

## TESS

Traffic Emissions, Socioeconomic valuation and Socioeconomic measures

Part 2:

**Exposure of the European population to atmospheric particles (PM) caused by emissions in Stockholm**

*Cover: Contributions of different sources in the Greater Stockholm area to the yearly average population exposure to PM<sub>2.5</sub> in Europe. For the residential heating source, two different estimates are given, one based on the emissions from Johansson and Eneroth (2007) and one based on emissions from SMED. Unit: person  $\mu\text{g} / \text{m}^3$ .*

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TESS - Traffic Emissions, Socioeconomic valuation and Socioeconomic measures PART 2: EXPOSURE OF THE EUROPEAN POPULATION TO ATMOSPHERIC PARTICLES (PM) CAUSED BY EMISSIONS IN STOCKHOLM						
Abstract/Sammandrag						
<p>Model calculations have been performed to estimate the effects of emissions in Stockholm on the population exposure to particulate matter (PM) outside the city.</p> <p>The impacts of five different emissions were investigated: Road traffic exhaust, split into Light Duty Vehicles (LDV) and Heavy Duty Vehicles (HDV), Sea Traffic, Power Plants and Residential Heating. The emissions from non-exhaust (mainly road wear due to use of studded tyres) were also treated, in addition to combustion sources.</p> <p>The calculated impact of the Stockholm emissions on atmospheric concentrations of particles were weighted by the European population distribution and the resulting yearly average <u>total European</u> population exposures for the different sources are summarised in the Table below (unit: person <math>\mu\text{g}/\text{m}^3</math>).</p>						
	Road traffic exhaust, light duty vehicles	Road traffic exhaust, heavy duty vehicles	Sea traffic	Power plants	Residential heating	Non-exhaust road traffic
Directly emitted combustion particles	150 000	55 200	17 000	86 800	ca178 000 - 886 000	-
Secondarily formed inorganic particles (nitrate, sulphate, ammonium)	315 000	117 000	70 100	193 000	ca26 000 - 70 300	-
Wear particles	-	-	-	-	-	2 520 000
<p>The uncertainties in the emission estimates for Residential Heating using biomass (wood) are very large but it seems that it is an important PM source in Stockholm. In this report two estimates of the emissions have been used. In the lowest estimate, which seems more realistic, the contribution to population exposure of <i>directly emitted</i> combustion particles from residential heating is of similar magnitude (37%) as the contribution from road traffic exhaust (42%). For all sources, except Sea Traffic, the total population exposure to combustion PM is much larger within Stockholm than outside; for shipping the total exposure is about as large outside the city as within.</p> <p>For all sources, except residential heating, the secondary inorganic aerosol (SIA) exposure is higher than the combustion particle exposure.</p> <p>Non-exhaust particles dominate the total impact on PM10 exposure, contributing about 60-70% to the total exposure, due to all the studied sources in Stockholm. The calculated population exposure due to the wear particles is to a very large extent (87%) occurring within the Greater Stockholm area.</p>						
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# Preface

This is a report resulting from the research project TESS – Traffic Emissions, Socioeconomic valuation and Socioeconomic measures that is financed by EMFO. In 2002 an agreement about the EMFO programme was reached between the partners from the Swedish Vehicle Research Council, PFF. EMFO is a sector-wide research competence to develop vehicles and vehicle components with emission levels that are sustainable in the long term. The aim of EMFO is to offer academia, industry and authorities access to necessary knowledge and pioneering solutions that are necessary if vehicle technology is to develop in the desired direction. One important task is to coordinate activities within the programme with both national and international research in the field.

EMFO comprises subsidiary programmes and two of these were: “Socio-economic evaluation of the health and environmental impact of different emissions” and “Optimal range of socio-economic measures”. TESS undertakes research in these two areas but it is also related to the subsidiary programme: “Health and Environmental Impact”. The application was approved in 2005 and the project runs during 2005-2008.

The basis for the research in TESS is the valuation methods developed in the EU funded ExternE projects where the external cost of emissions is calculated by tracing the effects that the emissions have on human health and then valuing these effects. The aim is to calculate the external costs related to particles that local emissions (from traffic and other sources) generate on a local and regional scale using Stockholm as a case study. Based on this information an analysis will be made on what reductions measures are likely to be efficient from an economic point of view.

The analysis undertaken in TESS requires collaboration between researchers from different research disciplines and therefore there are four parties involved in this project. Coordinator for the project is VTI, where Lena Nerhagen is project leader as well as responsible for the economic analysis. Christer Johansson and Kristina Eneroth at SLB analys (Environment and Health Administration, Stockholm) contribute with information about local emissions and perform dispersion model calculations for the Stockholm area. Bertil Forsberg at Umeå Universitet is responsible for the health impact assessment. Finally, Robert Bergström at SMHI undertakes the regional scale dispersion modelling.

This report is the result of the second part of the TESS project, the regional scale dispersion modelling for evaluating the impact of the Stockholm emissions in Sweden and the rest of Europe. This analysis has been undertaken by SMHI using the MATCH model and emission data from SLB (for Stockholm), SMED (for the rest of Sweden) and EMEP (for emissions outside Sweden).

# Summary

Model calculations have been performed to estimate the effects of emissions in Stockholm on the population exposure to particulate matter (PM) outside the city. In this report, the total European exposures to PM from different sources in Stockholm are presented and the results are compared to the earlier TESS-study of the exposure *within* Stockholm.

The impacts of five different emissions were investigated: Road traffic exhaust, split into Light Duty Vehicles (LDV) and Heavy Duty Vehicles (HDV), Sea Traffic, Power Plants and Residential Heating. The emissions from non-exhaust (mainly road wear due to use of studded tyres) were also treated, in addition to combustion sources.

The calculated impact of the Stockholm emissions on atmospheric concentrations of particles were weighted by the European population distribution and the resulting yearly average total European population exposures for the different sources are summarised in the Table below (unit: person  $\mu\text{g}/\text{m}^3$ ).

	Road traffic exhaust, ldv <sup>a</sup>	Road traffic exhaust, hdv <sup>b</sup>	Sea traffic	Power plants	Residential heating	Non-exhaust road traffic
Directly emitted combustion particles	150 000	55 200	17 000	86 800	ca178 000 - 886 000	-
Secondarily formed inorganic particles (nitrate, sulphate, ammonium)	315 000	117 000	70 100	193 000	ca26 000 - 70 300	-
Wear particles	-	-	-	-	-	2 520 000

<sup>a</sup> ldv = light duty vehicles

<sup>b</sup> hdv = heavy duty vehicles

The uncertainties in the emission estimates for Residential Heating using biomass (wood) are very large but it seems that it is an important PM source in Stockholm. In this report two estimates of the emissions have been used. In the lowest estimate, which seems more realistic, the contribution to population exposure of *directly emitted* combustion particles from residential heating is of similar magnitude (37%) as the contribution from road traffic (42%). For residential heating more than 90% of the exposure occur within the Greater Stockholm area. Further work is needed to validate this estimate. For all sources, except Sea Traffic, the total population exposure to combustion PM is much larger within Stockholm than outside; for shipping the total exposure is about as large outside the city as within.

For all sources, except residential heating, the secondary inorganic aerosol (SIA) exposure is higher than the combustion particle exposure (for example, both for LDV and HDV the SIA exposures are ca 110% higher than the directly emitted combustion PM exposures).

Non-exhaust particles dominate the total impact on PM10 exposure, contributing about 60-70% to the total exposure, due to all the studied sources in Stockholm. The calculated population exposure due to the wear particles is to a very large extent (87%) occurring within the Greater Stockholm area.

# Introduction

## General background

It has long been recognized that emissions from traffic have a negative impact on human health. In later years there has been emerging consensus that the main influence is due to particulate matter (WHO, 2005). From an economic point of view these negative effects are external costs caused by traffic that, if not accounted for in decision making regarding transport, will result in a non-optimal allocation of resources leading to welfare losses. There are however various measures in place aimed at reducing the negative health impact (i.e. the external costs) of the emissions from traffic. The measures include emission control legislation but also air quality objectives for local concentration levels in urban areas that if exceeded compels the local authorities to take action. Also road pricing measures are increasingly considered as an option since the new information technology has opened up for new technical solutions. One such example is the Stockholm trial where rush hour road pricing was implemented, resulting in reduced traffic to and within the city area and thereby reductions in emissions and concentration levels.

To be able to implement road pricing measures, but also for the evaluation of other control measures through benefit-cost analysis, information on the external cost of traffic emissions is needed. In the Impact pathway approach (IPA), that has been developed in the ExternE projects, the external cost is calculated as the product of exposure, effect and value. All these inputs are the result of ongoing empirical research and they are all related to uncertainties, hence the external cost that is calculated is not “the” cost. Regarding particles there is for example recognition among the research community that there are different types of particles and that it is likely that their impact on human health differ. Still the current practice is to treat fine particles (which are considered to be most detrimental to health) as equally harmful irrespective of origin. Hence, there is only one function used for the health impact of fine particles (so called PM<sub>2.5</sub>). However, what is mostly measured in urban areas is the concentration of PM<sub>10</sub> that contain both fine and coarse particles since the current air quality guidelines are based on these<sup>1</sup>. The most important local source of PM<sub>10</sub> in many urban areas in Sweden is coarse particles from road wear (Omstedt et al., 2005). In spring, when the roadways are dry, the contribution from road wear particles may be 30 times the direct emissions from the exhaust pipe. These mechanically generated road dust particles are however not considered in calculations of the external cost that is based on the original ExternE-methodology (Friedrich and Bickel, 2001; Bickel and Friedrich, 2007).

In reality the measured concentrations of fine particles and PM<sub>10</sub> in an urban area is composed of several types of particles such as combustion particles from different sources, non-exhaust particles from road wear and secondary particles from sources outside the city. Therefore it is not possible to assess the actual impact on health from local traffic emissions using measurement data of the total concentrations. The impact of different contributions to the total PM<sub>10</sub> concentrations at street canyon and urban background in central Stockholm is illustrated in Figure 1.

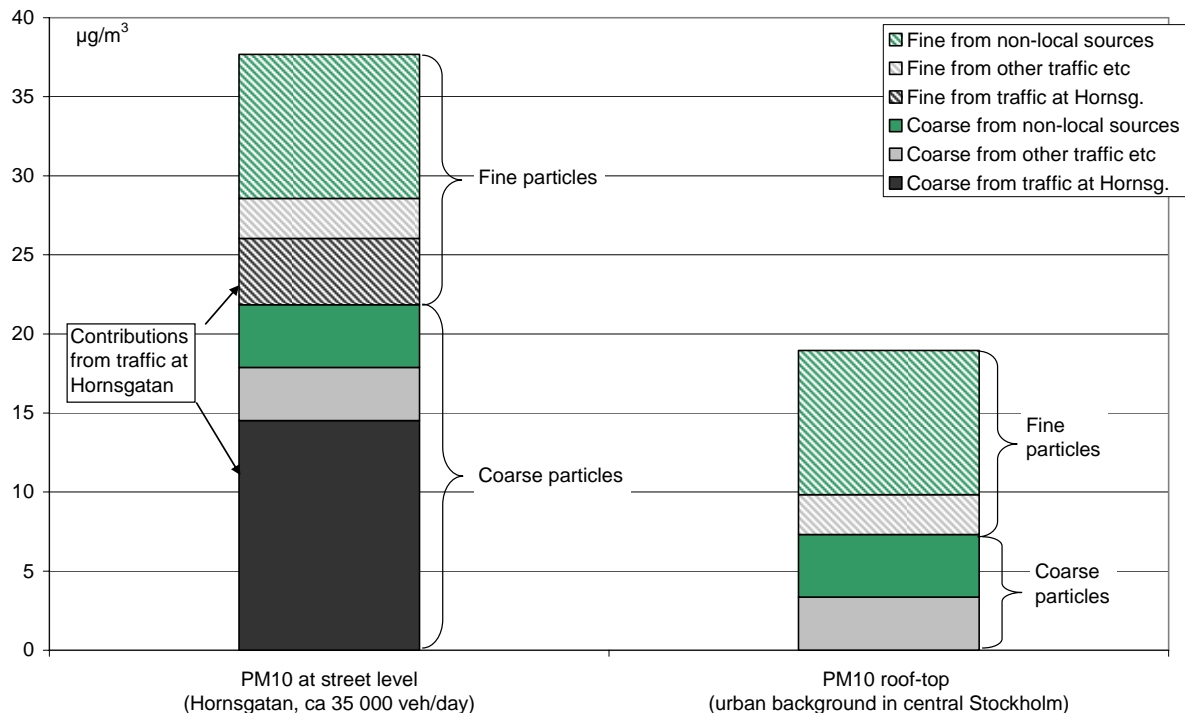
Hence, in order to undertake analysis of the influence of traffic emissions on human health dispersion models are needed. There is however an additional problem with the current measurement on which Figure 1 is based. If we are only interested in exhaust particles from local traffic, measurements or modelling of PM<sub>2.5</sub> are not relevant. This is because exhaust particles consist mainly of ultrafine particles (with diameters <0.1 µm) and hence their

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<sup>1</sup> There is a new EU-directive being prepared that will include limit values for both PM<sub>10</sub> and PM<sub>2.5</sub>

([http://ec.europa.eu/environment/air/cafe/pdf/com\\_2005\\_447\\_en.pdf](http://ec.europa.eu/environment/air/cafe/pdf/com_2005_447_en.pdf)).

contribution to the concentration of PM<sub>2.5</sub> is small. Therefore, in TESS we will not base the analysis on PM<sub>2.5</sub> or PM<sub>10</sub> but we will model the contribution of *exhaust* and *non-exhaust* and *secondary* particles.



**Figure 1.** The relationship between the contribution from traffic and other sources to the PM<sub>10</sub> concentration at a densely trafficked site (Hornsgatan) and at Urban background (a roof-top site) in central Stockholm (annual mean contributions). "Fine" particles refers to PM<sub>2.5</sub> and "Coarse" particles to PM<sub>10</sub>-PM<sub>2.5</sub>.

In TESS the purpose is to investigate how important the external cost of traffic generated particle emissions are in relation to the cost of other particle emission sources. To do this we will both investigate how the exposure varies between sources but also assess if it is reasonable to assume that the impact differs between particle emissions from different sources. Recent research studies which in various ways have tried to estimate the separate impact of traffic exhaust emissions on health have found larger effects than studies using PM<sub>2.5</sub> (Forsberg et al., 2005). One reason for this could be that although the mass concentration of exhaust particles is small the exhaust emissions largely contribute to the number of particles in urban air or that they are much more toxic than the particle fraction that dominate the PM<sub>2.5</sub> levels. Nitrogen oxide concentrations are highly correlated with the number of exhaust particles (Gidhagen et al., 2004; Olivares et al., 2007), therefore NO<sub>x</sub> or NO<sub>2</sub> can be a good indicator for the exposure to particle exhaust emissions. In the present study we will also investigate the influence of local traffic and other sources on a regional scale, for example by studying secondary particle formation due to local emissions of NO<sub>x</sub> and SO<sub>2</sub>. In the project we use Stockholm as a case study. The information on external costs will then be used to analyse the economic efficiency of different reduction measures.

### Impact of Stockholm emissions on the rest of Sweden and Europe

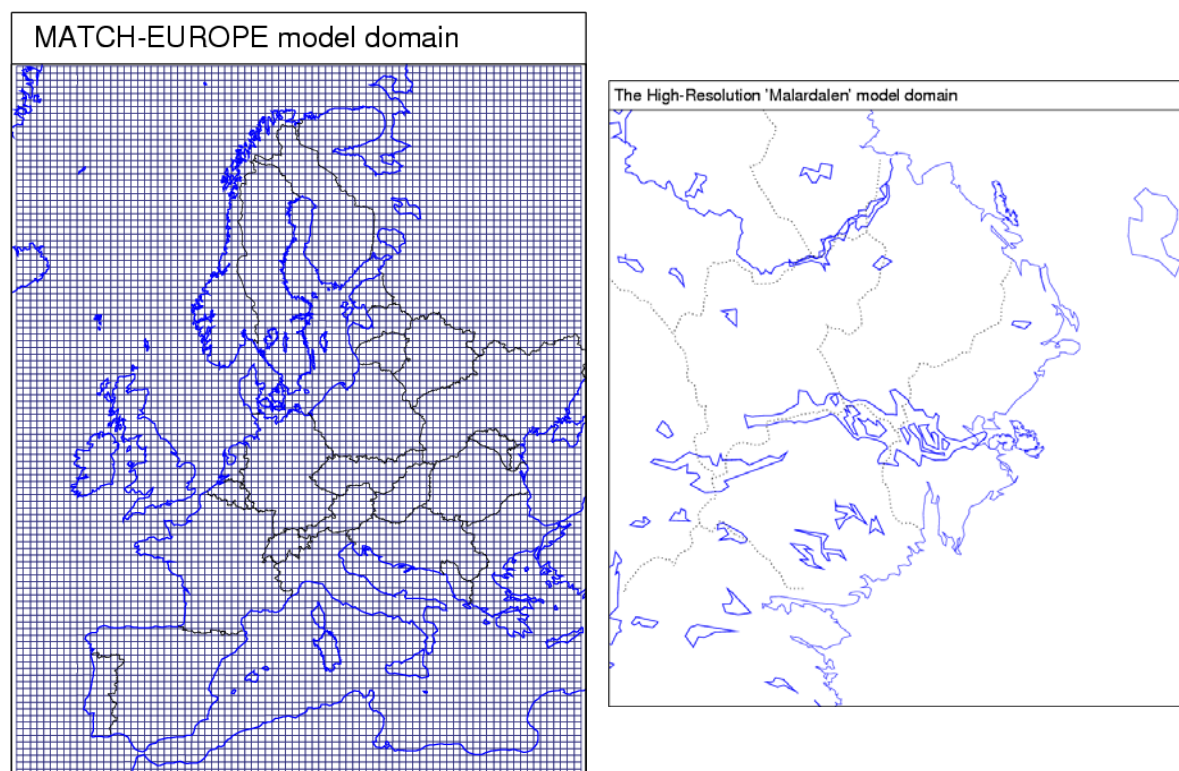
Particles emitted in Stockholm will, to some extent, be transported out of the city and contribute to the PM levels in other parts of Europe. The gaseous nitrogen oxides (NO<sub>x</sub>) and sulphur dioxide (SO<sub>2</sub>) emitted in Stockholm can also be oxidised to form particulate nitrate and sulphate and these secondary particles (denoted SIA, secondary inorganic aerosol) may be more important outside the emission area (Stockholm) than inside since it takes time for the SIA to form.

In this part of the TESS project the regional dispersion and chemistry of traffic exhaust air pollutants ( $\text{NO}_x$ ,  $\text{SO}_x$ , volatile organic compounds (VOC) and exhaust particles) and non-exhaust particles (road, brake and tyre wear) in the Stockholm area were investigated. Emissions from sea traffic, energy production and residential heating were also studied, using the same methodology as for road traffic.

Model calculations to determine the impact of emissions in Stockholm were performed on two different scales/regions. A large model domain covering most of Europe, with a horizontal resolution of ca 44km, was used to study impacts on a continental scale; the model area is illustrated in Figure 2.

For a more detailed investigation, of impacts closer to Stockholm, a smaller model domain was used, covering Greater Stockholm and the surrounding area, including the counties of Stockholm, Uppsala, Västmanland and Södermanland and the major cities in the surrounding counties (Linköping, Norrköping, Örebro, Gävle, Falun, Borlänge) as well as the Åland Islands. The simulations in the smaller domain were performed with a much higher resolution (ca 5km) than the European-scale calculations. The concentrations from the European scale simulations were used as boundary conditions in the higher resolution modelling. Hereafter, the high resolution domain will be called the Mälardalen domain.

All calculations were performed for one complete year (meteorology from 2003 was used).



**Figure 2.** The modelling domains. Left: The European scale model domain with ca 44km horizontal resolution (each square represents one model grid cell). Right: The high-resolution, "Mälardalen" model domain; the domain is split into 59 × 60 grid cells, with 5km horizontal resolution in the Stockholm region.

# Methods

## The MATCH Model

The Multi-scale Atmospheric Transport and Chemistry (MATCH) model is a three-dimensional, Eulerian model developed at the Swedish Meteorological and Hydrological Institute (SMHI). It is used in a range of applications from urban scale studies (e.g., Gidhagen et al., 2005) on ca. 5km, or higher, resolution to regional/continental scale studies on acidifying/eutrophying deposition and photochemistry (e.g., Andersson et al., 2007, Langner et al., 2005; van Loon et al., 2007). MATCH is used for air pollution assessment in Sweden and the Baltic Sea region; the air pollution budgets of nitrogen and sulphur compounds for Sweden are calculated annually, using a system combining the MATCH model calculations and monitoring data from Sweden and the neighbouring countries. The model is also used operationally to provide forecasts of radioactivity in case of nuclear emergencies in Europe (Langner et al., 1998a).

The MATCH model has participated in many international studies where different models have been evaluated against observational data and each other. Two examples of such intercomparison studies are the on-going EuroDelta-project (see e.g., van Loon et al., 2007) and the EuroTrac-2 study on aerosol modelling by Hass et al. (2003).

Some details about the MATCH model are given below; a thorough description of the model is given in Andersson et al. (2007).

## Transport

The basic transport model includes modules describing emissions, advection, turbulent diffusion and dry and wet deposition. Atmospheric weather data are taken from the numerical weather prediction (NWP) model HIRLAM (the operational version of SMHI was used in this study). Meteorological data are read every three hours. The data are then interpolated in time to yield hourly data. Special attention is given to interpolation of the horizontal wind where vector increments are applied. The vertical wind is calculated internally to assure mass consistency of the atmospheric motion after the time interpolation of the horizontal winds. The advection scheme is Bott-type (Bott 1989), using fourth-order scheme in the horizontal and a second-order scheme in the vertical. A complete description of the transport model can be found in Robertson et al. (1999).

The vertical resolution of the model is based on the resolution of the meteorological data. For the meteorological year 2003, used in this project, the number of model levels was 22 corresponding to a vertical extent of ca 5500 m. In the high resolution domain 16 levels were used, reaching ca 2900 m. In both cases the lowest model level was ca 60m thick.

## Deposition

The dry deposition of gaseous and particulate species is calculated using a resistance approach depending on land-use. For MATCH-Europe a simple scheme is used with only four different land-use classes (Water, Forest, Low vegetation and No vegetation). The dry deposition flux is proportional to the concentration of each component and the inverse of the sum of the aerodynamic resistance and a species specific surface resistance. For simplicity the same aerodynamic resistance is used for all surfaces within a grid square. For species with stomatal uptake as a major deposition route, surface resistance is calculated taking into account soil moisture, soil type, vegetation type, leaf area index, photosynthetic active radiation and temperature. For other species a simpler approach is used with only monthly varying surface resistances. For O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub> and NH<sub>3</sub> lower deposition velocities are used for snow covered surfaces.

For most species, precipitation scavenging is assumed to be proportional to the precipitation intensity and a species-specific scavenging coefficient:

$$\frac{dc_i}{dt} = -c_i \Lambda_i P$$

where  $c_i$  is the concentration of species  $i$ ,  $\Lambda_i$  is the scavenging coefficient ( $\text{s}^{-1} \text{mm}^{-1} \text{hour}$ ) and  $P$  is the precipitation rate (in  $\text{mm hour}^{-1}$ ).

For  $\text{O}_3$ , hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) and  $\text{SO}_2$  in-cloud scavenging is calculated by assuming Henry's law equilibrium; sub-cloud scavenging is neglected for these species. All particulate sulphate inside clouds is assumed to be dissolved in cloud droplets; in-cloud scavenging is proportional to the fraction of the cloud water that hits the ground as precipitation. Sub-cloud scavenging for sulphate is calculated as in Berge (1993).

### Particles

The size distributions of the directly emitted particles (or primary PM, "PPM", consisting of combustion particles and wear particles) are important since they determine the dry deposition velocities and wet scavenging efficiencies of the PM. Combustion particles are typically very small; in the MATCH simulations they are treated as consisting of a mixture of particles with two different mean aerodynamic diameters 50nm (ca 10-13%) and 200nm (ca 87-90%), this size distribution was chosen to represent effective emission sizes, i.e., sizes relevant for the relatively coarse horizontal resolution of the MATCH model. Road wear particles are much larger than combustion particles and in this project 20% of the emission is assumed to be fine particles, with an average emission diameter of  $1.75\mu\text{m}$ , and the rest (80%) are coarse particles, with a mean emission diameter of  $3.5\mu\text{m}$ . No aerosol dynamics is included (except deposition and hygroscopic growth; a growth function, specified for sulphuric particles, by Koutrakis (1989) and modified by Quinn and Ondov (1998) was used).

The dry deposition scheme for the PPM takes into account gravitational settling, Brownian diffusion, inertial impaction, interception and possible rebound from dry surfaces. The scheme is based on Zhang et al. (2001).

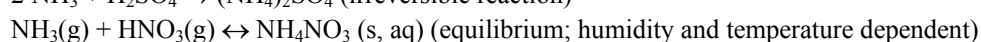
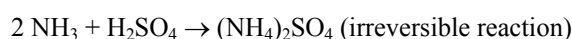
The wet deposition scheme for PPM takes into account 3-dimensional cloud cover and cloud water content. Wet deposition of PPM is split into in-cloud scavenging and below cloud scavenging. All particles inside clouds are assumed to go into cloud droplets. This means that the in-cloud scavenging coefficient is equal to the fraction of cloud water that hits the ground as precipitation. Below cloud scavenging of particles is based on an assumed lognormal distribution of the rain drop spectrum and takes into account collection of particles by Brownian diffusion, inertial impact and interception as formulated in Dana and Hales (1976).

### Chemistry scheme

Depending on the application specific modules describing, e.g., chemistry can be added to the basic transport model. In this study the photochemistry scheme proposed by Simpson et al. (1993) is used with some modifications and updates; the most notable being a modified production mechanism for isoprene chemistry, based on the so-called Carter-1 scheme (Carter, 1996, Langner et al., 1998b). The photochemical scheme includes about 130 reactions and 61 chemical components; the basic idea is to use a limited number of *representative* volatile organic compounds (VOC) to model all emitted VOC; the selection of model VOCs is based on Photochemical Ozone Creation Potentials (POCP).

#### *The aerosol chemistry:*

Only a few chemical reactions are considered for the ammonia-ammonium conversion in MATCH:



The sulphuric acid ( $\text{H}_2\text{SO}_4$ ), participating in the first reaction, can be directly emitted or formed in the model by gas phase oxidation of  $\text{SO}_2$ , by OH or  $\text{CH}_3\text{O}_2$ , or oxidation in cloud droplets, by  $\text{H}_2\text{O}_2$  or  $\text{O}_3$ , (a constant cloud water pH of 5 was used).

A bulk approach is used for secondary inorganic aerosol formation. This means that aerosol dynamics is not included and all SIA components are assigned to the fine particle mode  $\text{PM}_{2.5}$  (i.e., no part of the nitrate is included in the coarse mode).

### Boundary and initial concentrations

Boundary concentrations for the European scale model domain were based on data from earlier MATCH projects (e.g., Solberg et al., 2002, 2005; Tilmes et al., 2002; Roemer et al., 2003). Monthly or seasonally varying lateral boundary conditions were used for some species. The boundary conditions are partly based on observations at background locations and partly on large-scale model calculations. The lateral  $\text{O}_3$  boundary concentrations were based on measurement data for 2003 from EMEP measurement stations (Internet URL: <http://www.emep.int>) and the top concentrations on sonde data from Ireland, the United Kingdom and Norway (average for the year 1996-2001). Initial conditions were interpolated from the specified lateral boundary values.

### Emission data

Emission data for Greater Stockholm for  $\text{NO}_x$ ,  $\text{SO}_x$ , VOC, directly emitted combustion particles and road wear particles were provided by Johansson and Eneroth (2007). These data were produced within the TESS project and are described in detail in the TESS part 1 report (Johansson & Eneroth, 2007).

To describe the chemical evolution of the  $\text{NO}_x/\text{SO}_x/\text{VOC}$  emissions from Stockholm, and the resulting production of secondary inorganic aerosol, accurate emissions are also needed for the rest of Sweden and Europe. For Sweden  $\text{NO}_x$ ,  $\text{SO}_x$ , VOC, CO and  $\text{NH}_3$  emission data from the SMED (Swedish Methodology for Environmental Data, [www.smed.se](http://www.smed.se)) project, with ca 1km resolution, were used; the geographical distribution from 2001 was used but the total emissions were rescaled to be in line with estimated total national emissions in 2003. Passenger cars equipped with catalytic converters emit ammonia ( $\text{NH}_3$ ).  $\text{NH}_3$  emissions were taken from SMED, also for the Stockholm traffic (since  $\text{NH}_3$  data were not available from Johansson & Eneroth, 2007). Ammonia emissions from Heavy Duty Vehicles are expected to be small and were neglected in this study.

For emissions outside Sweden data from EMEP (<http://www.emep.int>) were used; the, so called, EMEP expert emission data for 2003 (Vestreng et al., 2006) were used; these data are based on official national data reported under CLRTAP to EMEP; missing/incomplete/erroneous data are corrected by EMEP. EMEP data are provided with a horizontal resolution of 50km. The EMEP data include anthropogenic emissions of  $\text{NO}_x$ ,  $\text{SO}_x$ , VOC,  $\text{NH}_3$  and CO as well as natural emissions of sulphur from oceans and volcanoes.

All emission data were regridded to the two different calculation grids used in MATCH.

EMEP provide emission data divided into 11 sectors (SNAP-codes, see e.g., EMEP/CORINAIR, 2002). The different sectors are given in Table 1.



**Table 1.** The SNAP sectors for emission data

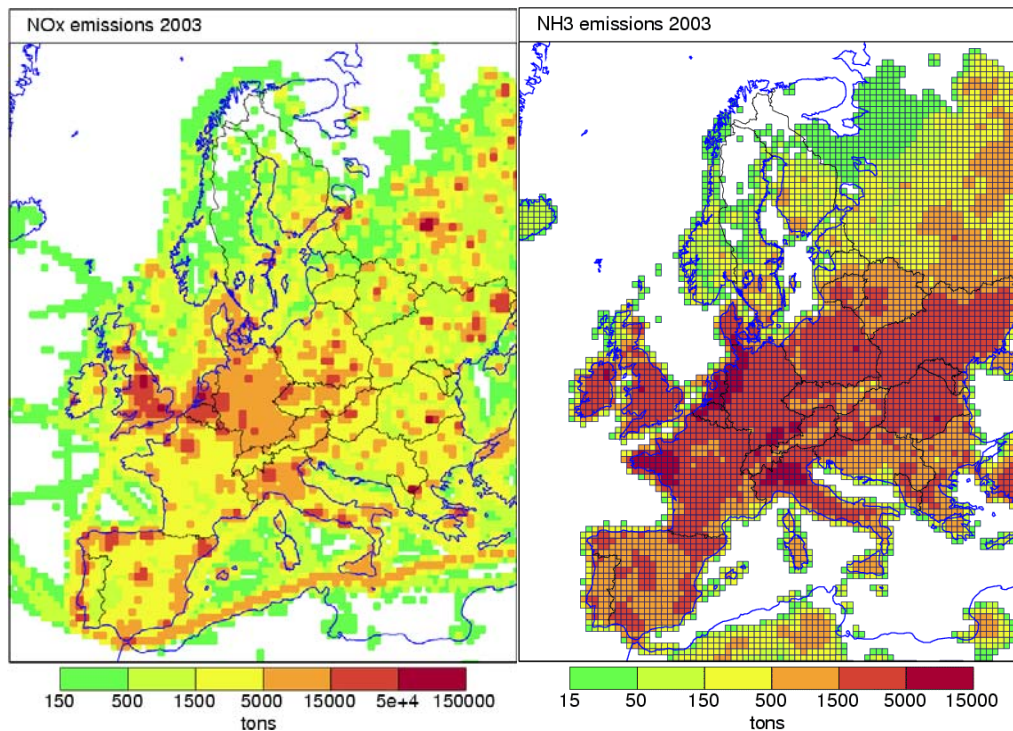
SNAP CODE	DESCRIPTION
1	Combustion in energy and transformation industries
2	Non-industrial combustion plants
3	Combustion in manufacturing industry
4	Production processes
5	Extraction and distribution of fossil fuels and geothermal energy
6	Solvent and other product use
7	Road transport
8	Other mobile sources and machinery
9	Waste treatment and disposal
10	Agriculture
11	Other sources

Anthropogenic SO<sub>x</sub> emissions are treated as 95% SO<sub>2</sub> and 5% sulphate. The NO<sub>x</sub> emissions are treated as 95% NO and 5% NO<sub>2</sub>. VOC emissions are split into the ten different model-VOC species of MATCH as indicated in Table 2.

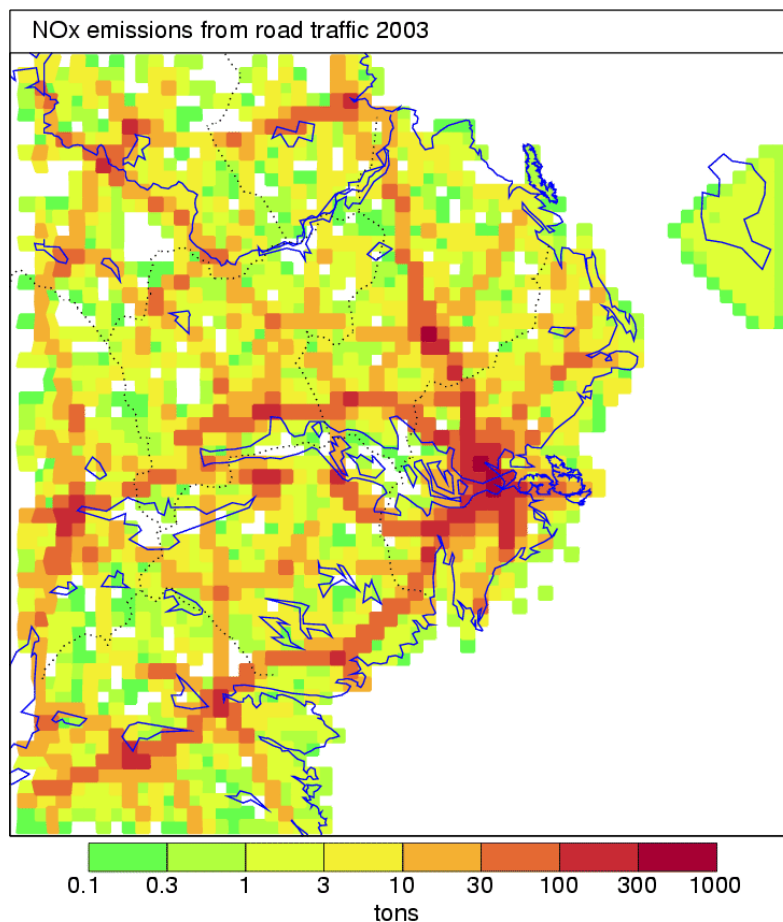
**Table 2.** Species split of VOC-emissions in MATCH. Unit: % of the total VOC emission in the respective SNAP sector

Specie	SNAP sector									
	1	2	3	4	5	6	7	8	9	10
<b>C<sub>2</sub>H<sub>6</sub></b>	4	12.1	5.4	6.1	9.0	9.7	4.8	5.0	28.5	12.5
<b>n-C<sub>4</sub>H<sub>10</sub></b>	14.25	53.2	36.5	33.7	87.9	23.3	39.55	50.5	38.2	12.5
<b>C<sub>2</sub>H<sub>4</sub></b>	1.9	8.5	10.6	5.8	0.5	0.6	10.45	13.9	2.9	12.5
<b>C<sub>3</sub>H<sub>6</sub></b>	3.3	8.6	2.2	5.4	2.0	0	6.6	4.4	0	0
<b>o-Xylene</b>	11.2	10.5	18.7	4.65	0.6	20.1	34.2	21.3	4.45	12.5
<b>HCHO</b>	65.25	4.2	21.4	0	0	0	2.2	2.8	18.5	0
<b>CH<sub>3</sub>CHO</b>	0	0	0	0.1	0	0	1.6	1.35	0	12.5
<b>CH<sub>3</sub>COC<sub>2</sub>H<sub>5</sub></b>	0	0	0	0	0	10.15	0	0	0.7	0
<b>C<sub>2</sub>H<sub>5</sub>OH</b>	0	2.95	5.0	35.2	0	22.3	0	0	4.0	12.5
<b>CH<sub>3</sub>OH</b>	0	0	0	1.2	0	6.8	0	0	0.5	12.5
<b>Unreactive</b>	0	0	0	7.6	0	6.6	0.8	0.8	2.0	12.5

Emissions of the reactive hydrocarbon isoprene (C<sub>5</sub>H<sub>8</sub>) from plants are calculated within the MATCH model using the E-94 isoprene emission methodology proposed by Simpson et al. (1995)). Isoprene contributes to formation of ozone and thereby indirectly to the oxidation of SO<sub>x</sub> and NO<sub>x</sub> to particulate sulphate and nitrate.



**Figure 3.** Examples of European emission data used in the MATCH simulations. Left: Total NO<sub>x</sub>-emissions in 2003 (unit: ton NO<sub>2</sub>) and Right: Total NH<sub>3</sub>-emissions in 2003 (unit: ton NH<sub>3</sub>).



**Figure 4.** Example of higher resolution emission data used in the MATCH simulations in the Mälardalen domain. NO<sub>x</sub>-emissions from road traffic in 2003. Unit: ton NO<sub>2</sub>.

Standard plume-rise calculations can be performed in MATCH based on stack parameters (stack diameter, effluent temperature, volume flux) but in the TESS project all emissions were treated as area sources with fixed *effective* emission heights according to Table 3.

**Table 3.** Emission fractions per model level for the different SNAP sectors (the approximate vertical extents of the different model levels are given in parenthesis)

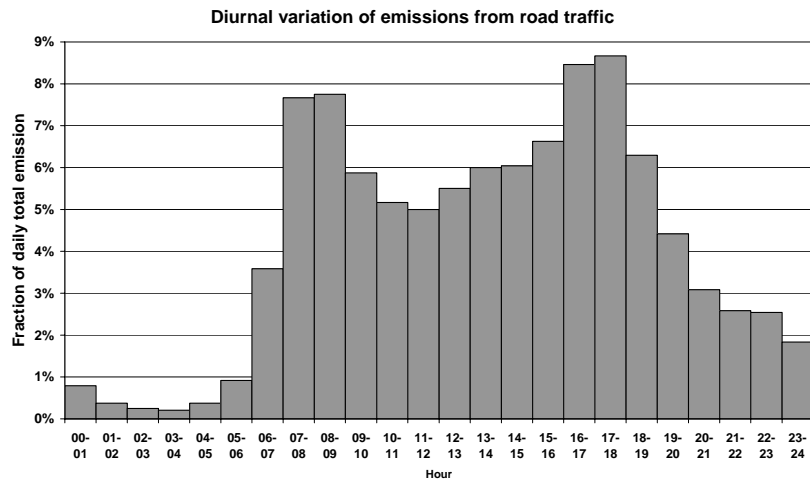
Model level	SNAP sector									
	1	2	3	4	5	6	7	8	9	10
<b>1 (0-60m)</b>	-	50%	-	90%	90%	100%	100%	100%	10%	100%
<b>2 (60-130m)</b>	-	50%	4%	10%	10%	-	-	-	15%	-
<b>4 (220-310m)</b>	8%	-	19%	-	-	-	-	-	40%	-
<b>5 (310-420m)</b>	46%	-	41%	-	-	-	-	-	35%	-
<b>7 (550-690m)</b>	29%	-	30%	-	-	-	-	-	-	-
<b>8 (690-850m)</b>	17%	-	6%	-	-	-	-	-	-	-

### Road traffic exhaust

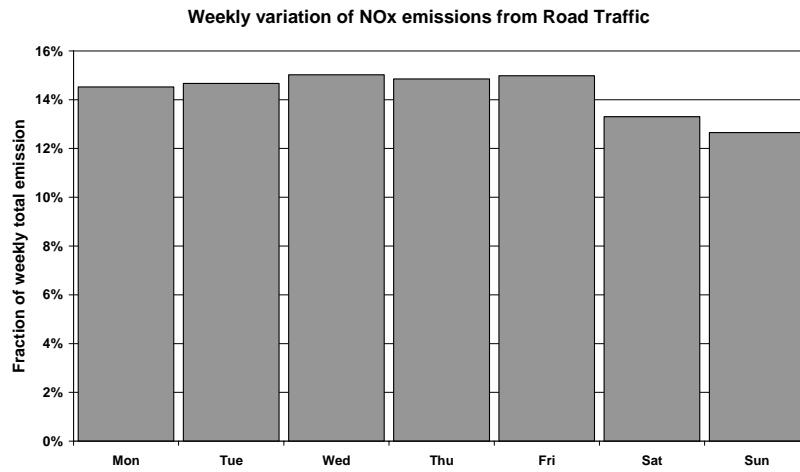
Emissions from road traffic were treated as surface level emissions in the MATCH model; all of the emissions are released in the lowest model level, which is ca 60m thick. The temporal variation of the road traffic emissions in Sweden is illustrated in Figure 5a-c. Variations on the seasonal scale<sup>2</sup> are rather small, with somewhat lower emissions in winter than in summer. Emissions during weekdays are a bit higher than those during weekends. The diurnal variation of the road traffic emissions is large with very small emissions during the night hours and distinct emission maxima during morning and afternoon rush-hours.

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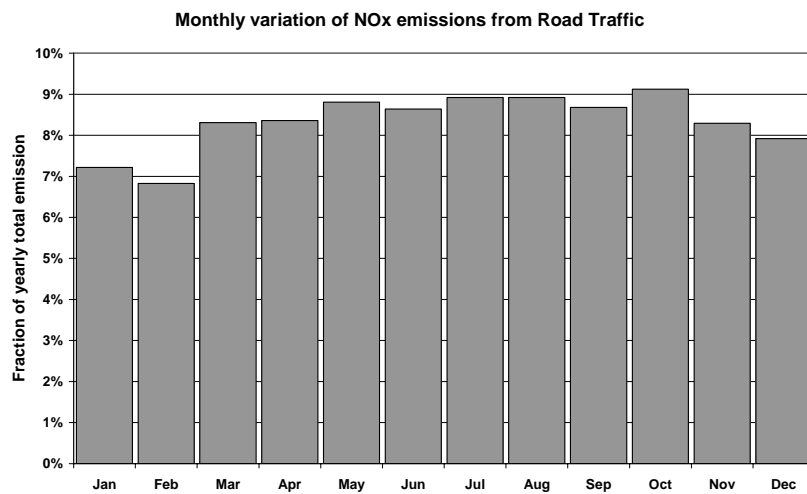
<sup>2</sup> The temporal variations were based on data from the GENEMIS project provided by the Institute of Energy Economics and the Rational Use of Energy ([IER](http://www.ier.uni-stuttgart.de/forschung/projektwebsites/genemis/)) at the University of Stuttgart ([www.ier.uni-stuttgart.de/forschung/projektwebsites/genemis/](http://www.ier.uni-stuttgart.de/forschung/projektwebsites/genemis/))



**Figure 5a.** Model variations of road traffic emissions with the time of day. The hourly fraction of the daily total emission is shown. Unit: % of the daily total emission.



**Figure 5b.** Model variations of road traffic emissions with the day of the week. The fraction of the weekly total emission is shown. Unit: % of the weekly total emission.



**Figure 5c.** Model variations of road traffic exhaust emissions with the month of the year. The fraction of the yearly total emission is shown for each month. Unit: % of the yearly total emission.

## Sea traffic

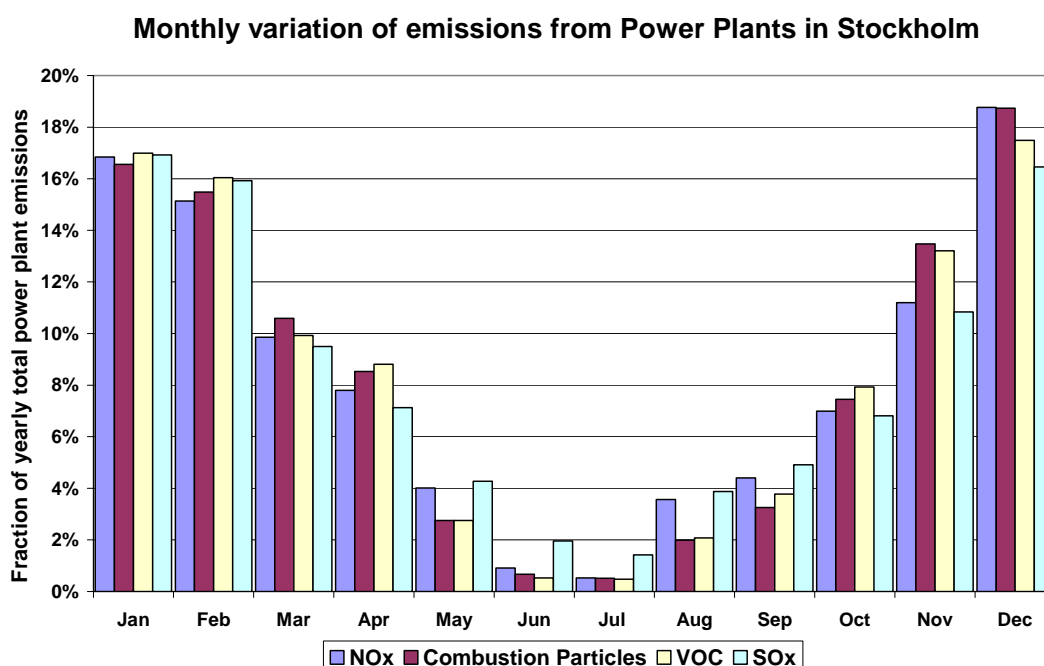
All sea traffic emissions were considered as surface level emissions, i.e., released in the lowest model level. Since shipping emissions are put in the same SNAP sector as machinery (8) the temporal variation applied were probably not optimal for the Sea traffic part in Stockholm; shipping emissions in grid points defined as open sea have no temporal variation (i.e., are constant in time) but for grid points in, and close to, Stockholm a seasonal variation was used with much lower emissions in July and August than for the other months. This, unrealistic, temporal variation of the shipping emissions is probably not of critical importance as long as only yearly average results are investigated.

No detailed geographical distribution of the emissions from leisure boats and “working” ships (service ships of different kinds, e.g., towing ships and icebreakers) was available; in the regional model simulations these emissions were distributed in the same way as the emissions from the ferries.

## Energy production

Emissions from the Energy production sector were treated in a simplified way in MATCH; typically these emissions occur from relatively high chimneys and in a *local scale* model they must be treated as point sources. In the *regional* MATCH model, emissions from this category are treated as area sources with an effective emission height distribution as given in Table 3 (SNAP sector 1). This distribution is based on studies of actual plumes and is considered reasonable for regional scale modelling in Europe (Simpson et al., 2003). Since the purpose of the regional scale modelling in this project is to estimate the impact of Stockholm sources outside the city we have used this simplified emission profile. For local scale studies (within the city) it is important to use the true emission heights for the individual chimneys.

The temporal variation of the emissions from Energy production in Greater Stockholm were based on data from Eneroth (2006) and are slightly different for the different species emitted from the sector; the seasonal variations are illustrated in Figure 6.



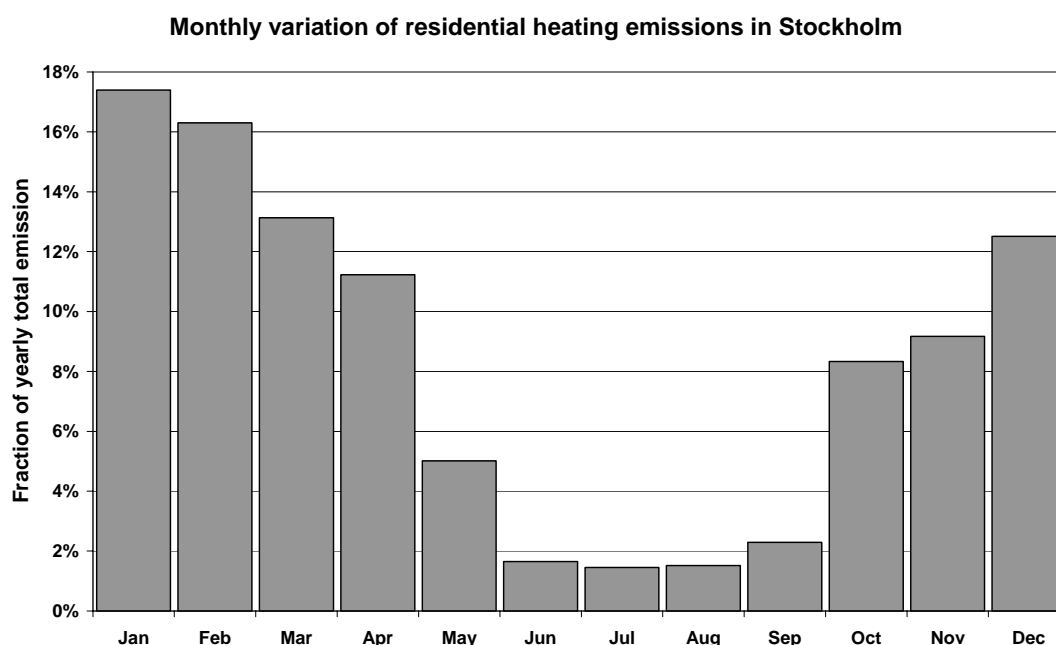
**Figure 6.** Variations of Stockholm power plant emissions with the month of the year. The fraction of the yearly total emission is shown for each month for NOx, Combustion particles, VOC and SOx. Unit: % of the yearly total emission.

## Residential heating

The emissions from residential heating are strongly dependent on temperature. In the MATCH simulations an assumed temporal variation depending on the temperature at 2-m height in Stockholm was used. The variation was simplified to only include variations on a monthly scale. The fraction of the yearly total emitted each month is shown in Figure 7. Emissions are largest in January and February and very low in June – September. The vertical distribution of the residential heating emissions in MATCH is treated in a simplified way with 50% of the emissions being released in the lowest model level and the remaining 50% in the second model level.

The emissions of sulphur dioxide from *wood burning* have been neglected due to lack of information.

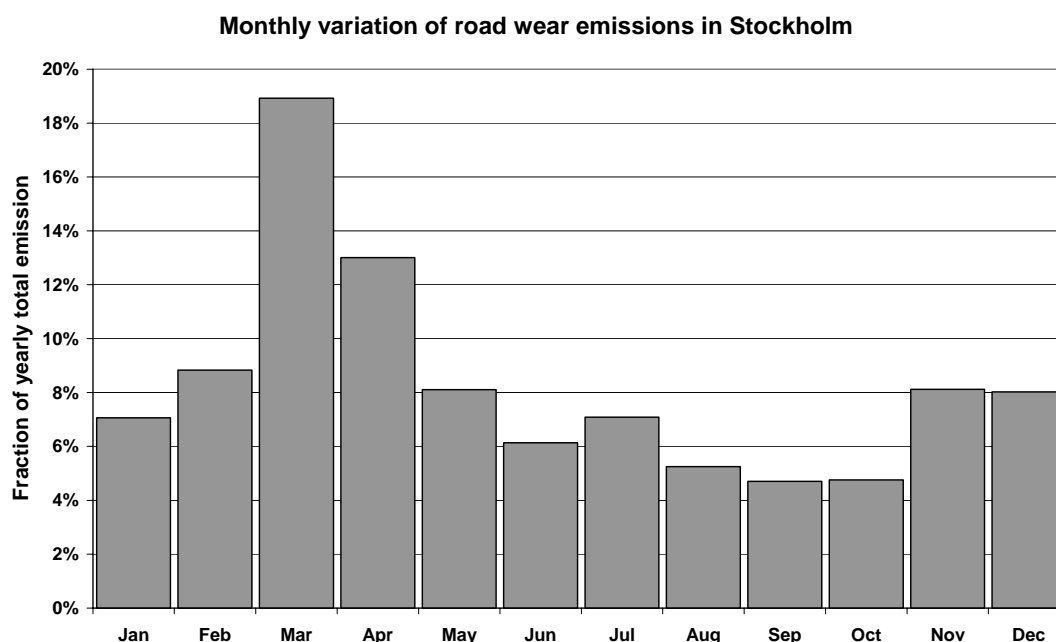
The emissions from residential biomass burning are very uncertain. In this study the emissions from Johansson and Eneroth (2007) were used. These were estimated using rather high emission factors for directly emitted PM from residential wood burning. Recently, the SMED consortium has published new emission estimates for the residential heating sector in Sweden (Paulrud et al., 2007). Their estimates of the direct PM emissions are only about 1/5 of the estimates from Johansson and Eneroth (2007). SMED also gives considerably lower NO<sub>x</sub> and SO<sub>2</sub>-emissions from residential heating in Stockholm (50-70% lower than the data used here).



**Figure 7.** Monthly variations of residential heating emissions in Stockholm used in the MATCH model. The fraction of the yearly total emission is shown for each month. Unit: % of the yearly total emission.

## Road traffic non-exhaust emissions

Emissions of particles due to road wear in Stockholm are much larger in winter and spring than in summer and early autumn, due to the use of studded tyres (and sanding of streets) in the winter season. In the MATCH simulations a seasonal variation was used for the wear particles, based on measurements in Stockholm for the years 2003-2005 (Johansson, 2007). The seasonal variation of the emissions is shown in Figure 8. Emissions are largest in March and April and lowest in August-October.



**Figure 8.** Monthly variations of road traffic non-exhaust (mostly road wear) emissions in Stockholm used in the MATCH model. The fraction of the yearly total emission is shown for each month. Unit: % of the yearly total emission.

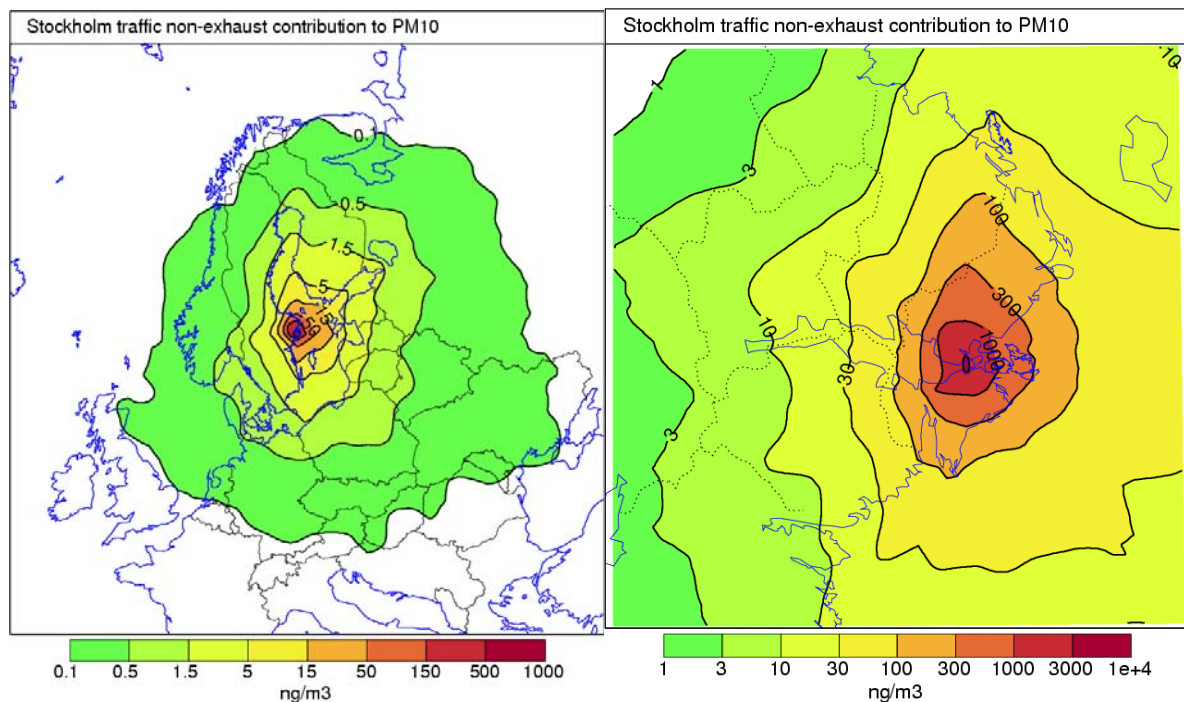
## Results and Discussion

### Concentrations – geographic distribution and contribution from different sectors

To estimate the impact of different emission sources in Stockholm a series of simulations were performed where one emission sector at a time was turned off. The results were then compared to the results from a simulation including all emissions. The emission sectors investigated in this way were: light duty vehicles (LDV), heavy duty vehicles (HDV), sea traffic (denoted Shipping in some maps), residential heating and power plants.

### Road traffic non-exhaust particles

Non-exhaust particles from road traffic (mostly road wear PM) were also simulated. The calculated contributions to the European and Swedish PM<sub>10</sub>-concentrations, from these relatively coarse particle emissions, are shown in Figure 9. The gradient from the emission area in Stockholm is very strong; the impacts outside Stockholm county are relatively small (below 0.3 µg/m<sup>3</sup> contribution to the yearly average PM<sub>10</sub> concentration).



**Figure 9.** Contribution from Traffic non-exhaust particles (mainly road wear) in Stockholm to yearly average PM10 concentrations. Left: Coarse scale simulation. Right: High resolution simulation. Unit:  $\text{ng/m}^3$  ( $10^{-9} \text{ g/m}^3$ ).

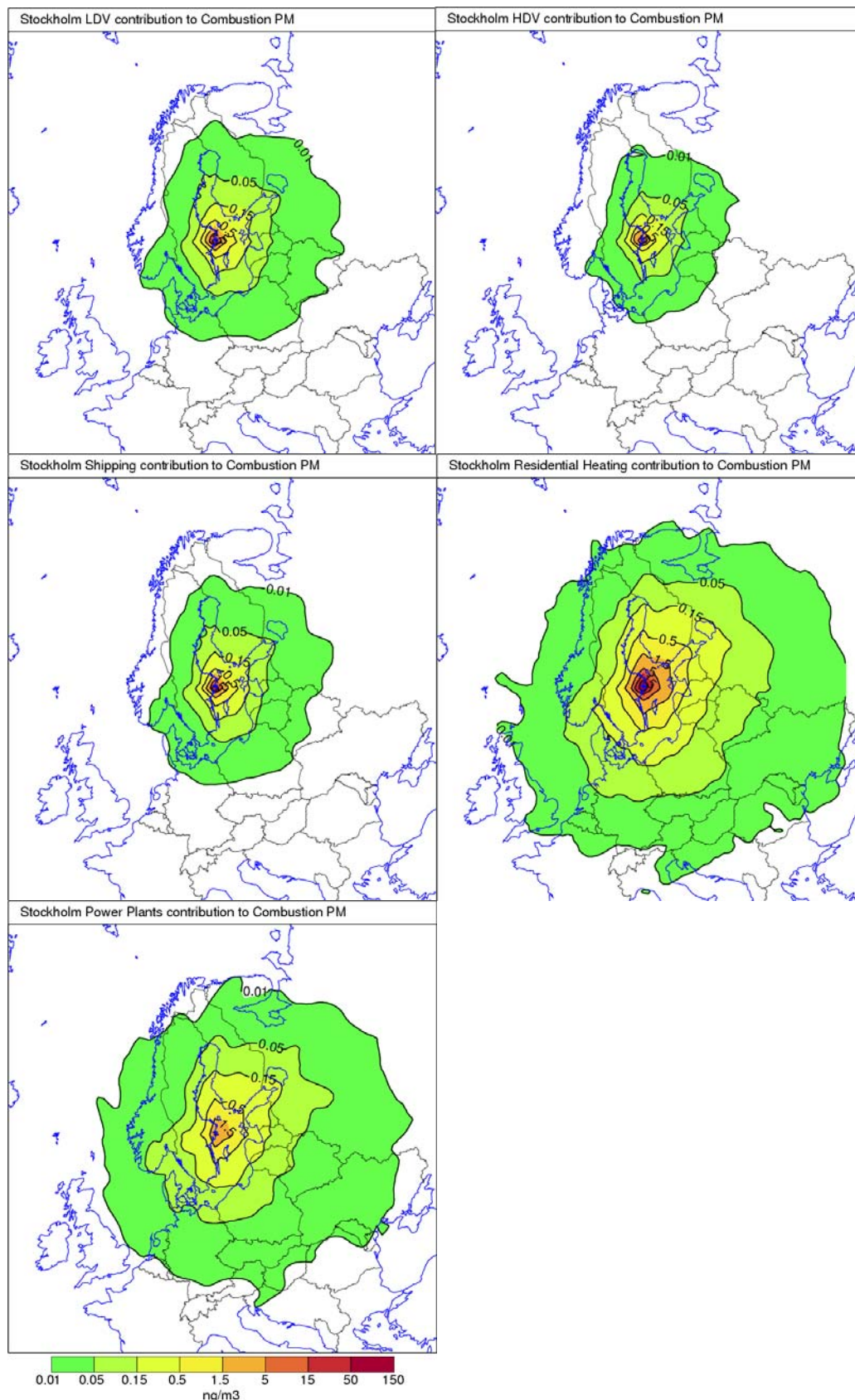
### Directly emitted combustion particles

Figures 10-15 show model calculated concentrations of directly emitted combustion particles and secondary particulate nitrate and sulphate. Results are shown both for the European scale and the Mälardalen scale; for ease of comparison each picture illustrates a particular pollutant for the five different emission sources.

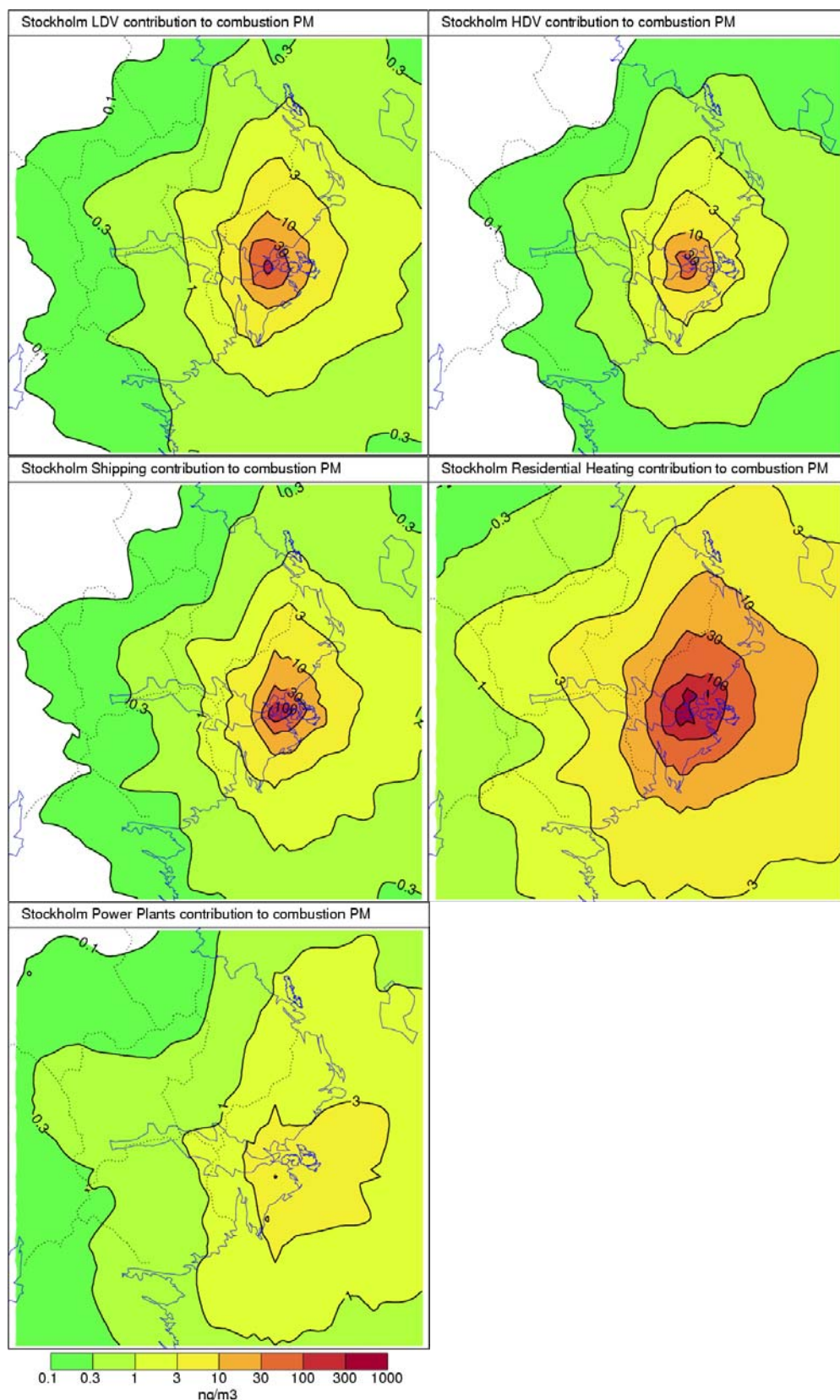
Due to the large emissions, residential heating gives the largest contributions to combustion particles; this is illustrated in Figures 10 and 11. As pointed out earlier, the uncertainties in the emission estimates for residential wood combustion are very large and the true emissions may be much lower than the ones used here<sup>3</sup>. Power plants give a low contribution close to Stockholm but, compared to the other emission sources, relatively large contribution far from the city; this is due to the high effective emission height used for this source sector.

<sup>3</sup> The emission estimates from Paulrud et al. (2007) are about 1/5 of the ones used here and this could be used as a rough estimate of the magnitude of the uncertainty for the residential heating source.





**Figure 10.** Modelled contribution to yearly average combustion particle concentrations due to different sources in Greater Stockholm in the European scale simulations. Upper Left: contribution from Light Duty Vehicles (LDV); Upper Right: contribution from Heavy Duty Vehicles (HDV); Middle Left: contribution from Sea Traffic; Middle Right: contribution from Residential Heating; Lower Left: contribution from Power Plants. Unit:  $\text{ng}/\text{m}^3$ .

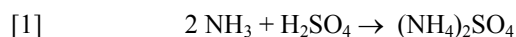


**Figure 11.** Modelled contribution to yearly average combustion particle concentrations due to different sources in Greater Stockholm in the high resolution model. Upper Left: contribution from Light Duty Vehicles (LDV); Upper Right: contribution from Heavy Duty Vehicles (HDV); Middle Left: contribution from Sea Traffic; Middle Right: contribution from Residential Heating; Lower Left: contribution from Power Plants. Unit:  $\text{ng}/\text{m}^3$ .

## Secondarily formed particulate matter due to emissions in Stockholm

The impact of the Stockholm emissions on particulate nitrate and sulphate are more complicated than the directly emitted combustion and wear particles. Since nitrate and sulphate are formed through non-linear chemical reactions, both increases and decreases are possible due to the different substances emitted in Stockholm.

For nitrate, the Stockholm emissions lead to increased concentrations far from the city (Figure 12) but, except for the road traffic sources, the *local* nitrate concentrations decrease (Figure 13); the local decrease is due to the relatively large sulphur emissions from sea traffic, residential heating and power plants. The emitted sulphur is partly in the form of sulphuric acid and this reacts with available ammonia (which may come from agricultural, industrial and traffic sources) to form ammonium sulphate:



This leads to a loss of available ammonia to form ammonium nitrate via the equilibrium reaction:



The loss of  $\text{NH}_3$  pushes the equilibrium towards more  $\text{HNO}_3(\text{g})$ , instead of particulate nitrate, and thereby the local effect of the sulphur rich emissions can be a decrease in particulate nitrate. However, this decrease in nitrate is countered by an increase in particulate sulphate.

It is interesting to note that the LDVs in Stockholm contribute much more to particulate nitrate formation than the HDVs.  $\text{NO}_x$  emissions are about equal from the two sectors but the LDVs also emit  $\text{NH}_3$  that can react with  $\text{HNO}_3$  to form ammonium nitrate [2].

The road-traffic exhaust emissions (LDV and HDV) contain very little sulphur compared to  $\text{NO}_x$ . This leads to increased nitrate concentrations due to road traffic both locally and at longer range (Figure 12 and 13).

For HDV the sulphur emissions are in fact so low (only 1.71 tons/year) that the calculated net effect of the HDV-emissions on *sulphate* concentrations is a small *decrease* (see Figures 14 and 15). The reason for the negative impact on particulate sulphate concentrations from the HDV traffic in Stockholm is that the *yearly average* impact of the large  $\text{NO}_x$ -emissions from this sector (ca 2645 tons/year) is to reduce the oxidation rate of gaseous  $\text{SO}_2$  to particulate sulphate; the reduction is due to the reaction of the emitted NO with ozone ( $\text{O}_3$ ):

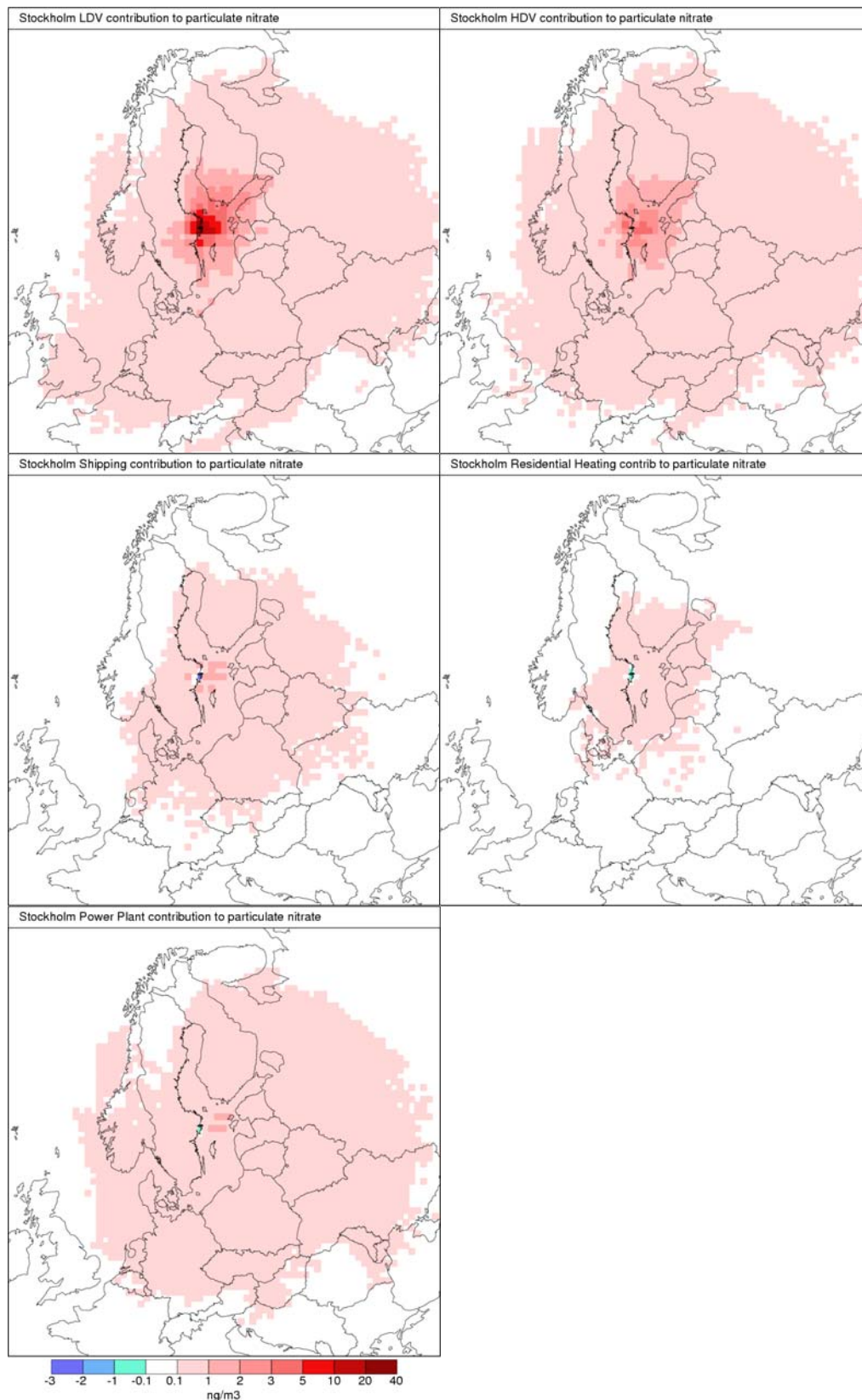


Ozone is a rather important oxidant for  $\text{SO}_2$  and also for generating other oxidants (OH and  $\text{H}_2\text{O}_2$ ) that produce sulphate from  $\text{SO}_2$ . The reduction in oxidation rate is larger than the small addition of  $\text{SO}_2$  and sulphate from the HDVs in Stockholm; this leads to somewhat lower particulate sulphate concentrations (but at the same time slightly higher  $\text{SO}_2$  concentrations) because of the total HDV emissions in Stockholm.

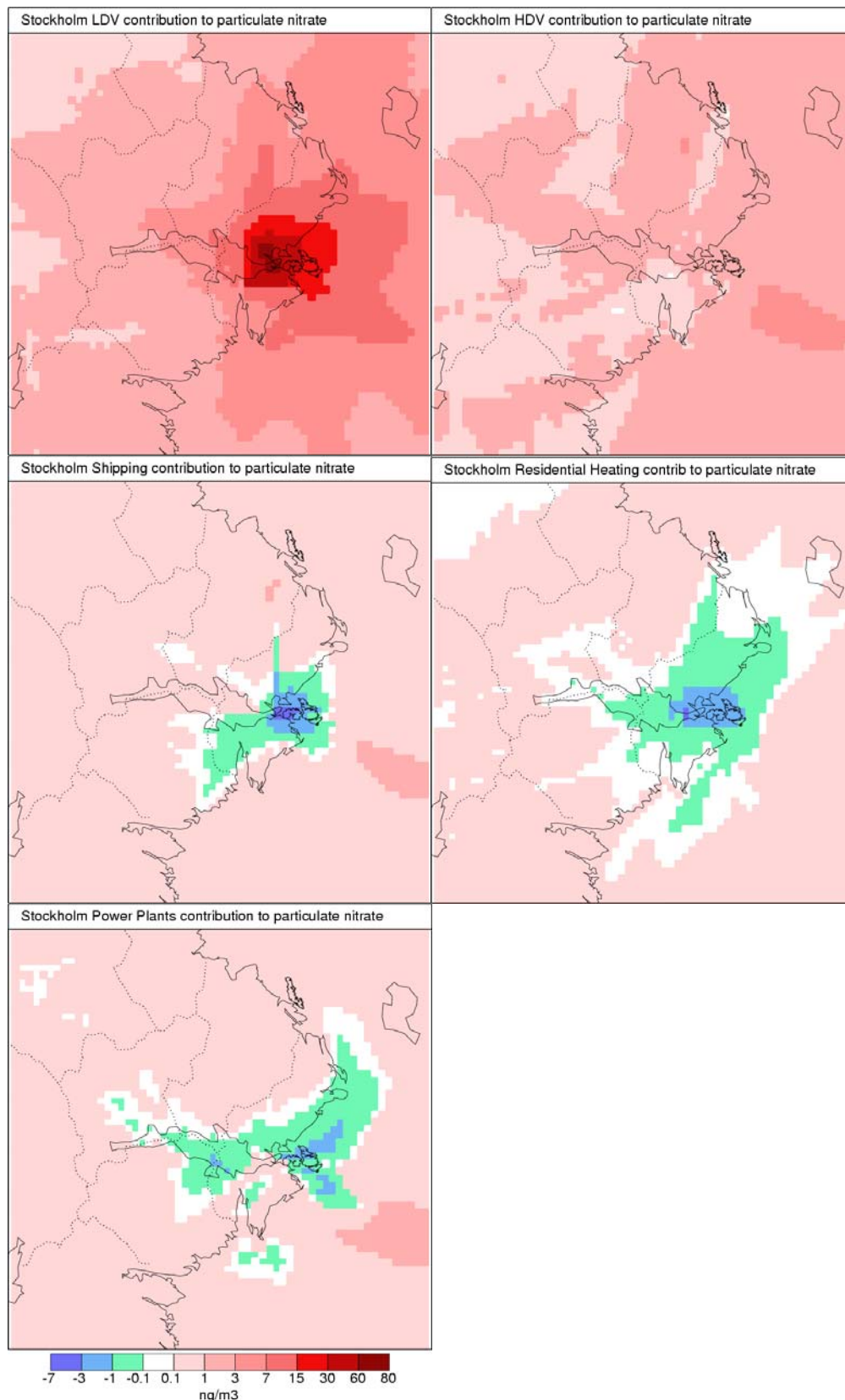
Due to the higher sulphur content in gasoline, compared to diesel, LDV emissions of sulphur are more than 20 times larger than those from HDVs (but still very small compared to the emissions from sea traffic, residential heating and power plants). The larger sulphur emissions from the LDVs compensate the reduced oxidation rate and the, yearly average, net effect is a very small increase in sulphate due to LDV emissions in Stockholm.

Emissions of  $\text{NO}_x$  and  $\text{SO}_x$  also contribute to the formation of particulate ammonium ( $\text{NH}_4^+$ ) from gaseous ammonia ( $\text{NH}_3$ ) via reactions [1] and [2]. The impacts of the different emission sources in Stockholm on particulate ammonium concentrations have been calculated and the results are illustrated in Figures 16 and 17. Most sources have a relatively small, and rather short-ranged, impact on the  $\text{NH}_4^+$  concentrations. The exception is the LDV traffic that emits relatively much  $\text{NH}_3$  and thereby contributes to particulate  $\text{NH}_4\text{NO}_3$  formation.

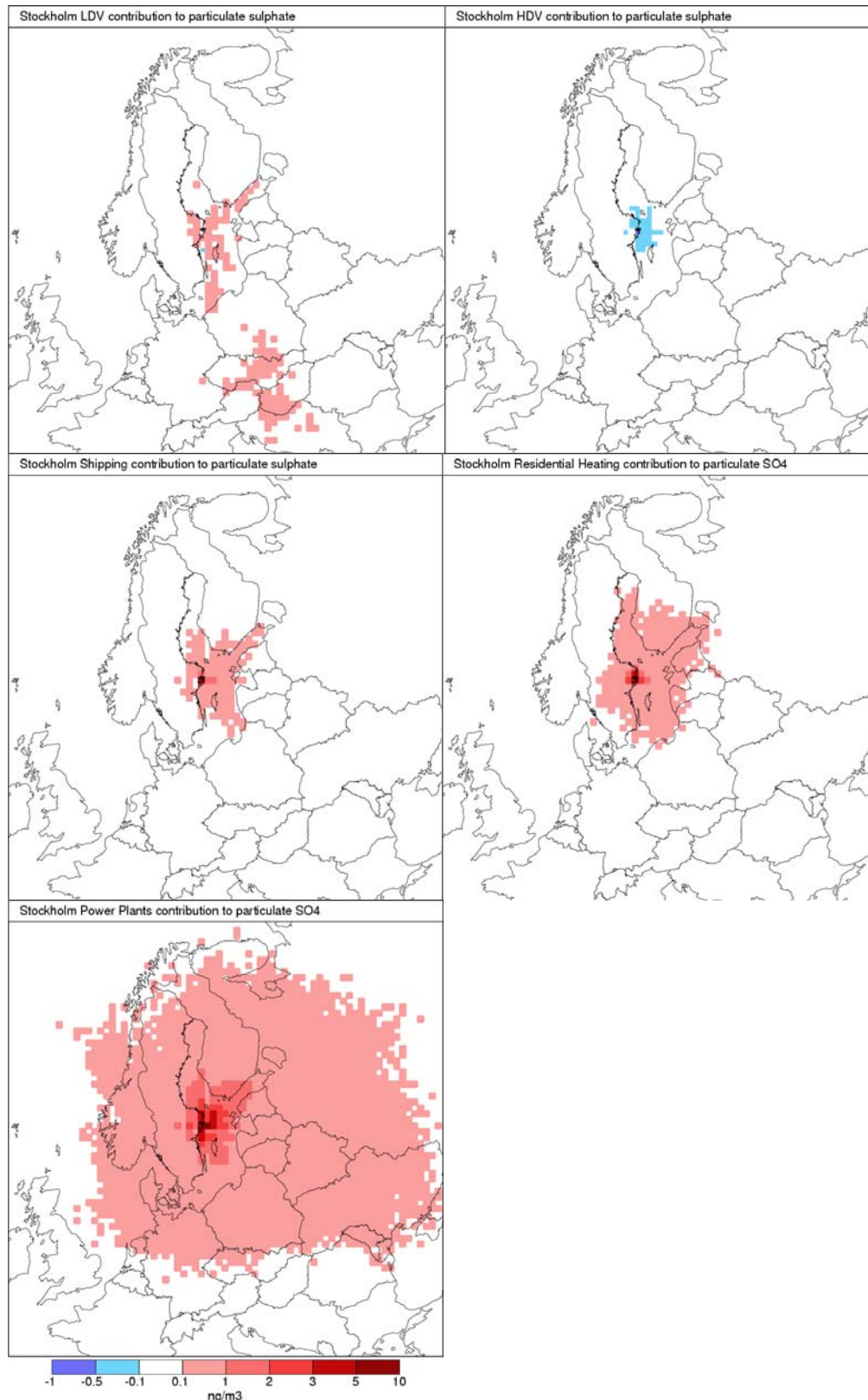




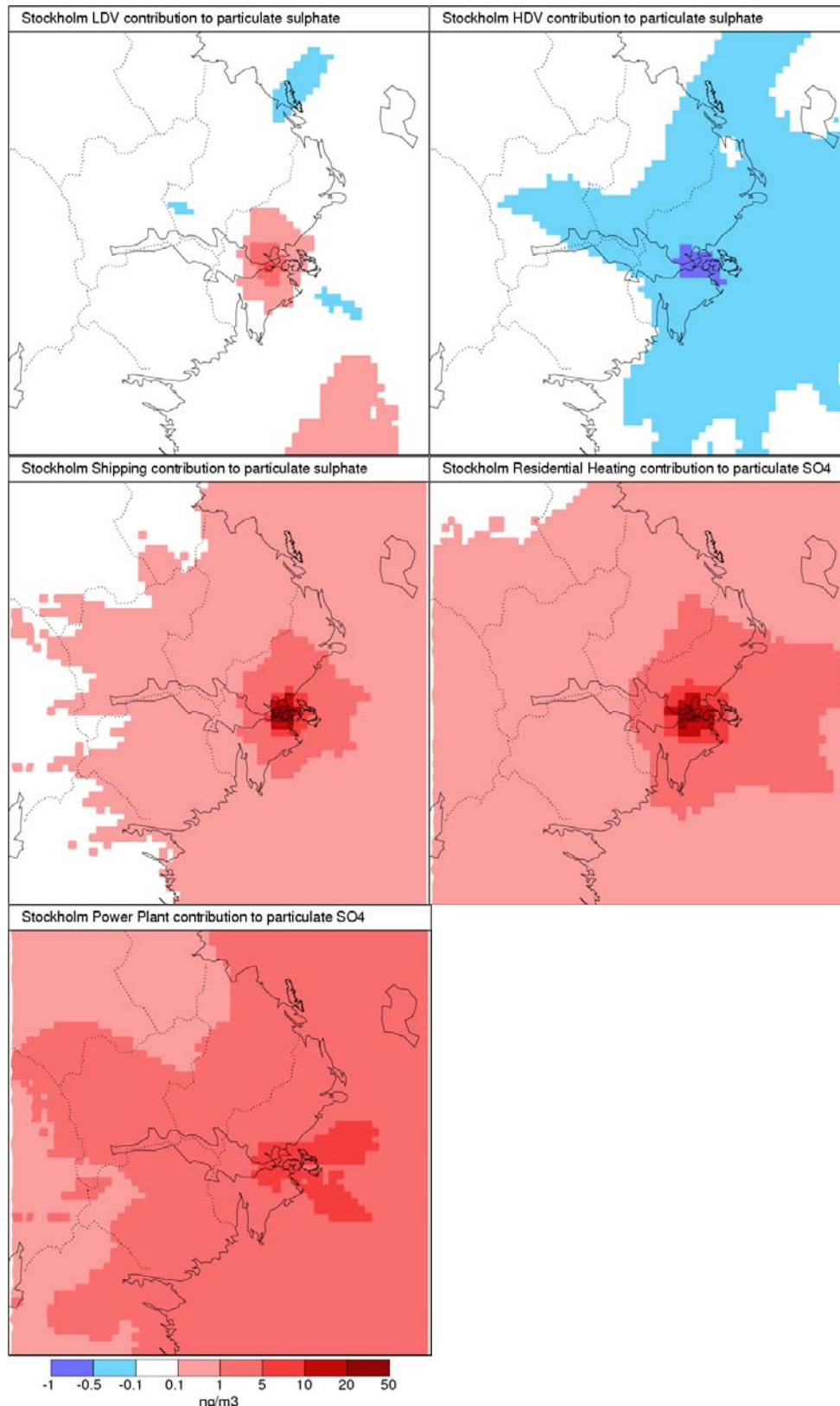
**Figure 12.** Modelled impact on yearly average particulate nitrate concentrations due to different emission sources in Greater Stockholm in the European scale simulations. Blue colours indicate decreases in nitrate due to Stockholm emissions. Upper Left: contribution from Light Duty Vehicles (LDV); Upper Right: contribution from Heavy Duty Vehicles (HDV); Middle Left: contribution from Sea Traffic; Middle Right: contribution from Residential Heating; Lower Left: contribution from Power Plants. Unit: ng/m<sup>3</sup>.



**Figure 13.** Modelled impact on yearly average particulate nitrate concentrations due to different emission sources in Greater Stockholm in the high resolution simulations. Blue/Green colours indicate decreases in nitrate due to Stockholm emissions. Upper Left: contribution from Light Duty Vehicles (LDV); Upper Right: contribution from Heavy Duty Vehicles (HDV); Middle Left: contribution from Sea Traffic; Middle Right: contribution from Residential Heating; Lower Left: contribution from Power Plants. Unit:  $\text{ng/m}^3$ .

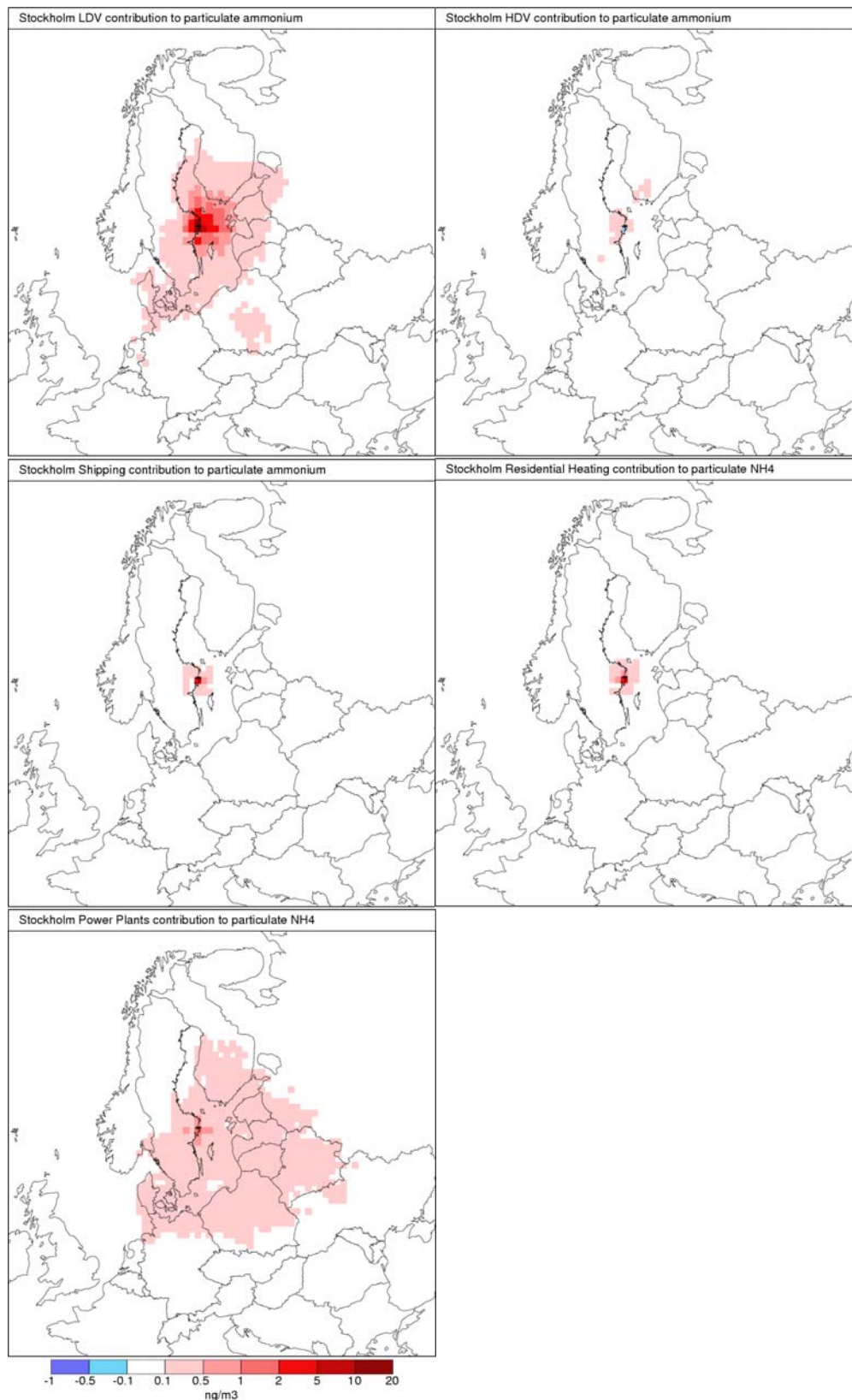


**Figure 14.** Modelled impact on yearly average particulate sulphate concentrations due to different emission sources in Greater Stockholm in the European scale simulations. Blue colours indicate decreases in sulphate due to Stockholm emissions. Upper Left: contribution from Light Duty Vehicles (LDV); Upper Right: contribution from Heavy Duty Vehicles (HDV); Middle Left: contribution from Sea Traffic; Middle Right: contribution from Residential Heating; Lower Left: contribution from Power Plants. Unit: ng/m<sup>3</sup>.



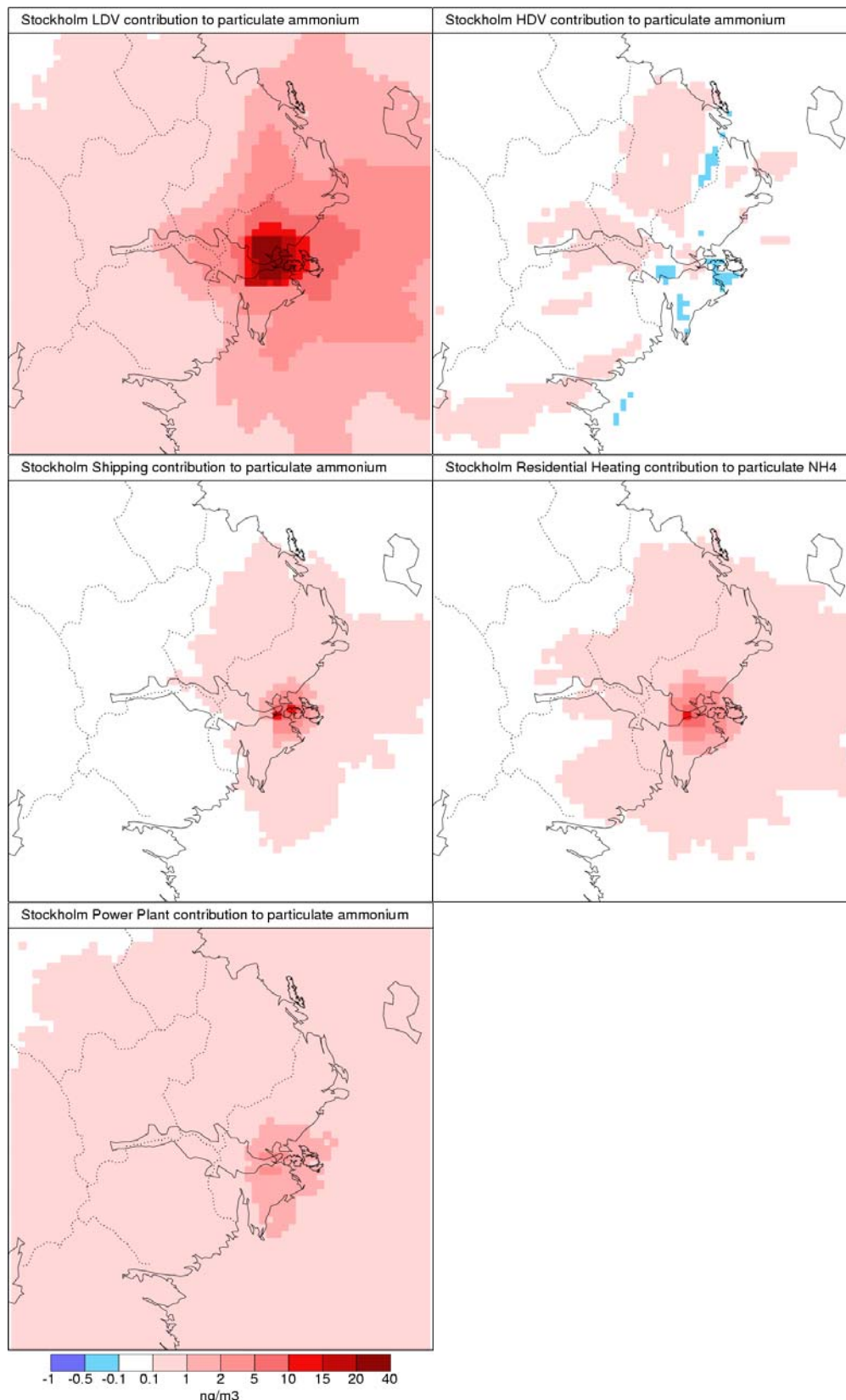
**Figure 15.** Modelled impact on yearly average particulate sulphate concentrations due to different emission sources in Greater Stockholm in the high resolution simulations. Blue colours indicate decreases in sulphate due to Stockholm emissions. Upper Left: contribution from Light Duty Vehicles (LDV); Upper Right: contribution from Heavy Duty Vehicles (HDV); Middle Left: contribution from Sea Traffic; Middle Right: contribution from Residential Heating; Lower Left: contribution from Power Plants. Unit:  $\text{ng}/\text{m}^3$ .





**Figure 16.** Modelled impact on yearly average particulate ammonium concentrations due to different emission sources in Greater Stockholm in the European scale simulations. Blue colours indicate decreases in particulate ammonium due to Stockholm emissions. Upper Left: contribution from Light Duty Vehicles (LDV); Upper Right: contribution from Heavy Duty Vehicles (HDV); Middle Left: contribution from Sea Traffic; Middle Right: contribution from Residential Heating; Lower Left: contribution from Power Plants. Unit:  $\text{ng/m}^3$ .

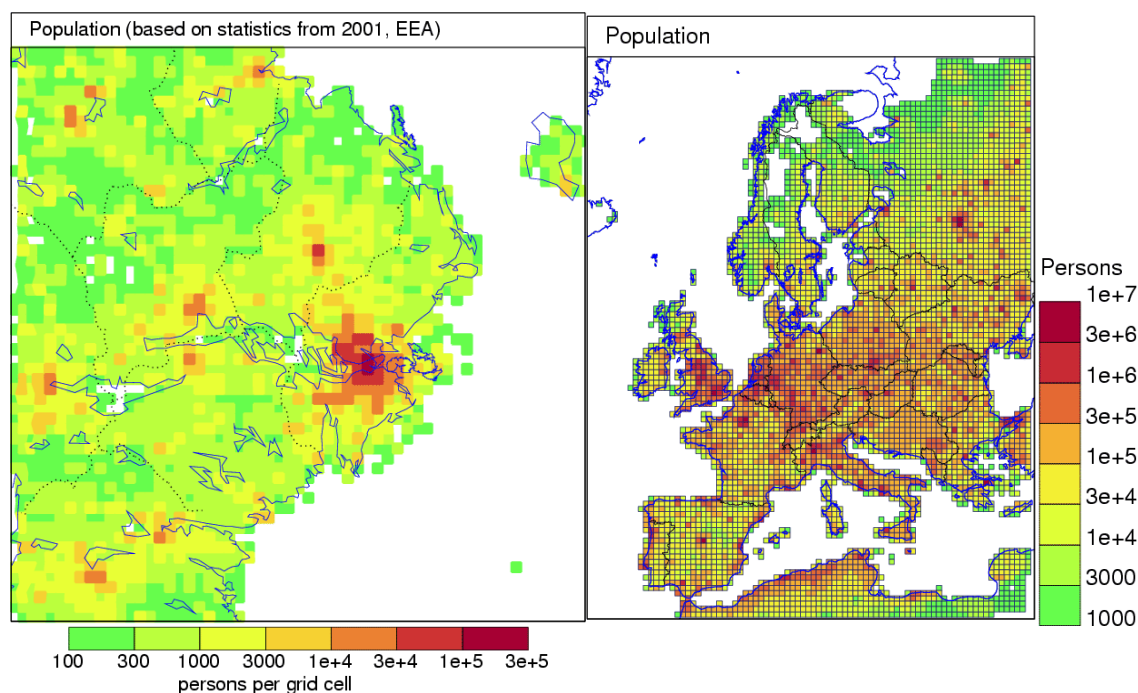




**Figure 17.** Modelled impact on yearly average particulate ammonium concentrations due to different emission sources in Greater Stockholm in the high resolution simulations. Blue colours indicate decreases in particulate ammonium due to Stockholm emissions. Upper Left: contribution from Light Duty Vehicles (LDV); Upper Right: contribution from Heavy Duty Vehicles (HDV); Middle Left: contribution from Sea Traffic; Middle Right: contribution from Residential Heating; Lower Left: contribution from Power Plants. Unit: ng/m<sup>3</sup>.

## Population exposure

Population data with approximately 1km resolution was used for the EU countries and Croatia; the data were produced by the Joint Research Centre (JRC) for EEA and are based on population figures for 2001 (EEA/JRC, 2006). For countries outside EU population data were taken from the Columbia University data base “Gridded Population of the World, version 3 (GPWv3)” (CIESIN, 2005); these data are based on population statistics for the year 2000. The population densities, used in the calculations of human exposure, are illustrated in Figure 18.



**Figure 18.** Population data used in the exposure calculations. Left: High resolution domain, data taken from EEA/JRC (2006) (based on 2001 statistics). Right: European scale domain, data taken from EEA/JRC (2006) for the EU countries (+Croatia) and from CIESIN (2005) for the rest of the domain. Unit: persons per grid cell.

## Directly emitted combustion particles

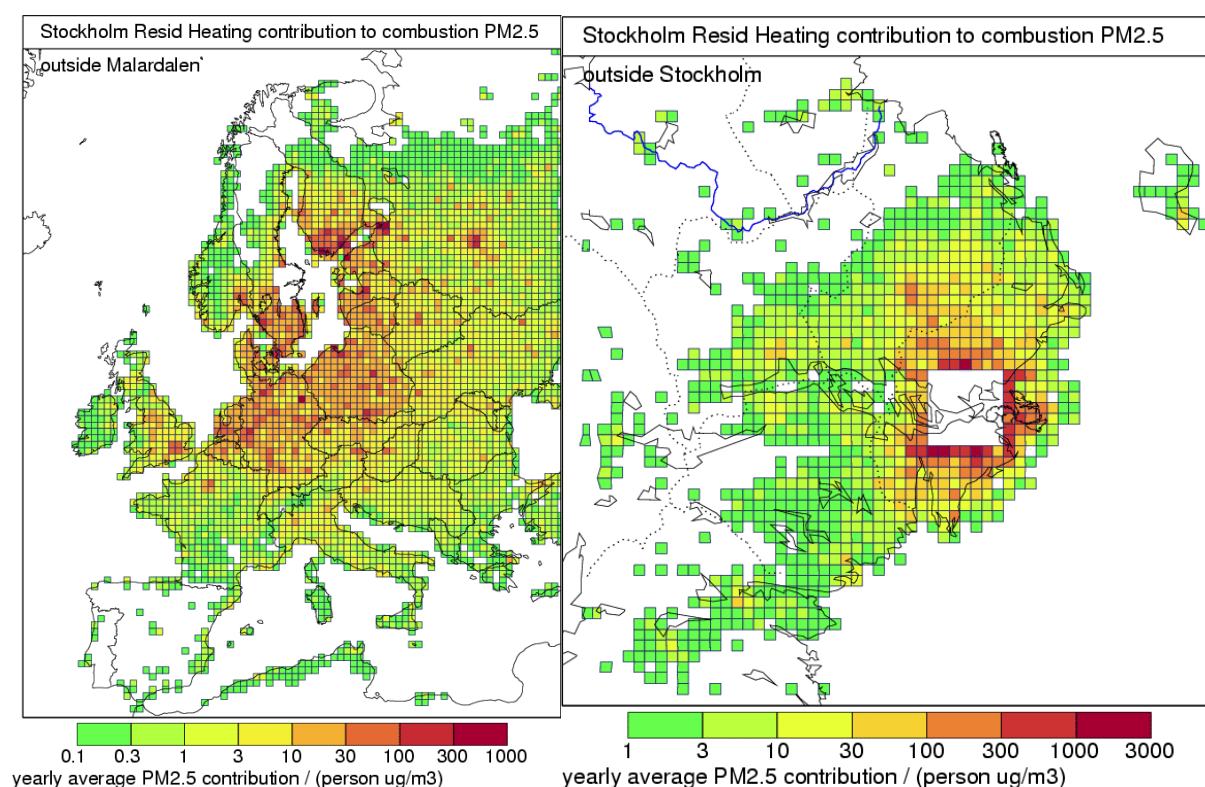
An example of the exposure results is given in Figure 19, which shows the calculated yearly average of the population exposure to directly emitted combustion particles from residential heating in Stockholm. Results from the European scale model and from the High resolution model are shown in the figure. On the European scale (outside Mälardalen) the Stockholm emissions have the largest population impact in the populated areas around the Baltic Sea and some larger cities relatively close to Sweden, such as, e.g., Berlin.

In Tables 4-8 the model calculated population exposures to various particle components due to emissions from the different sources in the Greater Stockholm area are given. Since exposure calculations were performed in three different model resolutions in TESS, results are presented for exposure within the Greater Stockholm area, within the Mälardalen area (except Greater Stockholm) and within Europe (except Mälardalen).

The population exposures to directly emitted combustion and wear particles *inside the emission area* are taken from Johansson and Eneroth (2007), assuming a population in the area of 1 405 600. The calculations of Johansson and Eneroth did not include secondary inorganic aerosols and therefore the contributions to these were calculated in this study, using results from the Mälardalen model domain simulations.

For directly emitted *combustion* particles the residential heating emissions dominate the population exposure, with ca 74% of the total calculated impact of the Stockholm sources, when the emissions from Johansson and Eneroth (2007) are used. Estimates of what the population exposures would have been with the SMED emissions (Paulrud et al., 2007) are also included in Table 4. Using these estimates the residential heating contribution to population exposure would be of about the same magnitude (37%) as the contribution from road traffic exhaust (42%). Even if the uncertainties in the emission estimates for Residential Heating are very large it seems that this is a very important PM source in Stockholm. More work is needed to quantify these emissions more accurately.

For residential heating more than 90% of the exposure occurs within the Greater Stockholm area. For all sources, except Sea Traffic, the total population exposure to combustion PM is much larger within Stockholm than outside; for shipping the total exposure is about as large outside the city as within.



**Figure 19.** Yearly average population exposure to directly emitted combustion particles from Residential Heating in Stockholm. Left: Coarse scale simulation for Europe, the Mälardalen region has been masked (since it is treated in more detail in the higher resolution simulation). Right: The high-resolution simulation for the Mälardalen region; the Stockholm region has been masked (results for this region are taken from the local scale study of Johansson & Eneroth, 2007). Unit: person  $\mu\text{g}/\text{m}^3$ .

**Table 4.** Population exposure to combustion particles, due to emissions in Greater Stockholm (Yearly average, unit: person  $\mu\text{g}/\text{m}^3$ )

REGION\SOURCE	ROAD TRAFFIC TOTAL	ROAD TRAFFIC, LDV <sup>A</sup>	ROAD TRAFFIC, HDV <sup>B</sup>	SEA TRAFFIC	POWER PLANTS	RESIDENTIAL HEATING
Greater Stockholm	192 000 <sup>c</sup>	141 000 <sup>c</sup>	50 600 <sup>c</sup>	8 860 <sup>c</sup>	71 700 <sup>c</sup>	ca167 000 <sup>d</sup> - 829 000 <sup>c</sup>
Mälardalen, except Greater Stockholm	7 560	5 000	2 560	4 690	2 190	ca6 000 <sup>e</sup> - 31 800 <sup>f</sup>
Europe, except Mälardalen	6 160	4 150	2 010	3 430	12 900	ca4 700 <sup>e</sup> - 25 200 <sup>f</sup>
Europe, total population exposure	205 000	150 000	55 200	17 000	86 800	ca178 000 <sup>d,e</sup> - 886 000 <sup>c,f</sup>

<sup>A</sup> LDV = light duty vehicles

<sup>B</sup> HDV = heavy duty vehicles

<sup>c</sup> from Johansson & Eneroth (2007), assumed population within the Greater Stockholm domain 1 405 600 persons

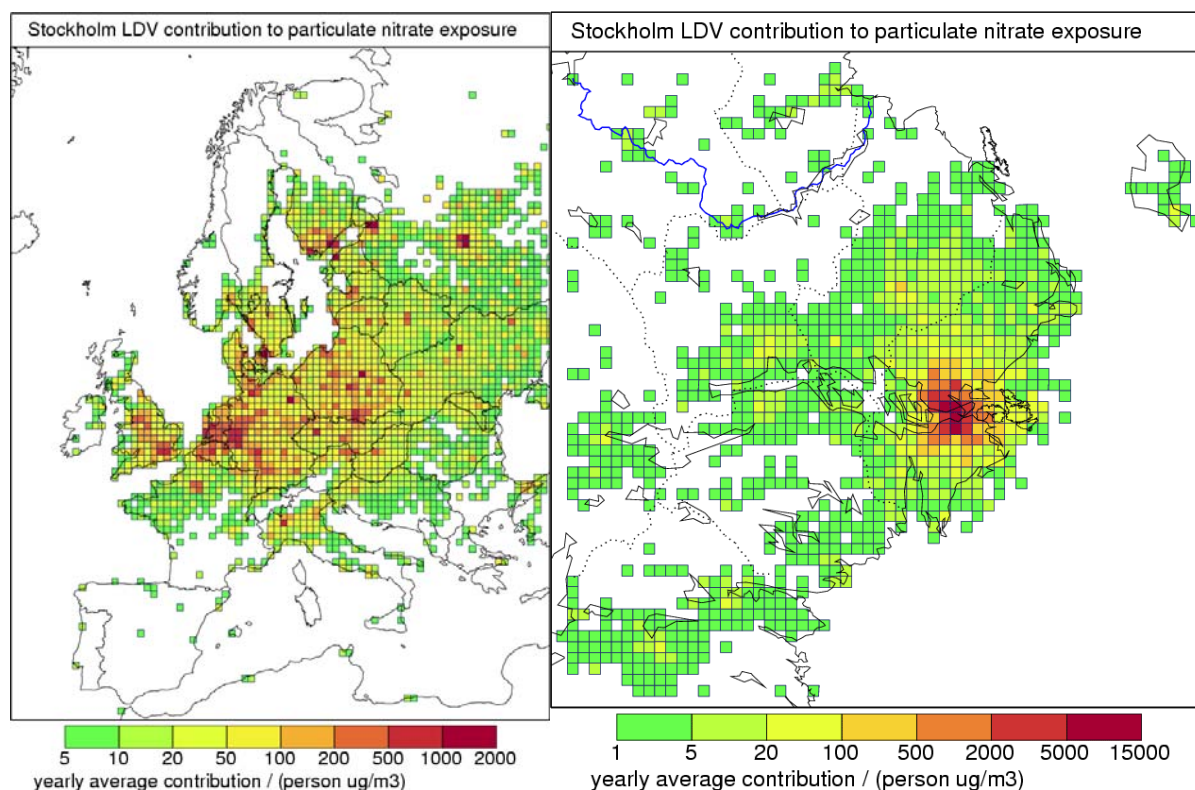
<sup>d</sup> based on scaling of the results from Johansson & Eneroth (2007), using the SMED estimated emissions, which are 1/5 of the emissions used in this study

<sup>e</sup> based on scaling of the results from this study, using the SMED estimated emissions, which are 1/5 of the emissions used in this study

<sup>f</sup> this study, based on emissions from Johansson & Eneroth (2007)

## Secondary Inorganic Aerosols

An example of the SIA exposure (nitrate) due to road traffic emissions (LDV) in Stockholm is shown in Figure 20.



**Figure 20.** Yearly average population exposure to particulate nitrate due to emissions from Light Duty Vehicles (LDV) in Stockholm. Left: Coarse scale simulation for Europe, the Mälardalen region has been masked (since it is treated in more detail in the higher resolution simulation). Right: The high-resolution simulation for the Mälardalen region. Unit: Person  $\mu\text{g}/\text{m}^3$ .

The calculated total population exposures to the different SIA components are given in Tables 5-7.

The total population exposure to secondary inorganic aerosols, due to the sum of the emissions in Stockholm, is considerably lower than the exposure to directly emitted combustion particles. This is, however, only due to the very large estimated direct emissions from residential heating used here. For all other emission sources the SIA exposure is higher than the direct PM exposure. For HDV exposure to SIA is ca 111% higher than to the directly emitted exhaust PM and for LDV exposure to SIA is about 24% higher than to direct exhaust PM. For shipping emissions and power plant emissions the calculated total SIA exposures are also much higher than the directly emitted PM exposures.

**Table 5.** Population exposure to particulate nitrate ( $\text{NO}_3^-$ ), due to emissions in Greater Stockholm (Yearly average, unit: person  $\mu\text{g}/\text{m}^3$ )

REGION\SOURCE	ROAD TRAFFIC TOTAL	ROAD TRAFFIC, LDV <sup>A</sup>	ROAD TRAFFIC, HDV <sup>B</sup>	SEA TRAFFIC	POWER PLANTS	RESIDENTIAL HEATING <sup>C</sup>
Greater Stockholm	78 900	76 800	2 110	-1 320 <sup>D</sup>	69	-2 730 <sup>C,D</sup>
Mälardalen, except Greater Stockholm	11 400	8 960	2 400	434	579	69 <sup>C</sup>
Europe, except Mälardalen	242 000	135 000	108 000	42 000	81 600	21 700 <sup>C</sup>
Europe, total population exposure	333 000	221 000	112 000	41 100	82 200	19 000 <sup>C</sup>

<sup>A</sup> LDV = light duty vehicles

<sup>B</sup> HDV = heavy duty vehicles

<sup>C</sup> The  $\text{NO}_x$  (and  $\text{SO}_2$ ) emissions from residential heating are very uncertain which means that the population exposure values for this sector are uncertain as well.

<sup>D</sup> The reason for the *decrease* in particulate nitrate within the Greater Stockholm region due to sea traffic and residential heating is that these sources emit fairly large amounts of  $\text{SO}_x$ , which reacts irreversibly with available ammonia to form ammonium sulphate:  $2 \text{NH}_3 + \text{H}_2\text{SO}_4 \rightarrow (\text{NH}_4)_2\text{SO}_4$ ; this leads to a loss of available ammonia to form ammonium nitrate via the equilibrium reaction  $\text{NH}_3(\text{g}) + \text{HNO}_3(\text{g}) \leftrightarrow \text{NH}_4\text{NO}_3(\text{s, aq})$ . The loss of  $\text{NH}_3$  pushes the equilibrium towards more  $\text{HNO}_3(\text{g})$  instead of particulate nitrate.

**Table 6.** Population exposure to particulate sulphate ( $\text{SO}_4^{2-}$ ), due to emissions in Greater Stockholm (Yearly average, unit: person  $\mu\text{g}/\text{m}^3$ )

REGION\SOURCE	ROAD TRAFFIC TOTAL	ROAD TRAFFIC, LDV <sup>A</sup>	ROAD TRAFFIC, HDV <sup>B</sup>	SEA TRAFFIC	POWER PLANTS	RESIDENTIAL HEATING <sup>C</sup>
Greater Stockholm	2 220	3 030	-810 <sup>D</sup>	10 300	8 650	24 200 <sup>C</sup>
Mälardalen, except Greater Stockholm	-177	62	-239 <sup>D</sup>	1 080	3 880	1 960 <sup>C</sup>
Europe, except Mälardalen	19 000	23 900	-4 890 <sup>D</sup>	7 330	66 500	11 500 <sup>C</sup>
Europe, total population exposure	21 100	27 000	-5 940 <sup>D</sup>	18 700	79 000	37 600 <sup>C</sup>

<sup>A</sup> LDV = light duty vehicles

<sup>B</sup> HDV = heavy duty vehicles

<sup>C</sup> The  $\text{SO}_2$  (and  $\text{NO}_x$ ) emissions from residential heating are very uncertain which means that the population exposure values for this sector are uncertain as well.

<sup>D</sup> The reason for the *decrease* in particulate sulphate due to the HDV traffic in Stockholm is that the sulphur emissions from this category are extremely low, only 1.71 tons/year, which is insignificant compared to the emissions of  $\text{NO}_x$  from the same sector, ca 2645 tons/year. The yearly average impact of the large  $\text{NO}_x$ -emissions from HDV is to reduce the oxidation rate of gaseous  $\text{SO}_2$  to particulate sulphate; the reduction is due to the reaction of the emitted  $\text{NO}$  with ozone,  $\text{NO} + \text{O}_3 \rightarrow \text{NO}_2 + \text{O}_2$ ;  $\text{O}_3$  is a rather important oxidant for  $\text{SO}_2$  and also for producing the other oxidants that efficiently produce sulphate. The reduction in oxidation rate is larger than the small addition of  $\text{SO}_2$  and sulphate from the HDVs in Stockholm; this leads to somewhat lower particulate sulphate concentrations (but at the same time slightly higher  $\text{SO}_2$  concentrations).

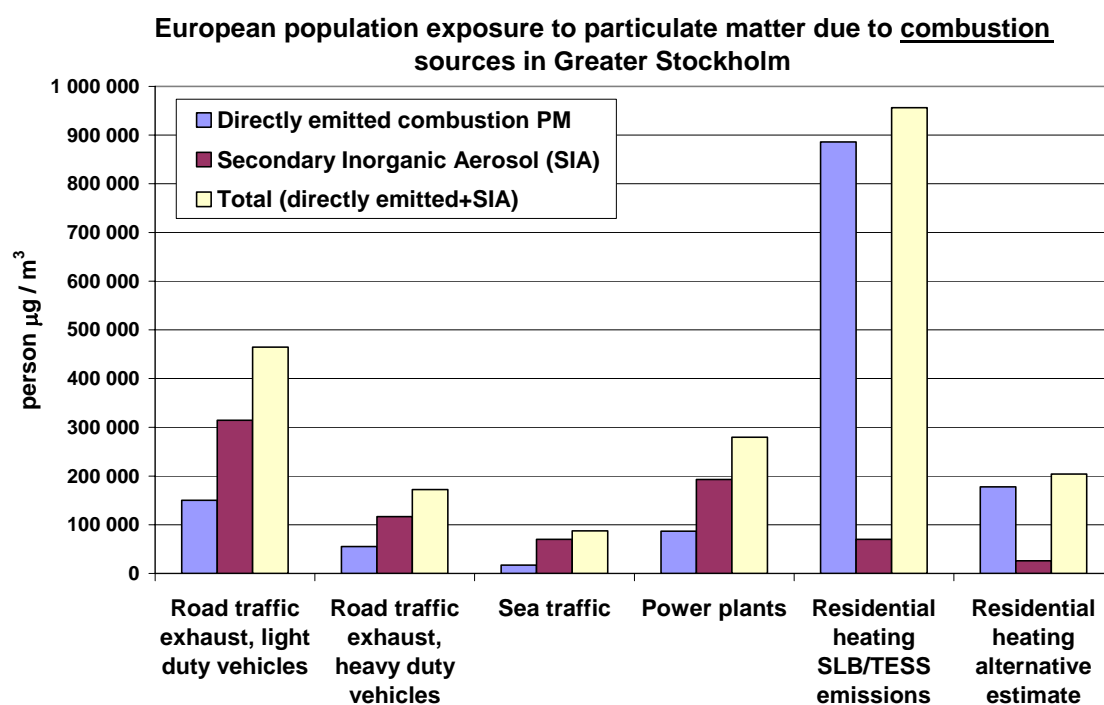
**Table 7.** Population exposure to particulate ammonium ( $\text{NH}_4^+$ ), due to emissions in Greater Stockholm (Yearly average, unit: person  $\mu\text{g}/\text{m}^3$ )

REGION\SOURCE	ROAD TRAFFIC TOTAL	ROAD TRAFFIC, LDV <sup>A</sup>	ROAD TRAFFIC, HDV <sup>B</sup>	SEA TRAFFIC	POWER PLANTS	RESIDENTIAL HEATING <sup>C</sup>
Greater Stockholm	36 000	35 800	196	2 890	2 690	7 280 <sup>C</sup>
Mälardalen, except Greater Stockholm	3 430	3 280	154	352	977	570 <sup>C</sup>
Europe, except Mälardalen	38 000	27 900	10 100	7 070	27 900	5 870 <sup>C</sup>
Europe, total population exposure	77 500	67 000	10 500	10 300	31 600	13 700 <sup>C</sup>

<sup>A</sup> LDV = light duty vehicles

<sup>B</sup> HDV = heavy duty vehicles

<sup>C</sup> The NO<sub>x</sub> and SO<sub>2</sub> emissions from residential heating are very uncertain which means that the population exposure values for this sector are uncertain as well.



**Figure 21.** Contributions of different sources in the Greater Stockholm area to the yearly average population exposure to combustion particles and secondary inorganic particles in Europe. For the residential heating source, two different estimates are given, one based on the emissions from Johansson and Eneroth (2007) and one based on emissions from SMED. Unit: person  $\mu\text{g}/\text{m}^3$ .



### Non-exhaust particles and total PM10 and PM2.5 exposure

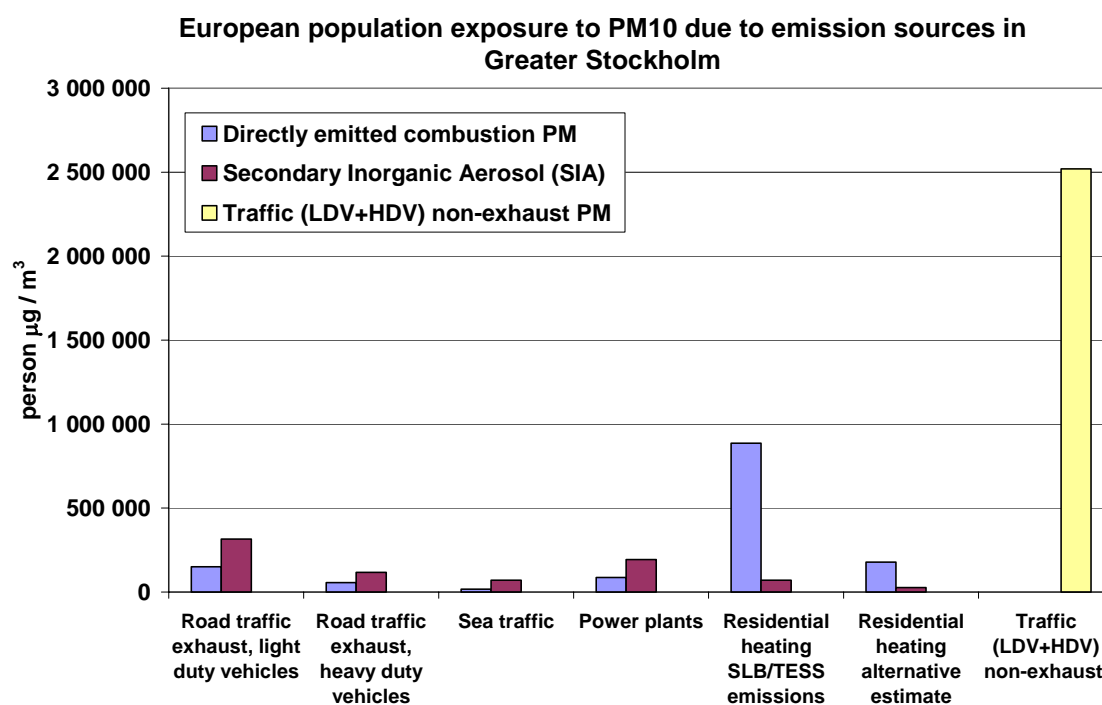
Non-exhaust particles dominate the total impact on PM10 exposure, contributing about 60-70% to the total exposure, due to the use of studded tires in Stockholm, as can be seen in Figure 22 (and Table 8). The calculated population exposure due to the non-exhaust particles is to a very large extent (87%) occurring within the Greater Stockholm area.

**Table 8.** Population exposure to non-exhaust particles, due to emissions in Greater Stockholm (Yearly average, unit: person  $\mu\text{g}/\text{m}^3$ )

REGION\SOURCE	TOTAL	COARSE FRACTION	FINE FRACTION (PM2.5)
Greater Stockholm	2 190 000 <sup>a</sup>	1 740 000 <sup>a,b</sup>	450 000 <sup>a,b</sup>
Mälardalen, except Greater Stockholm	196 000	153 000	42 500
Europe, except Mälardalen	133 000	101 000	31 700
Europe, total population exposure	2 520 000	2 000 000	524 000

<sup>a</sup> from Johansson & Eneroth (2007) , assumed population within the Greater Stockholm domain 1 405 600 persons.

<sup>b</sup> assuming that 20.5% of the wear particle exposure within the Greater Stockholm domain consists of fine particles.

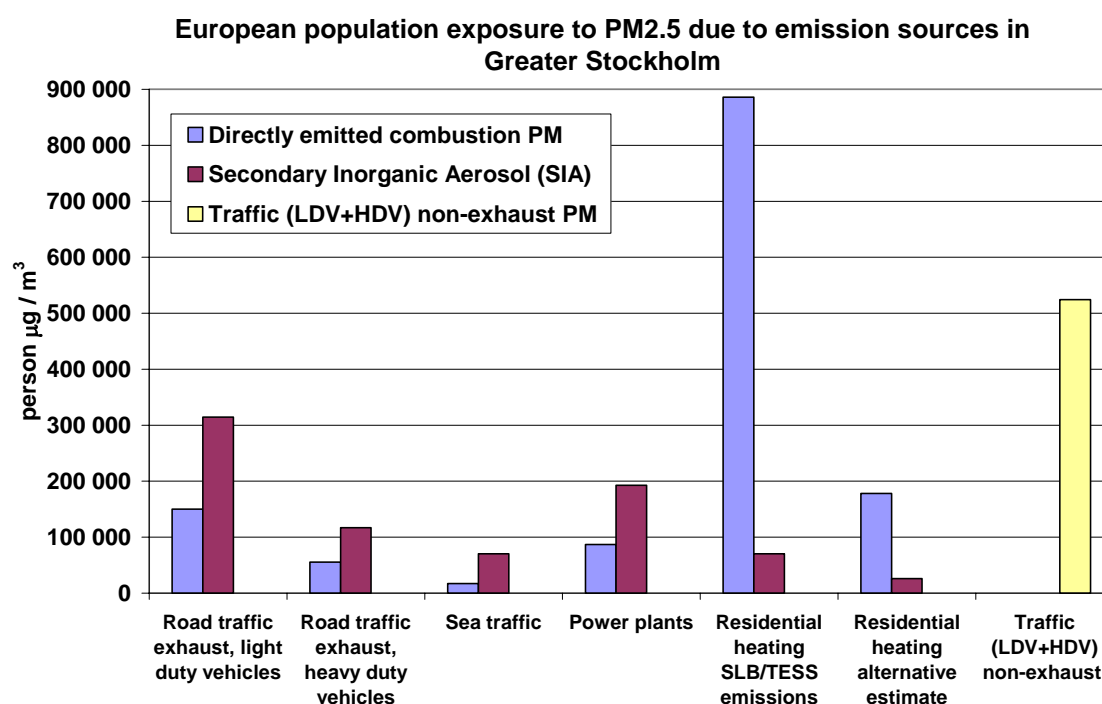


**Figure 22.** Contributions of different sources in the Greater Stockholm area to the yearly average population exposure to PM10 in Europe. For the residential heating source, two different estimates are given, one based on the emissions from Johansson and Eneroth (2007) and one based on emissions from SMED. Unit: person  $\mu\text{g}/\text{m}^3$ .



The total population exposures to *fine* particles (PM<sub>2.5</sub>) due to the different emissions in Stockholm are shown in Figure 23. Johansson and Eneroth (2007) only give total exposure to non-exhaust particles (not divided into fine and coarse fractions). Here it is assumed that the part of the population exposure due to the fine fraction inside Stockholm is 20.5% (based on Mälardalen scale simulation results). Outside Stockholm the relative amount of fine particles, compared to coarse, increases slightly due to longer atmospheric residence times for the finer particles.

The calculated total PM<sub>2.5</sub> exposure, due to Stockholm emissions, is dominated by directly emitted combustion particles from residential heating, and fine non-exhaust particles, when the emissions from Johansson and Eneroth (2007) are used. If the much lower SMED emissions would have been used for residential heating the results would have been quite different as can be seen in Figure 23, with the fine exhaust particles contributing about 1/3 to the total PM<sub>2.5</sub> exposure.



**Figure 23.** Contributions of different sources in the Greater Stockholm area to the yearly average population exposure to PM<sub>2.5</sub> in Europe. For the residential heating source, two different estimates are given, one based on the emissions from Johansson and Eneroth (2007) and one based on emissions from SMED. Unit: person µg / m³.

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