FRES version in Paper V (4.9 to 11.4 Gg a^{-1}).

Paper V presents an uncertainty analysis for FRES $PM_{2.5}$ emission calculation in two important source sectors: domestic wood combustion and road traffic. The emission factor and total activity confidence intervals were determined based on the variation in different measurement data sets and statistical uncertainties, respectively, i.e. they represent random uncertainty. However, the confidence interval values of activity disaggregation factors also represent the degree of deficiency of knowledge in the applied expert judgments, and thus include a component of systematic uncertainty.

Table 6 presents activity and emission factor uncertainty values in the main emission source sectors estimated in different literature sources. Activity uncertainties have been estimated relatively widely, mainly as part of national greenhouse gas inventories. Table 6 presents results from a Finnish analysis (Monni et al. 2004) and a summary from several studies (Rypdal and Winiwarter 2001). In general, the different estimates are in good agreement.

Emission factor uncertainties have been assessed variably for different pollutants. For primary PM, very few peer-reviewed studies at the regional or country level exist. In addition to Paper V of this study, Bond et al. (2004) presented a summary of several measurement studies of domestic wood combustion as a part of a global carbon emission inventory. As a part of the inventory report to UNECE/CLRTAP, the Finnish environment institute (2008) reported uncertainty estimates of air pollutants in power plants and the industrial sector based on extensive plant-basis material.

For the emission factors of gaseous pollutants, Rypdal (2002) presented an uncertainty assessment for the Norwegian CLRTAP inventory. The analysis of Schöpp et al. (2005) was carried out for the European wide RAINS model. Their results were mainly in agreement with the other values given in

Table 6. The values of Schöpp et al. (2005) were for unabated emission factors; additional uncertainty caused by removal efficiencies of control technologies were not presented.

Country-specific circumstances may have considerable effects on emission uncertainties in different source sectors. Therefore, the literature review in Table 6 is presented in a general and aggregated level, and the drawing of very specific conclusions is not possible. However, it can be concluded in general that:

- Uncertainties are mainly higher in emission factors than in activities
- Of the different pollutants, emission factor uncertainties are highest with PM and NMVOCs
- Of the different sources, domestic wood combustion shows the highest uncertainties. Emissions from small energy production plants and traffic non-exhaust emissions are also uncertain.

5. Conclusions

A national air pollution emission assessment tool, the Finnish Regional Emission Scenario (FRES) model, was developed in this study. The FRES model offers unique capabilities to assess the emission scenarios of multiple pollutants (primary TSP, PM_{10}, $PM_{2.5}$ and PM_1, SO_2, NO_x, NH_3 and NMVOCs) in a coherent framework. The most important model features include the possibility to (1) identify key emission sources, (2) estimate future emissions and cost-effective emission reductions and (3) provide emission data for air pollution impact assessment.

The study highlighted the advantages of the applied emission source description in the model; high spatial resolution and a combined top-down and bottom-up approach. To support air pollution impact assessment, emission estimates should be available with appropriate location information of release points, both in terms of emission altitude and geographical location. Representation of point sources is

Table 6. Activity and emission factor uncertainties of main emission sources based on this study and other literature (relative 95% confidence interval, %)

	Power plants and industry, large plants >50 MW / smaller plants	Domestic wood combustion	Traffic sources[1]	Other sources[2]
Total activity	±1-5 (±15-20)[a,3]; ±1-6[b]	±15[a] ±10-30[b]; ±10[c]	±1 (±5-30)[a,4] ±1-6[b]	±3-5[a,5] ±5-10[b,5]
Primary PM	±10 / ±50-60[e]	-54 to +88[c]; ±50-71[d]	-20 to +24 (-54 to +88)[c,6]	-[7]
SO_2	±10 / ±20-40[e] ±2-12 / -[f] ±5[g]	Nme	±1[f] ±5[g]	Nme
NO_x	±10 / ±50[e] ±7-20 / ±40-50[f] ±7.5[g]	±40-50[f] ±7.5[g]	±25-30[f] ±7.5-19[g]	Nme
NH_3	±10 / -[f,8] ±15[g,8]	Nme	Nme	±30[f,9] ±20[g,9]
NMVOCs	±45 / -[f]	±40-50[f]	±40-50[f]	±30-50[f,10]

[a)] Monni et al. (2004); [b)] several studies collected in Rypdal and Winiwarter (2001); [c)] Paper V of this study, $PM_{2.5}$ for primary PM; [d)] several studies collected in Bond et al. (2004), TSP for primary PM; [e)] Finnish environment institute (2008), TSP for primary PM; [f)] Rypdal (2002); [g)] Schöpp et al. (2005)

[1)] Road and off-road traffic; [2)] Agriculture and non-combustion NMVOC sources; [3)] ±15-20% for biomass; [4)] ±5-30% for off-road traffic sources; [5)] Livestock population (no uncertainty data found on solvents use and other non-combustion source activities); [6)] -54 to +88% for non-exhaust emission factors; [7)] No uncertainty data found on non-combustion primary PM sources other than traffic non-exhaust; [8)] Emission factors of fertilizer production; [9)] Emission factors of manure application; [10)] Emission factors of solvents use, oil loading and gasoline distribution

Nme = Not a major emission source

important for acidification which is caused to a large extent by emissions from large polluters with high stacks. Correct location information of stacks is important because of variable ecosystem sensitivities for acidification.

Spatial distribution of emissions is especially pronounced when assessing impacts on human populations. Especially primary PM is emitted to a considerable degree from low altitudes in urban areas. Therefore it is important to be able to assess the impacts near the emission sources. The spatial emission resolution of 1 × 1 km^2 applied in the FRES model, with relevant dispersion estimates, would enable the assessment of the impacts on populations inside urban areas.

The FRES model results pinpoint the most important emission sources at present and in the future. For SO_2, industrial and energy production plants are the major source, contributing to approx. 90% of the total emissions in 2000. For other pollutants, by contrast, relatively small sources are important, i.e. traffic vehicles (58%, 54% and 26% of total NO_x, NMVOCs and primary $PM_{2.5}$ in 2000, respectively), domestic combustion (25% and 12% for primary $PM_{2.5}$ and NMVOCs, respectively) and agriculture (97% for NH_3).

Air pollution emissions have decreased considerably in the past, especially on SO_2 (87% from the peak values in 1980 to 2000), but also on primary PM (46% from 1990 to 2000), NMVOCs (30% from 1990 to 2000) and NO_x (27% from 1989 to 2000). The emissions of NH_3, instead, have remained relatively stable. The emissions will continue to decrease in the future because of tightening emission standards in legislation. SO_2 emissions from industrial and energy production plants are expected to decrease slightly (approx. 25% from 2000 to 2020). However, the emissions are strongly dependent on future primary energy choices in power plants. Traffic tailpipe exhaust emissions will decrease considerably (76%, 74% and 60% for NMVOCs, primary $PM_{2.5}$ and NO_x, respectively). Unregulated non-exhaust emissions from traffic, i.e. fugitive dust, will become the dominant source of traffic-induced primary $PM_{2.5}$; thus a need for abatement measures for these emissions exists. Domestic combustion is not subject to tight emission standards and it will remain a major emission source of primary $PM_{2.5}$ and NMVOCs.

Of the factors influencing the development of future emissions, the majority of climate mitigation measures, e.g. energy saving and non-combustion based power production, brings co-benefits as reduced air pollution. The co-benefits appear most strongly for SO_2, 20 – 28% emission reduction in 2020 from no-action to climate-action pathways. In addition to co-benefits, potential trade-offs were also identified, e.g. in the form of PM emissions from increasing domestic wood combustion. Local air quality problems caused by domestic wood combustion and severe human health impacts of fine PM (e.g. Pope and Dockery 2006) emphasize the importance of this trade-off as one of the future challenges in Finland.

The FRES model enables the assessment of air pollution reduction potentials and costs. The results show that emission legislations set relatively strict standards for large energy production and industrial plants and the traffic sector, and further reductions are mainly costly. Smaller combustion plants and domestic heating sources, however, still have a cost-efficient remaining reduction potential, especially for primary $PM_{2.5}$.

Emission model verifications are important in the model development process. Emission comparisons against alternative estimates and measurements have guided FRES model development e.g. in the case of domestic wood combustion emission calculation. The performed uncertainty analyses led to the identification of key sources of model uncertainty. Highest relative uncertainties (above ±50%, 95% confidence interval) were estimated for primary PM emission factors of domestic wood combustion, traffic non-exhaust and small energy production plants below 50 MW_{th}.

National emission models can be used to compare and evaluate the results of large-scale model systems. They are especially valuable in assessing models used in international policymaking, e.g. the RAINS model applied within UNECE/CLRTAP and the EU/NEC

directive. The accuracy of national data in the RAINS model is crucial as it directly affects the quantification of legally binding commitments for each country. The FRES model results have been successfully used in the national assessment of international model systems and policy processes.

The FRES model has been acknowledged as a valuable tool for designing comprehensive emission scenarios. It has given integral input to the transfer of recent scientific development to decision making. The FRES model results have been used as a part of the Finnish Climate Strategy preparations (e.g. Hilden et al. 2001, 2005) and regional air quality assessments (e.g. Kousa et al. 2007, Osmo et al. 2005), and thus they have substantially supported Finnish air pollution and climate policies at both national and local levels. The role of the FRES model has become significant in supporting decision making on air pollution in Finland.

5.1 Contribution of the work

This study comprises the development and application of a mathematical modeling framework for the assessment of regional air pollution emissions. The FRES model is the first comprehensive system to calculate consistent emission scenarios of all major pollutants in high sectoral and spatial resolution in Finland. The model results enable the integrated assessment of air pollution and climate policies and their impacts in Finland.

6. Future work

This study has brought up results that can be used to direct future FRES model development and other air pollution research. The following emission source sectors with a need for future research were identified:

- **Domestic wood combustion:** A potential trade-off between climate and air pollution policies and potential for future emission reductions has been identified. Future research will focus on emission reduction possibilities and policy support in order to avoid negative impacts from increasing domestic wood heating.
- **Traffic non-exhaust:** Paper V identified a need for further improvement in primary PM calculation of traffic non-exhaust emissions. Furthermore, the primary $PM_{2.5}$ dispersion study showed the importance of traffic emissions from the point of view of population exposure. The FRES emission calculation will be developed to reflect specific Finnish seasonal characteristics (e.g. the effect of studded tires and traction sanding in winter) and differences between urban and non-urban roads.
- **Small energy production plants:** Relatively high PM emissions and considerable emission reduction potential have been identified for combustion plants in the size range 1 - 50 MW_{th}. Furthermore, because of their proximity to population clusters and lower stacks than in large power plants, they retain a potential for considerable population exposure effects. In the future, the FRES calculation will be improved by describing these plants as point sources. Furthermore, the population exposure effects of these plants should be studied.

In FRES model application, air pollution impact assessment will remain a major focus in the future. The source-receptor matrices applied with the FRES model will be developed further to include, in addition to current matrices for Finnish primary PM emissions, secondary PM and sulfur and nitrogen deposition, as well as LRT from outside Finland.

In the impact assessment on humans, this study indicated different emission – population exposure relationships for different emission sources. Furthermore, restrictions in the spatial resolution of applied source-receptor matrices in studying population exposure effects of low altitude emission sources (e.g. traffic and domestic combustion) in detail were revealed. In the future, 1 × 1 km^2 resolution matrices will be developed for low emissions for the area of Finland.

In the international air pollution assessment work of the EU and UNECE, one of the main focuses has been on integration of the air pollution framework with climate change

research (Sliggers 2004). Consequently, the RAINS model framework has been extended to greenhouse gas abatement in order to better assess the linkages between the two worlds. The extended framework, denoted as GAINS, includes the assessment of emission controls of greenhouse gases together with the emissions of air pollutants (Klaassen et al. 2004).

Integrations with and extensions towards different environmental aspects and other model frameworks will also be important future prospects in FRES development. First, extension towards climate change assessment frameworks, in parallel with RAINS/GAINS development, will be one objective. The second direction will be pronounced collaboration with other regional scale air pollution models; integration both with the RAINS framework and with other national IAMs to form model networks or clusters. Such clusters of national models could operate either as a collaboration network with integrated compilation of results in a coherent framework, or as a tighter coupling of models to form an operational cluster covering the area of several countries, e.g. the Nordic countries. Third, impact assessment would also benefit from the model integration towards higher spatial resolution, i.e. integration with local or urban scale air pollution models. The model integrations at these different scales will enhance the assessment of different aspects of different air pollution problems.

In the long term, the diverse features in FRES emissions open many possibilities. For climate change integration, the estimates for different chemical PM species, especially those for black and organic carbon and sulfate, would enable the use of the FRES model (or a wider cluster of similar models) as a provider of contributing aerosol emissions for climate change assessment frameworks.

For human health assessment, directions in emission modeling will depend largely on future findings about the impacts of different PM characteristics (e.g. size and chemical compounds). The FRES construction with size and chemically resolved PM estimates (e.g. ultrafine ($PM_{0.1}$) and carbonaceous particles and heavy metals) provides a good basis to meet these future challenges.

Tiivistelmä

Ilmansaasteiden päästöjä aiheutuu useista eri lähteistä. Ne vaikuttavat usein vahingollisesti ympäristöön ja ihmisten terveyteen. Ilmansaasteiden päästöjen ja vaikutusten arviointiin on kehitetty matemaattisia malleja. Tämä työ esittelee tuloksia Suomen alueellisen päästöskenaariomallin (Finnish Regional Emission Scenario, FRES) soveltamisesta. FRES-malli kuvaa ihmisperäiset rikkidioksidin (SO_2), typen oksidien (NO_x), ammoniakin (NH_3), ei-metaani haihtuvat orgaaniset yhdisteiden (NMVOC:t) sekä primääristen hiukkasten (TSP, PM_{10}, $PM_{2.5}$ ja PM_1) päästöt koko Suomen alueelta. Työn tarkoitus oli määrittää tärkeimmät nykyiset ja tulevaisuuden päästölähteet Suomessa, arvioida päästövähennysten mahdollisuuksia ja kustannuksia, sekä arvioida ilmastopolitiikan vaikutuksia ilmansaasteisiin. Myös päästöepävarmuuksia analysoitiin. Työssä arvioitiin mitkä päästömallin ominaisuudet ovat tärkeitä erilaisissa ilmansaasteiden vaikutustarkasteluissa.

Tärkeimmät päästölähteet SO_2:lle Suomessa ovat suuret energiantuotanto- ja teollisuuslaitokset (64 % vuoden 2000 kokonaispäästöistä 76 Gg a^{-1}). Liikenne on tärkein päästölähde NO_x:lle (58% 206 Gg a^{-1}:sta), NMVOC:eille (54% 152 Gg a^{-1}:sta) ja primääri-$PM_{2.5}$:lle (26% 31 Gg a^{-1}:sta). Maatalous aiheuttaa korkeimmat NH_3-päästöt (97% 33 Gg a^{-1}:sta). Muita ilmansaasteiden tärkeitä päästölähteitä ovat puun pienpoltto primääri-$PM_{2.5}$:lle (25%) ja NMVOC:eille (12%), sekä liikenteen ja muiden ihmisaktiviteettien nostattama pöly primääri-PM_{10}:lle (30% 46 Gg a^{-1}:sta). Tulevaisuudessa liikenteen pakokaasupäästöt laskevat tiukentuvien päästömääräysten ansiosta merkittävästi, 60% - 76 % vuodesta 2000 vuoteen 2020. Suurten polttolaitosten päästöt riippuvat tulevaisuuden polttoainevalinnoista; pääosin päästöt laskevat lievästi. Lähteiden, joihin ei kohdistu tiukkoja päästöraja-arvoja, kuten pienpoltto tai liikenteen nostattama pöly (ts. liikenteen ei-pakokaasuperäiset päästöt), päästöt uhkaavat kasvaa tulevaisuudessa. Päästöjen lisävähennyspotentiaalia maltillisilla vähennyskustannuksilla löytyy pääasiassa primääri-$PM_{2.5}$:lle pienissä energiantuotantolaitoksissa

ja pienpolttosektorilla. Suurin osa kasvihuonekaasujen rajoituskeinoista, esim. energiansäästö ja ei-polttoperäinen energiantuotanto, vähentävät myös ilmansaasteiden päästöjä, erityisesti SO_2:ta (20% - 28%). Jotkut kasvihuonekaasujen rajoituskeinoista, esim. puun pienpoltto, saattavat kuitenkin lisätä ilmansaastepäästöjä. Suurimmat päästöepävarmuudet arvioitiin puun pienpolton, liikenteen pölypäästöjen ja pienten energiantuotantolaitosten primääri-PM päästökertoimille.

Tärkeimmät päästömallin ominaisuudet ovat suurten teollisuus- ja voimalaitosten oikeat sijaintitiedot arvioitaessa happamoittavien päästöjen vaikutuksia, ja pienten päästölähteiden kuvaus tarkalla alueresoluutiolla arvioitaessa vaikutuksia ihmisiin. Erityisesti primääriset $PM_{2.5}$ päästöt aiheutuvat merkittävissä määrin pienistä matalan päästökorkeuden lähteistä kaupunkialueilla, joten on tärkeää pystyä arvioimaan näiden päästöjen vaikutuksia päästöjen lähialueilla. FRES-mallin suurten pistelähteiden yksityiskohtainen kuvaus ja 1 $\times$ 1 km^2 alueresoluutio pienille päästölähteille mahdollistavat mallin käytön monien ympäristövaikutusten ja niiden vähentämismahdollisuuksien arvioinnissa.

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Appendix A. Source sector and fuel division in the FRES model

Table A1. Source sector division in the FRES model

Main source sector		
1st level sub-sectors	2nd level sub-sectors	Technologies
Power plants		
New Existing	> 50 MW_{th}, District heat > 50 MW_{th}, Condensating 5 – 50 MW_{th} < 5 MW_{th}	Pulverized fuel Fluidized bed Grate Burner Turbine
Industrial combustion		
Industrial boilers Fuel conversion Other industrial comb.	> 50 MW_{th} 5 – 50 MW_{th} < 5 MW_{th}	Pulverized fuel Fluidized bed Grate Burner Turbine
Industrial processes		
Cement Chipboard and plywood Coking plants Sintering plants Iron and steel Non-ferrous metal Lime Mineral extraction and proc. N-fertilizer and N-acid Pulp and paper Black liquor recovery Oil refining Sulfuric acid Titanium oxide Glass and mineral wool Printing Organic chemicals Other processes		
Domestic combustion		
Residential buildings Recreational buildings Agricultural buildings Commerc./public buildings Industrial buildings	Boilers Stoves Fireplaces	Automatic boilers Manual boil. with heat storage Man. boil. without heat storage Modern masonry heaters Conventional masonry heaters Masonry ovens Kitchen ranges Sauna stoves Iron stoves Open fireplaces
Road traffic		
Light duty vehicles Heavy duty vehicles	Motorcycles and mopeds Passenger cars Vans Buses Trucks Road wear and resuspension Brake wear Fuel evaporation	4-stroke 2-stroke GDI Non-GDI

Machinery		
Forestry Agriculture Construction		4-stroke 2-stroke
Other traffic sources		
Snowmobiles Shipping Recreational vessels Rail traffic Domestic aviation		4-stroke 2-stroke
Agriculture		
Field harvesting Field tilling Cattle and horses Pigs and sheep Poultry Fur animals Fertilizer use		
Peat production		
Harrowing Loading Mechanical harvesting Milling Pneumatic harvesting, new Pneumatic harvesting, old Ridging Stockpile shaping Stockpiling		
Storage and handling		
Coal Iron ore Other bulk material		
Construction		
Dwellings Utility buildings		
Solvents and paints use		
Solvents use Paints use	Domestic and architectural Industrial	
Other sources		
Meat frying, Barbecues Meat frying, Other Tobacco smoke Fireworks Asphalt paving Gasoline distribution		

Table A2. Fuel division in the FRES model

Fuels	
Coal Peat Coke Black liquor Wood chips Wood pellets Wood logs Other wood fuels	Waste Light fuel oil Heavy fuel oil Diesel oil Gasoline Natural gas Process gas

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