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N O R D I C W O R K I N G P A P E R S

Emissions and air exposure of carcinogens and co-carcinogens in four Nordic countries

Fausser, P., Plejdrup, M., Ketzel, M., Hertel, O., Loft, S., Gidhagen, L., Omstedt, G., Skårman, T., Kittilsen, M., Walker, S-E., Schwarze, P., Karvosenoja, N., Kukkonen, J., Karppinen, A., Pukkala, E., Salonen, R.

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Abstract: This project (KoL 12-08) was performed for the Climate and Air Quality Group (Klima- og Luftgruppen, KoL), Nordic Council of Ministers by atmospheric emission, exposure and epidemiology experts from Denmark, Finland, Norway and Sweden. Emission inventory methods and exposure models were presented. A list of carcinogenic and co-carcinogenic pollutants (particles, heavy metals and organic compounds) emitted from energy production, industrial activities, road transport, navigation, agriculture, residential heating and product use was compiled. Pollutant emissions levels for 2010 and trends for 1990 to 2010 were compiled and discussed, and modelled and measured atmospheric concentrations for 2010 were compiled on regional, urban and local scales. Nordic maps of emissions and air concentrations of PM_{2.5}, PM₁₀, NO_x, NMVOC, benzene, BaP, dioxin, cadmium and nickel were compiled for all aggregated main sources, traffic and residential wood combustion. An overview of local studies on exposure for cities or communities with emphasis on wood combustion and traffic and a discussion of existing epidemiological studies on cancer and environment were given.

Keywords: Carcinogenic pollutants, air, particles, organic compounds, heavy metals, emissions, exposure, GIS, residential wood combustion

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Abstract

This project (KoL 12-08) was performed for the Climate and Air Quality Group (Klima- og Luftgruppen, KoL), Nordic Council of Ministers by atmospheric emission, exposure and epidemiology experts from Denmark, Finland, Norway and Sweden. Emission inventory methods and exposure models were presented. A list of carcinogenic and co-carcinogenic pollutants (particles, heavy metals and organic compounds) emitted from energy production, industrial activities, road transport, navigation, agriculture, residential heating and product use was compiled. Pollutant emissions levels for 2010 and trends for 1990 to 2010 were compiled and discussed, and modelled and measured atmospheric concentrations for 2010 were compiled on regional, urban and local scales. Nordic maps of emissions and air concentrations of PM_{2.5}, PM₁₀, NO_x, NMVOC, benzene, BaP, dioxin, cadmium, nickel were compiled for all aggregated main sources, traffic and residential wood combustion. An overview of local studies on exposure for cities or communities with emphasis on wood combustion and traffic and a discussion of existing epidemiological studies on cancer and environment were given.

Summary

The Nordic countries, and Denmark in particular, have according to the WHO relatively high female total cancer and female lung cancer incidence rates among the 27 EU member states, see Table 1. It is well known that particularly exposure to air pollution is associated with adverse health effects in the population. The environmental exposure-health associations remain relatively ill-defined due to lack of information integration, and existing studies have focused on only a few pollutants and sources, such as particles and NO_x from traffic. However, emission inventories and air modelling and monitoring show that there are a number of sources that emit many critical pollutants all contributing, possibly in a synergistic way, to the overall cancer risk, see Table 2 and 3.

This study compiles existing data, information and maps on the state and trends of emissions and air concentrations of carcinogens and co-carcinogens in Denmark, Finland, Norway and Sweden. To present a more realistic and complete picture of the human exposure via air this study considers all relevant sources (energy production, industrial activities, road transport, navigation, agriculture, residential heating and product use) and their associated carcinogens. Emissions and air concentrations are compiled from the national emission reporting to United Nations Economic Commission for Europe (UNECE) and Convention on Long Range Transboundary Air Pollution (CLRTAP), and from various scientific studies.

Due to the novelty of this approach and the scarcity of data, priority has been given to sources and pollutants that have relatively high, and for some pollutants increasing, emissions, low cancer threshold values, and high availability of data; i.e. particles (PM_{2.5} and PM₁₀), organic compounds (Non-Methane Volatile Organic Compounds (NMVOC), Poly Aromatic Hydrocarbons (PAHs) incl. benz(a)pyrene (BaP), dioxin and benzene), heavy metals (cadmium and nickel), and furthermore NO_x. Focus is on traffic due to the relatively high amount of existing information and on residential wood combustion due to the high and still increasing emissions of many carcinogens, vicinity of large population, large uncertainty of emission amounts and location, and thus potential high impact on human health. Furthermore all sources have been considered as an aggregated group.

Summarized emission state and trends for the Nordic countries for the time period 1990 to 2010, unless otherwise stated, are;

The largest emission sources for particles (TSP, PM_{2.5} and PM₁₀) are residential plants and transport. Emissions have decreased for all particulate fractions, however, the PM_{2.5} emissions have increased during the last ten years due to increasing residential wood combustion.

NMVOC emissions originate from many different sources through incomplete combustion and evaporation, and have decreased more than 45 %. Road transport, navigation and solvent and product use are the main sources, and emission decreases are largely due to the increased use of catalyst cars and reduced emissions from use of solvents. Few data for benzene are available, however the predominant source of benzene in the air is road traffic and benzene emissions have decreased.

Emissions of PAHs (incl. BaP) have decreased slightly, however emissions from combustion of wood in the residential sector, being the predominant source with between 40 % and 80 % of the total emission in 2010, have increased more than 15 %. Aluminium production was the predominant PAH-4 (BaP, chrysene, benz(a)anthracene and benzo(b)fluoranthene) source in Norway, and shows decreasing emissions.

Overall emissions of dioxins have decreased by more than 20 % mainly due to a decrease in emissions from the metal industry. On the other hand increased wood burning within the energy sector has resulted in an increase in emissions. Between 10 % and 60 % of the dioxin emissions are from residential wood combustion, approximately 25 % are from the waste sector due to accidental fires, especially building fires. Emissions from navigation and metal production account for approximately 20 % and 10 % of the total, respectively.

Emissions of cadmium have decreased with more than 50 %, the main sources being combustion in energy industries (combustion of wood, wood waste and municipal waste), manufacturing industries (combustion of residual oil), wood processing industries (burning of refuse wood and waste liquor) and residential wood combustion. Decreasing emission from energy industries are seen due to decreasing combustion of coal and reduced emissions from metal production. Emissions from residential wood combustion have increased more than 100 % in some countries.

For NO_x, where transport (road and navigation) is highest contributor, the total emission have decreased more than 40 %, due to the increasing use of catalyst cars and installation of low-NO_x burners and denitrifying units in power plants and district heating plants. However in Norway, the emissions from diesel fired engines used within oil extraction increased during the latter years and curbed the overall reduction in NO_x emissions.

Even though an overall decrease in emissions to air is seen for many pollutants and sources across the four countries, emissions from residential wood combustion are relatively high and still increasing for NO_x, PM_{2.5}, arsenic, cadmium, mercury, lead, PAHs (incl. BaP), dioxins/furans and HCB. In general, levels and trends of air concentrations follow that of the emissions, and accordingly air concentrations have been decreasing, however, with stagnation during the latest 5 to 10 years, for many pollutants. On a local scale the air concentration threshold values may be exceeded, which is seen in studies of areas with extensive residential wood burning. Given the current trend, residential wood combustion will become more predominant in the emission picture, and the overall decreasing trend of carcinogens in air may shift to an increase, on a local and urban scale, in the future.

Only existing data are presented and used in this report and a conclusion is that there are important data gaps that need to be investigated and filled in order to gain an even more complete and realistic picture of the human exposure to airborne carcinogens. There is knowledge and lines of evidence that residential wood combustion is an important source to particles and several organic pollutants and heavy metals, and given the considerable uncertainties in data and methodologies it is recommended that this source is given priority especially regarding more accurate spatial mapping of sources, improved emission factors as a function of oven type and wood type, and modelling of air concentrations. This is necessary in order to in-

crease the knowledge on the environmental influence on human health, which will give high quality information to decision makers to facilitate define appropriate regulative actions and monitoring activities.

Outlook

The results from this study can serve as input to an (epidemiological) study where correlation between environmental exposure and cancer incidents can be investigated and possible causations of specific chemicals and sources to specific cancers can be identified. The maps compiled within the current project give a visual impression of emission and air concentration levels between urban and rural areas and on national levels. Zooming will give emission and exposure levels at grid scale, which is the most differentiated available spatial scale. From maps it is possible to identify localities with critically high exposures, and identify sources and pollutants that have high contribution to the air pollution. To understand if the concentrations are critical they can be compared with air quality limit threshold values and risk levels (Margin of Exposure), with different safety margins, can be calculated and mapped.

A next phase Nordic study can contribute to answer the questions:

What is the risk of this atmospheric exposure with respect to cancer?

Are the Nordic countries high-risk communities for some specific cancers, such as female total and lung cancer due to atmospheric exposures, also seen in comparison with other comparable cities and countries?

Are there significant correlations and possible causation between observed atmospheric pollutant concentrations and increased incidence rates of cancers, such as female cancers in the Nordic countries?

Are environmental cancer risks due to pollutants related to specific activities (e.g. residential wood burning) or locations (hot spots)?

1 Foreword

This study was performed during 2012 and 2013 for the Climate and Air Quality Group (Klima- og Luftgruppen, KoL), Nordic Council of Ministers, by the following people:

- Lars Gidhagen and Gunnar Omstedt, Swedish Meteorological and Hydrological Institute (SMHI)
- Tina Skårman, Swedish Environmental Research Institute (IVL)
- Marte Kittilsen, Statistics Norway (SSB)
- Sam-Erik Walker, Norwegian Institute for Air Research (NILU)
- Per Schwarze, Norwegian Institute of Public Health (FHI)
- Niko Karvosenoja, Finnish Environment Institute (SYKE)
- Jaakko Kukkonen and Ari Karppinen, Finnish Meteorological Institute (FMI)
- Eero Pukkala, Finnish Cancer Registry, Institute for Statistical and Epidemiological Cancer Research and University of Tampere
- Raimo Salonen, National Institute for Health and Welfare (THL)
- Steffen Loft, University of Copenhagen (KU)
- Matthias Ketzel, Ole Hertel, Marlene Plejdrup and Patrik Fauser, DCE – Danish Centre for Environment and Energy/Institute for Environmental Science, Aarhus University (ENVS).

These people are experts within emission inventories, air concentrations modelling, human health and epidemiological studies in their respective countries. They have access to data and methodologies and they participate in generating new data and develop new approaches for investigating and describing the state and trends of pollutant emissions and human exposure to air pollution and its consequences on human health.

The study is intended as the first part of a more comprehensive (epidemiological) study on the influence of airborne carcinogens on certain cancers in the Nordic countries.

2 Introduction: Air pollution and human health

It is well known that the environmental quality, and particularly exposure to air pollution, is associated with adverse health effects in the population (EU Environment Health Action Plans; EU Parma Declaration on Environment and Health) and although it is commonly known that carcinogenetic environmental exposures play an important role in the cancer aetiology in western societies, the environmental exposure-health associations remain relatively ill-defined due to lack of information integration. The 2010 report by the U.S. Presidential Cancer Panel concluded that the estimated environmentally related cancer burden of 4-6 % (WHO) is most likely a significant underestimation. Moreover the report concluded that some 40 % of the total cancer burden is still unaccounted for when life-style, heredity, etc. have been uncovered. How much of these 40 % can be ascribed to environmental exposures is unknown.

Table 1 Cancer incidences excl. non-melanoma skin for all cancers men, all cancers women and lung cancer women (<http://globocan.iarc.fr/>). ASR (W) is the age-standardised rate.

All cancer sites excl. non-melanoma skin, Men, GLOBOCAN2008			All cancer sites excl. non-melanoma skin, Women, GLOBOCAN2008			Lung cancer, Women, GLOBOCAN2008		
POPULATION	Numbers	ASR (W)	POPULATION	Numbers	ASR (W)	POPULATION	Numbers	ASR (W)
France (metropoli- tan)	188918	360.6	Denmark	16265	325.3	Denmark	1974	34.6
Ireland	10378	355.9	Ireland	9076	285.1	Hungary	3051	30.7
Hungary	26085	352.3	Netherlands	39691	276.5	Iceland	77	29.4
Belgium	32610	351.3	Belgium	26662	275.7	Netherlands	4000	27.2
Czech Republic	28510	348.8	Norway	11390	270.3	United Kingdom	17974	25.8
Norway	13210	338.4	Iceland	608	265.6	Norway	1122	24.7
Denmark	15924	334.7	United Kingdom	152408	260.5	Ireland	860	24.4
Germany	261173	330.7	Czech Republic	25453	259.1	Poland	6513	18.6
Luxembourg	1241	324.0	France (metropolitan)	143783	254.9	Albania	403	18.6
Slovakia	11142	320.2	Luxembourg	1185	254.4	Serbia	1525	18.5
Slovenia	5125	319.6	Italy	157109	251.6	Luxembourg	81	18.2
Lithuania	7281	316.5	Germany	218688	245.7	Belgium	1714	17.5
Croatia	11531	315.0	Sweden	21166	241.2	Czech Republic	1826	17.1
Switzerland	20578	313.2	Finland	12696	239.8	Switzerland	1292	17.0
Italy	183328	310.0	Switzerland	17033	236.0	Austria	1392	16.9
Spain	119590	309.9	Hungary	23532	235.6	Montenegro	90	16.7
Netherlands	42107	306.3	Slovenia	4517	232.9	Sweden	1589	16.4
Iceland	698	305.5	Croatia	10115	229.3	Germany	15070	16.4
Latvia	4825	304.0	Slovakia	10206	223.4	Slovenia	341	16.2
Estonia	2734	285.7	Lithuania	6668	207.8	France (metropolitan)	8194	14.7
Poland	75925	280.5	Bulgaria	14677	206.6	Croatia	675	13.8
United Kingdom	151827	280.0	Serbia	16463	205.2	Finland	718	11.7
Austria	19310	275.4	Estonia	2822	203.7	Italy	8493	11.4
Finland	12849	271.3	Austria	16635	200.6	Slovakia	512	10.6
Sweden	23385	269.6	Malta	715	199.7	Romania	1997	9.8
Belarus	15653	269.0	Albania	3909	196.6	Bosnia Herzegovina	326	9.5
Portugal	24030	266.8	FYR Macedonia	2893	196.5	Greece	1127	9.5
FYR Macedonia	3528	262.1	Latvia	4495	193.4	Estonia	150	9.0
Bulgaria	16024	252.9	Portugal	19254	190.8	Bulgaria	655	8.5
Russian Fed.	206370	247.1	Spain	77312	187.0	FYR Macedonia	122	7.7
Romania	37386	240.6	Belarus	15535	186.7	Spain	3126	7.7
Serbia	17720	238.5	Poland	64853	185.5	Republic of Moldova	220	7.1
Malta	775	233.2	Montenegro	911	185.2	Latvia	188	7.1
Montenegro	1006	231.2	Romania	32876	179.5	Russian Fed.	10247	7.0
Ukraine	70186	231.1	Russian Fed.	226826	178.9	Lithuania	257	6.5
Republic of Moldova	4702	217.0	Cyprus	1109	174.5	Ukraine	3173	6.2
Albania	3823	213.9	Ukraine	72774	173.0	Portugal	674	6.0
Bosnia Herzegovina	5535	198.3	Republic of Moldova	4693	167.1	Malta	23	5.9
Greece	21054	190.0	Greece	16035	136.2	Cyprus	33	4.9
Cyprus	1173	187.2	Bosnia Herzegovina	4243	133.6	Belarus	468	4.7

In areas with intensive industrial activities and traffic there is often a concern among the public and local authorities of the risk caused by emissions of chemicals to the surrounding environment. In many cases this concern is based on visual impressions of industrial buildings, lorries and trucks, ships bunkering in harbours or chimneys emitting smoke. In other cases there may be odour and taste problems of air and bathing and drinking water, and the

final consequence can be occurrence of illnesses such as asthma or bronchitis or eventually life threatening diseases such as cancer.

It may be the case that the visual or odour concern is not the dominating source of a health problem, but that some other source(s) or pollutant(s), however not so apparent, may bear the primary burden of pressure. There is therefore a need for a reliable procedure for scientists and decision makers to assess and communicate the state of environmental contamination and risk towards the public and surrounding environment. A careful definition and scoping of the model scenario is important in order to capture the important and critical sources in terms of pollutants and their emissions, environmental concentrations and human exposure concentrations. Many studies deal with these issues individually but there is a need for integrating and assessing the abundant data and not least to identify critical data gaps that impede a reliable conclusive understanding of environmental exposures and health effects.

To be able to act upon a concern in a scientifically reliable and accurate way there are a number of issues that must be considered. The pressure, or emission rates, from industries on the surrounding environment must be established. Industries are obliged to report emissions of harmful chemicals to various national and international bodies. On an international scale Persistent Organic Pollutants (POP) and heavy metal emissions are under the existing reporting obligations of the Convention on Long-Range Transboundary Air Pollution (CLRTAP) and the EU requires that certain industries must report the facilities environmental pressure to the European Pollutant Release and Transfer Register (E-PRTR). The E-PRTR is the most comprehensive and readily accessible register for industry emissions on an EU scale and is designed to provide information to citizens, industries, scientists, insurance companies, local authorities, non-governmental organisations and other decision-makers on the state and trends of the environment in relation to social, economic and health issues as well as the general raising of awareness of the environmental implications put on the environment and humans by industrial activities (E-PRTR, 2010).

Emission rates are introduced in atmospheric dispersion models to predict environmental concentrations at a target site. These environmental concentrations represent the exposure towards humans, where the polluted environmental medium can be ingested, inhaled or in dermal contact with humans. The choice of model follows the criteria of being accurate, transparent and reliable and must have a complexity that reflects the actual problem; simple models may have high model uncertainty and more complex models may introduce high input parameter uncertainty. It is essential to select an exposure model that represents the actual site and tidal scale and the input in terms of e.g. meteorological data in a realistic manner. Furthermore, model results must be evaluated against air monitoring data sampled on-site in order to ensure an accurate simulation of the pollutant fate and occurrence.

A number of cities where the combination of rapid growth, dirty fuels, and polluting technologies are overwhelming the capacities to control air pollution, source apportionment techniques offer policymakers practical tools for identifying and quantifying the different sources of air pollution, and thereby increasing the ability to put in place effective policy measures to reduce air pollution to acceptable levels. Source apportionment aims to explain the chemical composition of contributions from different sources. In doing so,

source apportionment quantifies the relative contributions of these different sources.

Epidemiology is the discipline to link the distribution of exposures and determinants of health-related states or events (including disease), and the application of this study to the control of diseases and other health problems (WHO, <http://www.who.int/topics/epidemiology/en/>). Epidemiological studies thus present an interdisciplinary challenge as emission data, exposure modelling and measurements are analysed in conjunction with health, sociological and life style data.

A number of epidemiological studies have already been done, also by participants in this project, and it has been shown that life-style factors are the predominant source to most cancer types, e.g. smoking is the predominant cause of lung cancer. Previous studies on the influence of chemicals via the environment have, however, focused on a limited number of pollutants such as particles, SO₂ and NO_x, often as indicators for specific sources such as traffic. The pollutants act individually with specific limit values.

A significant new contribution in this project is to investigate and present an estimate of the total cumulative exposure of the high number of organic pollutants and heavy metals based on existing data. This will give a more realistic estimate of the exposure, and also give input to the discussion regarding synergistic effect of mixtures and qualify the estimate of environmentally related cancer, as raised by the US presidential panel.

Based on the fact that the Nordic countries have high quality GIS-based emission data for a number of (co)carcinogens and sources, GIS-based exposure models, and high quality cancer registries, there is an obvious opportunity to do a Nordic case study on the influence of the exposure of chemicals via the environment on certain cancers.

The four countries do not have the same level of detail on emissions and exposures of pollutants and the extent of mapping varies between countries. A joint project will facilitate to obtain a common high level of emission and exposure mapping and modelling, and sharing of data between countries enables filling of data-gaps. From an international perspective the Nordic data and methodologies may be extrapolated to regions or countries with a worse situation, i.e. higher incidence rates and lesser developed data and methods.

The main results for Denmark, Finland, Norway and Sweden, presented in this report are:

- Identification of carcinogenic pollutants that are emitted from main sources (Chapter 3).
- Description of emission inventory methods and exposure models (Chapter 4 and country chapters).
- Presentation of state and trends of (co)carcinogen emissions in main sectors, as reported to UNECE and CLRTAP (country chapters).
- Tables with annual mean 2010 emissions of all (co)carcinogens in main sectors and total pr. inhabitant (Appendix A).
- Overview of available emission and exposure modelled spatial grids (maps) of (co)carcinogens. Model grids cover entire respective country and spatial scales are stated (Appendix B).

- Nordic maps of 2010 emissions on regional or urban scale of PM_{2.5}, PM₁₀, NO_x, NMVOC, benzene, BaP, dioxin, cadmium, nickel for; 1) all aggregated main sectors, 2) traffic and 3) residential wood combustion (Appendix C).
- Nordic maps of 2010 exposures on regional or urban scale of PM_{2.5}, PM₁₀, NO_x, NMVOC, benzene, BaP, dioxin, cadmium, nickel for; 1) all aggregated main sectors, 2) traffic and 3) residential wood combustion (Appendix D).
- Tables with modelled and/or measured annual mean 2010 air concentrations. Selected data are shown for nickel, cadmium, arsenic, PAHs (BaP), benzene, PM_{2.5}, PM₁₀, NO_x (NO₂) (country chapters).
- Overview of local studies on exposure for cities or communities with emphasis on wood combustion and traffic (country chapters).
- Discussion of existing epidemiological studies on cancer and environment. Application of available data in environmental health studies (Chapter 9).

3 Carcinogens and co-carcinogens in air

Carcinogens are defined according to the principles, procedures, and scientific criteria that guide the evaluations that a pollutant can increase the risk of cancer. These are described in the preamble to the IARC Monographs (2011). Co-carcinogens are compounds or particles that by themselves are not known carcinogens but may potentially enhance the carcinogenicity of other carcinogens.

A literature review of all available official emission data from relevant sources has been made with respect to identifying potential (co)carcinogens. The sources and their associated pollutants that are relevant to cancer and exposure via the environment are listed in Table 2. We have drawn on emission data that industries are committed to report to e.g. the United Nations Economic Commission for Europe (UNECE) Convention on Long Range Transboundary Air Pollution (CLRTAP, 2012), and data from the Pollutant Release and Transfer Registers (E-PRTR, 2012). Furthermore the gross list of pollutants is compiled from emission inventory guidelines for road traffic and ships (EMEP/CORINAIR, 2007), literature on flaring (Stroscher, 1996; Seebold, 2009; CONCAWE, 2009; EMEP/CORINAIR 2007) and from municipal and national reporting of various diffuse sources (EMEP/CORINAIR, 2007; Junta de Andalucia, 2010; US EPA, 1995). The pollutants in red are included in the national CLRTAP reporting.

Apart from the reported pollutants in Table 2 Nordic product registers comprise use amounts and emissions of pollutants that are used in consumer products and industrial processes and that are classified by the risk phrases R45 (may cause cancer) or R49 (may cause cancer by inhalation). A comprehensive list can be made but the bulk of these have small or no use amounts, typically with confidentiality of the pollutants with small use amounts. In Table 3 pollutants with emissions larger than 1 t, for the period 2002 – 2010 in Norway, are compiled. When large uses are associated with small emissions the pollutants are typically used as raw material in closed systems in the industry. The use amounts and emissions of pollutants can in some cases be differentiated in product categories according to the UCN system and industrial use categories according to the NACE system.

The carcinogens and co-carcinogens in Table 2 are considered in this study. To assess if air concentrations are critical with respect to cancer, they can be compared with the threshold values compiled in Table 4. These are mechanistic estimates since, in most cases, the shape of the risk curve is not known. Threshold values are 1:10,000 risk concentrations for carcinogens (US-EPA IRIS, 2013; EAQG) and EU Air Quality Guidelines second edition (WHO, 2000) for co-carcinogens.

Table 2 Sources and pollutants which are relevant in relation air and to cancer (carcinogens and co-carcinogens). Red pollutants are covered by CLRTAP.

	Energy production incl. waste incineration	Road traffic (diesel and gasoline)	Ships	Flaring	Industries	Various diffuse sources	Private wood combustion
Pollutant	Point sources	Line source	Point/line sources	Point sources	Point sources	Area source	Point sources
Chlorine and its inorganic compounds					C	D	
Particles (not specified)	A	A	A	B	C	D	E
PM _{2.5}	A	A	A			D	E
PM ₁₀	A	A	A		C	D	E
Arsenic	A		A	B	C	D	E
Cadmium	A	A	A		C	D	E
Chromium	A	A	A	B	C	D	E
Lead	A	A	A		C	D	E
Mercury	A		A	B	C	D	E
Nickel	A	A	A	B	C	D	E
NM VOC (not specified)	A	A	A	B	C	D	E
1,3-butadiene	A1	A					
Acetaldehyde	E	A		B			F
Acrolein	E	A		B			F
Benzene	A1	A	A	B	C	D	F
Crotonaldehyde	A1	A					F
Ethylbenzene	A1	A	A	B			F
Formaldehyde	E	A		B			F
Furans (unspecified)	E	A					E
Hexachlorobenzene	E				C	D	E
Hexachlorocyclohexane (Lindane)					C	D	
Isoprene	A1	A					
PCDD/F (dioxins not specified)	A	A	A	B	C	D	E
Styrene	A1	A		B			F
Tetrachloroethylene	F				C	D	
Trichloroethylene					C	D	
Trichloromethane					C		

Table 2 (continued).

	Energy production incl. waste incineration	Road traffic (diesel and gasoline)	Ships	Flaring	Industries	Various diffuse sources	Private wood combustion
Pollutant	Point sources	Line source	Point/line sources	Point sources	Point sources	Area source	Point sources
SO _x (SO ₂ for PRTR data)	E	A	A	B	C	D	E
NO _x (NO ₂ for PRTR data)	E	A	A	B	C	D	E
PAHs (not specified) ¹⁾	E	A	A	B	C	D	E
Benzo(b)fluorene	A1	A	A				F
Benzo(a)anthracene	E	A	A	B			E
Benzo(a)pyrene	E	A	A	B	C		E
Benzo(b)fluoranthene	E	A	A		C		E
Benzo(j)fluoranthene	E	A	A				E
Chrysene	E	A	A	B			E
Dibenzo(a,h)anthracene	E	A	A				E
Dibenzo(a,j)anthracene	A1	A	A				A1
Dibenzo(a,l)pyrene	A1	A	A				A1
Indeno(1,2,3-cd)pyrene	E	A	A	B	C		E
Naphthalene	E	A		B			E

¹⁾ Six PAHs are covered by PRTR: benz(a)pyrene, benzo(ghi)perylene, benzo(k)fluoranthene, fluoranthene, indeno(1,2,3-cd)pyrene and benzo(b)fluoranthene.

A: Potential pollutants from EMEP/CORINAIR (2007). It is not possible to calculate emissions of some pollutants due to data gaps in emission factors and information of e.g. vehicle traffic speciation.

A1: Pollutants are emitted from road traffic and assumed also emitted from energy production and wood combustion.

B: Flaring; Information on pollutants are from specific installations in Alberta as described in (Stroscher, 1996) and Canadian Public Health Association (2000). Fugitive emissions and leaks are also relevant and are comprised in pollutants from flaring.

C: E-PRTR (2010) holds reported emission data for facilities that exceed specific pollutant threshold values. The thresholds have been fixed at a level that aims to cover about 90 % of the emissions. This implies that only large and medium sized industrial plants, which are covered in the Annex I of the PRTR regulation (E-PRTR Annex I, 2010) and not low emissions from smaller installations, are reported. Some potentially relevant pollutants may not be covered by the E-PRTR pollutant list.

D: Diffuse sources, comprising many SNAP categories, are, e.g. extraction and treatment of minerals, agriculture, road paving, biogenic, fuel distribution, use of refrigerants and propellants, animal husbandry, petrol stations, roof waterproofing, forest fires, dry cleaning, agricultural machinery, commercial and institutional sector, households, solid waste treatment, solvent use. Diffuse emissions are described as area sources.

E: A study on decentralized combined heat and power plants, <25MW_e, for the plant technologies: Municipal solid waste incineration plants, plants combusting straw or wood, natural gas fuelled reciprocating engines, biogas fuelled engines, natural gas fuelled gas turbines, gasoil fuelled reciprocating engines, gasoil fuelled gas turbines, steam turbines combusting residual oil and reciprocating engines combusting biomass producer gas based on wood (Nielsen et al., 2010). Wood combustion plants are used as proxy for residential wood combustion emissions. F: US EPA (1995).

Table 3 Pollutants, used in consumer products and industrial activities, classified with R45 or R49 with uses larger than 1 t in total for the period 2002-2010 from Norwegian Product register.

CAS number	Pollutant name	Risk phrases	Total emissions (tonnes)	Net amount consumed in Norway (tonnes)
75-01-4	vinyl chloride	45-12	1173	1 295 331
107-06-2	1,2-dichloroethane	45-11-22-36/37/38	1164 ¹⁾	3 173 272
1333-82-0	chromium trioxide	45-46-9-24/25-26-35-42/43-48/23-62-50/53	95.4	213
79-01-6	trichloroethylene (TRI)	45-36/38-52/53-67	94.0	401
106-89-8	1-chloro-2,3-epoxypropane	45-10-23/24/25-34-43	58.8	31 152
50-32-8	Benz(a)pyrene	45-46-60-61-43-50/53	58.1	5 249
56-55-3	benzo(—)anthracene	45-50/53	54.6	546
205-99-2	benzo(e)acephenanthrylene	45-50/53	43.8	438
207-08-9	benzo(k)fluoranthene	45-50/53	41.5	415
205-82-3	Benzo(j)fluoranthene	45-50/53	39.3	393
25321-14-6	dinitrotoluene	45-23/24/25-48/22-62-68-51/53	36.4	87.7
101-77-9	4,4'-methylenedianiline	45-39/23/24/25-43-48/20/21/22-68-51/53	34.9	46.4
1344-37-2	lead sulfochromate yellow (C.I.Pigment Yellow 34)	49-61-33-50/53-62	33.4	126
302-01-2	hydrazine	45-10-23/24/25-34-43-50/53	26.1	50.6
122-60-1	2,3-epoxypropyl phenyl ether	45-20-37/38-43-68-52/53	20.9	26.0
7440-41-7	beryllium	49-25-26-36/37/38-43-48/23	15.6	156
12656-85-8	lead chromate molybdate sulfate red (Pigment red 104)	49-61-33-50/53-62	9.68	69.4
71-43-2	benzene	45-46-11-36/38-48/23/24/25-65	9.34	3 811
79-06-1	acrylamide	45-46-20/21-25-36/38-43-48/23/24/25-62	7.65	9.5
1313-99-1	nickel monoxide	49-43-53	3.8	34.7
106-99-0	1,3-butadiene	45-46-12	3.17	2 202
606-20-2	2,6-dinitrotoluene	45-23/24/25-48/22-62-68-52/53	2.33	23.4
121-14-2	2,4-dinitrotoluene	45-23/24/25-48/22-62-68-51/53	1.79	17.8
75-56-9	methyloxirane	45-46-12-20/21/22-36/37/38	1.7	3.1
Total			3 030	4 514 073

¹⁾ An updated number for this substance for will be much lower, due to a correction of an emissions factor (from 0.001 to 0.00002 for the last 3 years of the time series).

Table 4 Threshold values are 1:10.000 risk concentrations for carcinogens and air quality guideline values for other pollutants. Red pollutants are covered by CLRTAP.

	Threshold values ¹⁾
Chlorine and its inorganic compounds	RfC = 0.2 µg/m ³ WHO (no IARC class)
PM _{2.5}	WHO: 10 µg/m ³ annual mean; 25 µg/M3 24 hr mean - lung
PM ₁₀	WHO: 20 µg/m ³ annual mean; 50 µg/m ³ 24 hr mean - lung
Particles (unspecified)	NA
Arsenic	1 in 10.000 = 0.02 µg/m ³ (IARC class 1) - lung
Cadmium	1 in 10.000 = 0.06 µg/m ³ (IARC class 1) - lung
Chromium	1 in 10.000 = 0.0025 µg/m ³ (IARC class 1 Cr 6 and 3 for others) – lung
Lead	IARC class 2A-3
Mercury	RfC = 1 µg/m ³ WHO (no IARC class)
Nickel	1 in 10.000 = 0.25 µg/m ³ (IARC class 1) – lung
NMVOC (unspecified)	IARC class 3
Benzene	1 in 10.000 = 13 µg/m ³ (IARC class 1) - leukaemia
Hexachlorobenzene	1 in 10.000 = 0.2 µg/m ³ (IARC class 2B)
Hexachlorocyclohexane (Lindane)	1 in 10.000 = 0.06 µg/m ³ (IARC class 2B)
PCDD/F (dioxins unspecified)	1 in 10.000 = 0.00008 µg/m ³ (IARC class 1)
Tetrachloroethylene	RfC = 250 µg/m ³ (IARC class 2A)
Trichloroethylene	1 in 10.000 = 230 µg/m ³ (IARC class 2A)
Trichloromethane	1 in 10.000 = 4 µg/m ³ (no IARC class)
SO _x (/SO ₂ for PRTR data)	WHO: 20 µg/m ³ 24 hr; 500 µg/m ³ 10 min mean (IARC class 3 SO ₂)
NO _x (/NO ₂ for PRTR data)	WHO: 40 µg/m ³ annual mean; 200 µg/m ³ 1 hr mean (no IARC class)
PAH ¹⁾	BaP: 1 in 10.000 = 0.0012 µg/m ³ (IARC class 1)
Ethyl benzene	NA
1,3-butadiene	1 in 10.000 = 3 µg/m ³ (IARC class 2) - leukaemia

¹⁾ 1:10,000 risk concentrations for carcinogens (US-EPA IRIS, 2013; EAQG) and EU Air Quality Guidelines second edition (WHO, 2000) for other pollutants. The lowest, most conservative, values are used. NA: not applicable.

4 Emissions and exposure modelling

Although many studies have demonstrated that air pollution is hazardous to human health, there are still many unknowns regarding the impact of air pollution on the population. Difficult aspects in this context are to generate reliable and accurate emission data and perform a proper assessment of the human exposure to air pollution. Here an overview of the methodologies and data for emissions and exposure assessments that are used in the Nordic countries, is given. Exposure models are summarized with Denmark as an example, other countries can be seen in their respective chapters.

4.1 Emissions

Denmark, Finland, Norway and Sweden are obliged to report atmospheric emissions to a number of international bodies and the most comprehensive with respect to covering carcinogenic pollutants is the United Nations Economic Commission for Europe - Convention on Long-Range Transboundary Air Pollution (UNECE - CLRTAP). The emissions are reported as national totals for a number of sectors and sub-sectors as defined by the Nomenclature For Reporting (NFR). CLRTAP comprises the (co)carcinogenic pollutants; sulphur dioxide (SO₂), nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOC), total suspended particulates (TSP), particulate matter with an aerodynamic diameter less than 10 µm (PM₁₀), particulate matter with an aerodynamic diameter less than 2.5 µm (PM_{2.5}), arsenic (As), cadmium (Cd), chromium (Cr), mercury (Hg), nickel (Ni), lead (Pb), dioxins and furans (PCDD/F), hexachlorobenzene (HCB) and the polycyclic aromatic hydrocarbons (PAHs): benzo(b)fluoranthene, benzo(k)fluoranthene, benz(a)pyrene and indeno(1,2,3-c,d)pyrene.

The distribution in the CLRTAP reporting is based on GNFR categories and the corresponding NFR codes to assure the most accurate distribution of the emissions. There is a mandatory reporting every five years of gridded emissions. The latest was in 2012, covering the emission in year 2010 and the historical years 1990, 1995, 2000 and 2005. The reporting includes a number of sub-categories representing separate sources or groups of sources; Large Point Sources, Stationary combustion for point sources, Stationary combustion for area sources, Mobile sources, Aviation, Fugitive emissions, Industry, F-gases, Solvents, Waste and a number of sub-models for the agricultural sector.

In addition to the national reporting of emissions there are scientific investigations where emissions are assessed for specific sources and pollutants, typically on a local (address) scale often in relation to human impact analysis. In this report such Nordic studies with relevance to carcinogens are stated.

4.2 Exposure modelling (air concentrations)

Human exposure is here defined as “an individual’s contact with a pollutant concentration” (Hertel et al., 2001). For personal exposure to take place two events need thus to occur simultaneously; pollution concentration at a particular time and place must be nonzero and the person must be present in that same place and at that time. To obtain an estimate of personal exposure,

data for time-activity pattern as well as data for air pollution levels are therefore crucial.

Emission data from the national inventories are often used as input to exposure modelling, which in turn will serve as input in assessment and evaluation of health effects. Emission data can be used at the most differentiated level or aggregated to more overall sectors, and depending on the design of the exposure model and on the desired output scale, exposure modelling is done on three spatial scales:

- Regional modelling is typically done on 50x50 km² or 16x16 km² grids. Modelling can be based on national emissions or on emissions on the northern hemisphere of a number of sources and CLRTAP pollutants.
- Urban modelling is done on a 1x1 km² scale and typically covers traffic and (co)carcinogens NO_x and particles (PM, PM_{2.5}, PM₁₀).
- Local scale (address level) studies can focus on specific sources such as traffic or residential wood burning and pollutants such as NO_x, particles, PAHs, heavy metals and benzene. Localities are related to specific cohorts, e.g. the Danish KHK cohorte comprising 50,000 persons.

In assessment of human exposure to air pollution various types of exposure proxies may be applied (emission density in a region, distance to trafficked road etc.). However, exposure may also be estimated on basis of measurements from routine monitoring network and/or calculations from air quality models, it may be determined by tracing people along routes using personal exposure monitors (PEMs) or by applying exposure models, and finally it may be determined on basis of various types of biomarkers.

Air pollution is a trans-boundary problem where pollutants may be transported over thousands of km. A wide range of different types of models is currently available for describing these pollution phenomena. Such models may have different parameterizations of the physical and chemical processes (emission, dispersion, transport, transformation and dry and wet deposition), but they all demand proper input data for meteorology, emissions and land use for the selected model domain. Integrated model systems, where a hierarchy of models is combined, are being developed at many institutes these years. The THOR system (Brandt et al., 2001 and <http://www.au.dk/thor>) is an example of such an integrated model system, which has been applied for providing long-range transported pollution concentrations in the exposure assessments.

Various air quality models have been developed for describing the pollution levels in the urban background. The dispersion conditions are strongly affected by the city structure, which may generate motions at scales as large as the whole city area itself. Within the Danish exposure studies, measurements from urban background stations have been applied whenever possible. However, in the case when observations have not been available, a simple model for the urban background pollution (UBM; Berkowicz, 2000a) has been applied. In UBM, contributions from the individual area sources in the urban area, subdivided into a grid net of a resolution of typically 1x1 km², are integrated along the wind direction path assuming linear dispersion with the distance to the receptor point. Horizontal dispersion is accounted for by averaging the calculated concentrations over a certain wind direction dependent sector, centred on the average wind direction.

Since a significant part of the exposure of the population is caused by emissions from traffic in urban streets, street pollution models may serve as important tools in exposure assessment. Examples of these models are STREET, CPBM, OSPM and CAR. In the Danish model OSPM (Berkowicz, 2000), which has been applied for exposure assessment in a number of studies, concentrations of exhaust gases are calculated hour by hour, combining a plume model for the direct impact of vehicle emitted pollutants with a box model for computation of the additional impact due to pollutants being re-circulated within the street by the vortex flow. OSPM makes use of a simplified parameterisation of flow and dispersion conditions in a street canyon, but has been found to reproduce the observed hourly mean concentrations of traffic pollutants in urban streets well.

For assessing exposures in the Danish population, a GIS based modelling system, AirGIS (<http://AirGIS.dmu.dk>), has been developed (Jensen et al., 2001). AirGIS is originally aimed for traffic air pollution, but is under steady improvement and development (Jensen et al., 2009; Ketzel et al., 2011) e.g. also to handle other pollutant emissions. The central part is Operational Street Pollution Model (OSPM) (Berkowicz, 2000) that is described above and is currently applied in >17 countries worldwide (Kakosimos et al., 2010), among them Norway, Sweden and Finland. Within the Danish research centre AIRPOLIFE (www.airpolife.dk), the AirGIS system was applied for exposure assessment for a variety of DK cohorts including the diet, cancer, & health cohort of 50,000 people.

In Appendix B a summary of existing atmospheric emissions and exposure modelling of (co)carcinogens with spatial scales, is shown for the Nordic countries.

5 Denmark

5.1 State and trend of (co)carcinogen emissions to air

5.1.1 Sulphur dioxide (SO₂)

The main part of the SO₂ emission originates from combustion of fossil fuels, i.e. mainly coal and oil, in public power and district heating plants. From 1980 to 2010, the total emission decreased by 97 %. The large reduction is mainly due to installation of desulphurisation plants and use of fuels with lower content of sulphur in public power and district heating plants. Also emissions from industrial combustion plants, non-industrial combustion plants and other mobile sources are important. National sea traffic (navigation and fishing) contributes with about 13 % of the total SO₂ emission in 2010. This is due to the use of residual oil with high sulphur content.

5.1.2 Nitrogen oxide (NO_x)

The transport sector is the sector contributing the most to the emission of NO_x and, in 2010, 45 % of the Danish emissions of NO_x originates from road transport, national navigation, railways and civil aviation. Also emissions from national fishing and off-road vehicles contribute significantly to the NO_x emission. For non-industrial combustion plants, the main sources are combustion of gas oil, natural gas and wood in residential plants. The emissions from energy industries have decreased by 72 % from 1985 to 2010. In the same period, the total emission decreased by 53 %. The reduction is due to the increasing use of catalyst cars and installation of low-NO_x burners and denitrifying units in power plants and district heating plants.

5.1.3 Non-methane volatile organic compounds (NMVOC)

The emissions of NMVOC originate from many different sources and can be divided into two main groups: incomplete combustion and evaporation. Road vehicles and other mobile sources such as national navigation vessels and off-road machinery are the main sources of NMVOC emissions from incomplete combustion processes. Road transportation vehicles are still the main contributors, even though the emissions have declined since the introduction of catalyst cars in 1990. The evaporative emissions mainly originate from the use of solvents and the extraction, handling and storage of oil and natural gas. The emissions from the energy industries have increased during the nineties due to the increasing use of stationary gas engines, which have much higher emissions of NMVOC than conventional boilers. The total anthropogenic emissions have decreased by 55 % from 1985 to 2010, largely due to the increased use of catalyst cars and reduced emissions from use of solvents.

5.1.4 Total Suspended Particulate matter (TSP) and Particulate Matter (PM)

The PM emission inventory has been reported for the years 2000-2010. The inventory includes TSP, emission of particles smaller than 10 µm (PM₁₀) and emission of particles smaller than 2.5 µm (PM_{2.5}). The largest PM_{2.5} emission sources are residential plants (71 %), road traffic (9 %) and other mobile sources (8 %). For the latter, the most important sources are off-road vehicles and machinery in the industrial sector and in the agricultural/forestry sector (32 % and 37 %, respectively). For the road transport sector, exhaust emis-

sions account for the major part (64 %) of the emissions. The PM_{2.5} emission increased by 30 % from 2000 to 2010 due to an increasing wood consumption in the residential sector. The largest TSP emission sources are the residential sector and the agricultural sector. The TSP emissions from transport are also important and include both exhaust emissions and the non-exhaust emissions from brake and tyre wear and road abrasion. The non-exhaust emissions account for 62 % of the TSP emission from road transport.

5.1.5 Heavy metals

In general, the most important sources of heavy metal emissions are combustion of fossil fuels and waste. The heavy metal emissions have decreased substantially in recent years, except for Cu. The reductions span from 29 % to 91 % for Zn and Pb, respectively. The reason for the reduced emissions is mainly increased use of gas cleaning devices at power and district heating plants (including waste incineration plants). The large reduction in the Pb emission is due to a gradual shift towards unleaded gasoline, the latter being essential for catalyst cars. The major source of Cu is automobile tyre and break wear (93 % in 2010) and the increase from 1990 to 2010 is caused by increasing mileage.

5.1.6 Cadmium (Cd)

The main sources of emissions of Cd to air are combustion in energy industries (mainly combustion of wood, wood waste and municipal waste) and manufacturing industries (mainly combustion of residual oil). In the transport sector emissions from passenger cars is the main source contributing with 53 % of the sectoral emission in 2010. The emission from non-industrial combustion is dominated by wood combustion in residential plants which accounts for 78 % of the sectoral emission in 2010. Emissions from combustion in residential plants have increased by 119 % since 1990. The decreasing emission from energy industries are related to the decreasing combustion of coal.

5.1.7 Mercury (Hg)

The largest sources of Hg emissions to air are waste incineration and coal combustion in energy industries. Due to improved flue gas cleaning and decreasing coal combustion the emissions from Energy industries decreased by 76 % from 1990-2000. Non-industrial combustion is dominated by wood combustion in residential plants while emissions from the waste sector mainly stems from cremation. The variations in emissions from industrial processes are caused by a shutdown in 2002 followed by re-opening and a second shut in 2005 down of the only Danish electro-steelwork.

5.1.8 Lead (Pb)

The main Pb emission sources are combustion in residential plants and energy industries and transport. In earlier years combustion of leaded gasoline was the major contributor to Pb emissions to air but the shift toward use of unleaded gasoline for transport has decreased the Pb emission from transport by 93 %. In the non-industrial combustion sector the dominant source is wood combustion in residential plants. The trend in the Pb emission from non-industrial combustion from 1990 to 2010 is almost constant. This is due to the shift towards unleaded gasoline, as this sector includes other mobile sources in household, gardening, agriculture, forestry, fishing and military. However this is counteracted by an increase in emission from

residential plants. The decreasing emission from Energy industries (97 % from 1990 to 2010) is caused by the decreasing coal combustion.

5.1.9 Polycyclic Aromatic Hydrocarbons (PAHs)

The emission inventory for Polycyclic aromatic hydrocarbons (PAHs) includes four PAHs: benz(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene and indeno(1,2,3-cd)pyrene. Benzo(b)fluoranthene and Benz(a)pyrene contribute the major PAH emission by 31 % and 30 %, respectively. The most important source of PAH emissions is combustion of wood in the residential sector making up 88 % of the total emission in 2010. The increasing emission trend is due to increasing combustion of wood in the residential sector. The PAH emission from combustion in residential plants has increased by 17 % from 1990 to 2010.

5.1.10 Dioxins and furans (PCDD/ PCDF)

The major part of the dioxin emission owes to wood combustion in the residential sector, mainly in wood stoves and ovens without flue gas cleaning. Wood combustion in residential plants accounts for 64 % of the national dioxin emission in 2010. The contribution to the total dioxin emission from the waste sector (24 % in 2010) stems from accidental fires, especially building fires. The emission of dioxins from energy industries owes mainly to the combustion of biomass such as wood, wood waste and to a less extend agricultural waste.

5.1.11 Hexachlorobenzene (HCB)

Stationary combustion accounts for 98 % of the estimated national Hexachlorobenzene (HCB) emission in 2010. This is mainly due to combustion of municipal solid waste in heating and power plants. The HCB emission from stationary plants has decreased 81 % since 1990 mainly due to improved flue gas cleaning in MSW incineration plants. Wood combustion in households is also an important source, and has increased by 302 % since 1990, due to increasing wood consumption.

In Table A1 (Appendix A) Danish emissions for the year 2010 are shown for main source categories.

5.2 Emission inventory

Previously the Danish emissions inventory has been available on 50x50 km² EMEP grid for reporting of air pollutants to CLRTAP every fifth year, latest in 2007 for the emission year 2005 and the historical years 2000, 1995 and 1990. The methodology is described in Jensen et al. (2008a). Besides the emission distribution on 50x50 km² resolution, a distribution on the 17x17 km² EMEP grid has been set up and used in research projects combined with detailed distributions for relevant sectors or subsectors. The 17x17 km² distribution has e.g. been used in combination with a detailed distribution of emissions from road traffic on 1x1 km² resolution in the on-going research project REBECa (Renewable Energy in the transport sector using Biofuels as Energy Carriers) and the completed project HYSCENE (Environmental and Health Impact Assessment of Scenarios for Renewable Energy Systems with Hydrogen). In the research project WOODUSE (Residential wood combustion and the interaction between technology, user and environment) the emission distribution on 17x17 km² has been combined with a detailed dis-

tribution of emissions from wood combustion in the residential sector at address level, see more detailed description below.

In 2010 the new spatial high resolution distribution model for emissions to air, SPREAD, is completed at NERI (Department of Policy Analysis). SPREAD enables distribution of the Danish emissions for all pollutants and all sectors in the national emission database on a 1x1 km² grid covering Denmark and its national sea territory. The model is set up in Microsoft Access databases and the distribution keys are set up in GIS (ArcMAP). Output tables are transformed to shape files for visualisation in GIS. The projection for all output shape files are UTM zone 32 N and the datum is ETRS89.

The emissions on 1x1 km² grid are reported to CLRTAP and is carried out on both source and pollutant level, which makes it possible to extract gridded data for a single source or a single pollutant. The following pollutants are included: SO₂, NO_x, NMVOC, TSP, PM₁₀, PM_{2.5}, As, Cd, Cr, Cu, Hg, Ni, Pb, dioxin, benz(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene and indeno(1,2,3-cd)pyrene. The gridding is carried out on the most disaggregated level possible. For many sources the gridding is carried out on SNAP-3 level. For sources or sectors with very scarce data on spatial location of the sources, the gridding is made for a group of sources. On address level emissions are available for point sources that are included in "Energiproducenttællingen", i.e. power plants delivering electricity and heat to the public net, and smaller power-heat plants and industries delivering surplus electricity and/or heat.

In Appendix C Nordic maps of emissions are shown for regional scale emissions.

5.2.1 Gridding of residential wood combustion emissions (WOODUSE)

As a part of the integrated research project WOODUSE, Danish Centre for Environment and Energy (DCE) developed a method for high resolution geographical emission mapping of particulate matter for residential wood combustion, designed for input to local air quality models used for human exposure simulations. The method was applied for a case study area covering four municipalities adjacent to Copenhagen. The method could be extended to cover the whole of Denmark, but this is presently not feasible due to missing data and discrepancies in data quality.

Several geo-referenced basic datasets were applied; most extensively the central building and dwelling register (BBR) and extracted data from chimney sweepers. The BBR is a countrywide register with data on every building and residence, which can accurately position all information at address level by coordinates. The Chimney sweep registers contain accurate information on the number and type of facilities used for wood combustion in every household. The chimney registers enumerate every boiler, stove or fireplace that is connected to chimney(s) on the property. They furthermore contain information on heights of chimneys exceeding 10m.

The major challenge was to combine and link geo-referenced data from different hierarchical levels and to calculate fuel (wood) consumption for each household as a basis for emission factors estimation. Emission mapping was made based on calculated emissions factors and aggregated point-based emissions at address level in a 100x100 m² grid, see Figure 1.

Figure 1 Estimated annual emissions of PM₁₀ (kg/year) from residential wood combustion in Værløse. Purple areas are district heating and blue areas are heated by natural gas. Grids are 100x100 m². (Tranekjær and Hansen, 2007).

5.3 Exposure modelling

Regional modelling is done with DEHM on a 5.6x5.6 km² grid and is based on emissions on the northern hemisphere of all major sources and pollutants (Christensen 1997 and <http://envs.au.dk/videnudveksling/luft/model/dehm/>). Urban modelling is done with UBM on a 1x1 km² scale and at present only covers traffic and the carcinogens NO_x and PM. Modelling on street scale (address level) only covers traffic as source and the carcinogens: NO_x and PM. Using the AirGIS to generate input automatically and the efficient OSPM exposure can be calculated at a large number of localities and for long time periods (back to 1960ies until present) related to specific cohorts, e.g. the Danish KKH cohort comprising 50,000 persons.

For specific pollutants (e.g. NMVOC, BaP, dioxin, Cd, Ni) no detailed emission inventories exist and no modelling can be performed for this reason. Exposure estimates can be derived from measuring campaigns at various locations e.g. with focus on traffic emission (Ellermann et al. 2012b) or on residential wood burning (Wåhlin et al. 2010; Ellermann et al. 2011).

In Appendix D Nordic maps of modelled air exposures of PM_{2.5}, PM₁₀, NO_x for all sources are shown for the regional scale and urban scale (1x1 km²). In Figure 2 and 3 air concentrations from these maps zoomed in on the Copenhagen area for NO₂ and PM₁₀ are shown. In Figure 4 NO₂ concentrations calculated at address level for the town of Roskilde in 2010 is shown for traffic emissions only.

Figure 2 Annual mean 2010 NO_2 air concentrations in Copenhagen area ($1 \times 1 \text{ km}^2$) estimated with UBM with contribution from all sources (based on Jensen et al. 2010).

Figure 3 Annual mean 2010 PM_{10} air concentrations in Copenhagen area including Øresund ($1 \times 1 \text{ km}^2$) estimated with UBM. Unknown mass is not included (based on Jensen et al. 2010).

Figure 4 Example of NO₂ concentrations calculated at address level for the town of Roskilde in 2010. The AirGIS system is used together with OSPM, only considering traffic emissions (See: www.au.dk/OSPM).

5.4 Air concentration measurements

In addition to modelled air concentrations examples of measured air concentrations in urban and rural regions of Denmark are shown below for selected heavy metals and organic pollutants.

Figure 5 Annual averages measured at selected stations for nickel (Ni) in particulate matter. Until 2000 in TSP and later in PM₁₀ – except for Copenhagen (HCAB: HC Andersens Boulevard in the centre of Copenhagen) where PM₁₀ replaced TSP from the beginning of 2006. The heavy metals including Ni are usually found in fine particles (diameter < 2.5µm), which make the TSP and the PM₁₀ values comparable. The dashed line indicates that the analysis method has been changed from 2009 to 2010 (Ellermann et al. 2012b).

Figure 6 Annual average concentrations of measured heavy metal concentrations in ambient air since 1979. Values represent the average at the regional background stations Keldsnor and Tange in the period until 2010. From 2011 values show the average of two other background stations Anholt and Risø. The vertical line indicates a shift in the measuring method in 2010 (Ellermann et al. 2012a).

Figure 6.1. Annual average concentrations of benzene and toluene on the kerbside station Jagtvej, Copenhagen/1257.

Figure 10.2. Annual average concentrations of benzo[a]pyrene at H.C. Andersens Boulevard (Copenhagen/1103).

Figure 10.1. Monthly average concentrations in 2011 of benzo[a]pyrene and the sum of the analysed PAH.

Figure 7 Measured concentration levels and trends of annual mean of benzene (top), annual mean of benz(a)pyrene (middle) and monthly mean of benz(a)pyrene and sum(PAH) (bottom) in ambient air at Jagtvej and H.C. Andersens Boulevard, which are busy traffic locations in Copenhagen (Ellerman et al. 2012b).

Table 5 Measured daily minimum, maximum and monthly mean concentrations (ng/m³) of benz(a)pyrene during 2011, measured in ambient air in Copenhagen (Ellerman et al. 2012b).

Month	Minimum conc.	Maximum conc.	Mean conc.
January	0,08	1,39	0,49
February	0,06	1,74	0,44
March	0,08	0,41	0,23
April	0,06	1,68	0,24
May	0,03	0,16	0,06
June	0,03	0,25	0,07
July	0,02	0,17	0,07
August	0,02	0,18	0,06
September	0,06	0,29	0,14
October	0,07	0,33	0,13
November	0,16	1,47	0,63
December	0,06	1,04	0,33
Annual	0,02	1,74	0,21

Figure 8 Measured total PAH (top) and benz(a)pyrene (BaP) (bottom) in ambient air in background area and near a place affected by wood stove emissions, using 24h samples on quartz filter. The sampling began in the morning of the indicated day (Wählin et al. 2010).

The measured concentrations (ng/m³) of benz(a)pyrene in an area with high usage of wood stoves (Figure 8) are about 2-3 times higher during the winter season compared to a busy street location in Copenhagen (H.C. Andersens Boulevard) (Figure 7b and 7c and Table 5). Daily averages of benz(a)pyrene are measured up to 12.5 ng/m³ at the wood stove area, while the annual av-

erage is about 0.61 ng/m³ compared to 0.21 ng/m³ at the street location. (Ellermann et al. 2011). See summary in Table 6.

Benzene concentrations in four large cities in Denmark are measured at about 0.9-1.4 µg/m³ as annual average in 2010 (Ellermann et al. 2011). See Figure 7a and Table 6.

Table 6 Particles, NO₂ and NO_x: Measured ranges in annual mean pollutant concentration in the period 2000 to 2011 (µg/m³ except for particle number #/cm³. For NO_x the concentration is in µg NO₂/m³. Values are derived from measurements within the Urban Air Quality Monitoring Programme. Based on Hertel (2009) and Ellermann (2012b). Heavy metals: Annual statistics for Chromium (Cr), Nickel (Ni), Copper (Cu), Arsenic (As), Cadmium (Cd) and Lead (Pb) measured in PM₁₀ during 2011 in Copenhagen, Århus and Odense (street) and Copenhagen (urban). For comparison is shown heavy metal concentrations measured in total suspended particulate (TSP) at Risø (rural) (ng/m³).

Pollutant	Rural	Sub-urban	Urban	Street
PM ₁₀	20 - 25	20 - 25	20 - 27	27 - 40
NO _x	10 - 13	13 - 16	25 - 45	75 - 175
NO ₂	8 - 11	11 - 13	15 - 25	30 - 55
Particle number	-	3.000 - 5.000	5.000 - 10.000	15.000 - 40.000
Cr	0.5		1.2	7.0 3.7 3.7
Ni	2.0		3.4	2.9 2.8 5.7
Cu	2.2		13	97 42 37
As	0.7		0.8	0.9 1.0 0.7
Cd	0.1		0.1	0.2 0.2 0.1
Pb	4.1		4.5	5.9 5.8 3.9
BaP		0.61		0.21
Benzene			0.9 - 1.4	

6 Finland

6.1 State and trend of (co)carcinogen emission to air

The main sources of air pollutants in Finland are energy production, industrial processes, solvent and other product use, agriculture and waste. For the land use change and forestry sector no air pollutant emissions have been estimated thus far.

6.1.1 Energy

Combustion of fuels in the energy and heat production sectors is the main source of SO₂, NO_x, particulate matter and heavy metal emissions. NMVOC and POP compounds are released especially from small combustion sources. The transport sector is a significant source of NO_x and NMVOC emissions. Emissions from the energy sector are related to the production, distribution and consumption of fuels and fluctuate from year to year due to the economic trends and variations in the energy supply structure. The availability of hydropower in the integrated Nordic electricity market has a notable effect on the emissions. In the transport sector, emissions have a decreasing trend though the use of fuels is increasing. One of the most essential emission reduction measures in the transport sector is the EU level agreement with car manufacturers on reducing vehicles' fuel consumption. Emissions from the off-road sector are increasing.

6.1.2 Industrial Processes

Emissions estimated from the industrial processes sector include, among others, NMVOCs from pulp and paper, chemical and food and drink industries, total reduced sulphur compounds' (TRS) emissions from chemical and pulp and paper industries (reported as SO₂), heavy metal, POP and particle emissions from metal industry, and POP emissions from mineral and chemical industries. The trends are in general decreasing but variations due to fluctuations in production occur annually.

6.1.3 Solvent and other product use

The inventory of the solvent and other product use sector covers NMVOC compounds, particles, heavy metals and POP compounds. Paint application and printing are the most significant NMVOC sources. The trends of emissions are decreasing while emissions due the use of products have not yet been fully studied and included in the inventory. Efforts have been made to include those emissions that can be calculated with the existing information, but in many cases there is lack of both methods and activity data to quantify emissions from many product use sources. Several projects are, however, under way to study emissions from the use of products.

6.1.4 Agriculture

Agriculture is a source of particle emissions arising from animals and field activities. The emissions trends are decreasing due to decreases in the numbers of livestock.

6.1.5 Waste

Emissions from the waste sector include SO₂, NO_x, NMVOC, particulate matter, heavy metals and POPs. The trends of these emissions are generally declining.

6.2 Emission inventory

Finland has a comprehensive emission reporting system that produces annual inventories to the use of, e.g., CLRTAP. At the moment however, the inventory does not include spatial emission allocation apart from the 50 x 50 km² EMEP gridding. In addition to the inventory system, there is a separate national level integrated assessment model system called Finnish Regional Emission Scenario (FRES) model (Karvosenoja 2008) which includes partly the same pollutants as the CLRTAP inventory and, furthermore, spatial and temporal allocation to 1 x 1 km² and 1 hour resolutions, respectively. The (co)carcinogens that are included in both systems are particulate matter (TSP, PM₁₀, PM_{2.5}, PM₁), SO₂, NO_x and NMVOC. In addition, FRES calculates emissions of black carbon (BC) and organic carbon (OC) that are not yet part of the CLRTAP reporting.

The FRES system, being a scenario oriented emissions-to-effects model, operates with annual average emission factors, and therefore the FRES emission amounts are not necessarily identical to CLRTAP inventory reports. Relatively big difference is in residential wood combustion emissions, which is due to outdated emissions factors in the CLRTAP inventory. The FRES emission factors for residential wood combustion will be updated to the CLRTAP inventory in the near future. Furthermore, there are some sectoral differences in emission source definitions. The biggest difference is in road traffic non-exhaust emissions, where CLRTAP inventory includes only the direct wear products from road, tyres and brakes, while FRES includes also the resuspension of road dust. In the submission, road dust emissions are based on FRES calculation.

Table A2 (Appendix A) shows annual 2010 emissions from the CLRTAP inventory. The 1x1 km² gridded emission set (Appendix C) includes 2010 emissions based on the FRES model. The exception here is traffic exhaust emissions, that were taken, for this project, from CLRTAP inventory as Finnish sum emissions, and then gridded with the FRES model gridding procedure to 1x1 km² resolution. This can be seen as a demonstration of possibilities to grid emissions from other inventories or other sources.

The spatial allocation (i.e. gridding) procedure in the FRES model relies on various GIS and municipality level information for different emission source sectors, e.g. road network and traffic density information for the allocation of traffic emissions (see details in Karvosenoja et al. 2011; Tainio et al. 2009).

6.3 Exposure modelling

The Finnish Meteorological Institute (FMI) has during the past 10 years been involved in many studies for evaluating the population exposure in the Helsinki Metropolitan Area, Finland, Baltic regions and Europe (e.g. Karppinen et al., 2012; Kukkonen et al., 2008; Kousa et al., 2002 & 2005; Loh et al., 2009ab; Soares et al., 2010). Especially the exposures to NO_x and particulate matter (PM_{2.5} and PM₁₀) have been assessed frequently for different spatial domains and years and also for different source categories. Below an overview of Finnish exposure studies during the past 10 years are presented to-

gether with selected concentration and exposure results from the most recent studies including various spatial domains and emission sources; for local scale traffic is modelled and for regional scale all sources are modelled.

6.3.1 Local scale studies: Exposure assessments for Helsinki metropolitan area

Exposure assessment studies in Helsinki Area were initiated in the early 2000's. Kousa et al. (2002) describes the development of the EXPAND model (EXPosure to Air pollution), which incorporates spatial and temporal information on population activity patterns as well as urban-scale and street canyon dispersion models to predict spatial population exposure distributions, see Figure 9.

Figure 9 EXPAND-FMI exposure model (Kousa et al., 2002).

Figure 10 shows results from one practical application of the model for exposure assessment in Helsinki metropolitan area (Kukkonen et al., 2008).

The number of population ($>30 \mu\text{g}/\text{m}^3$)
2002 klo 7-8

Figure 10 The predicted number of persons that are exposed to the concentration of nitrogen dioxide exceeding $30 \mu\text{g}/\text{m}^3$ during the morning rush hour in 2002 in the Helsinki Metropolitan Area, on a characteristic winter day (upper map), and during a day with a prevailing surface temperature inversion (lower map) (Kukkonen et al., 2008).

The exposure modelling was later extended to cover also intake fractions (iF). The iF concept gives a measure of the portion of a source's emissions that is inhaled by an exposed population over a defined period of time, being defined as intake via relevant exposure pathways divided by the emissions of the pollutant and source (or source category) of interest. The numerator can be calculated using exposure estimates for the microenvironments of interest and the average breathing rate of a population while in each microenvironment. For instance, an intake fraction of 1 in a million (10^{-6}) means that for every ton of a pollutant emitted, 1 g is inhaled by the exposed population. iF can be useful in both screening-level order-of-

magnitude estimates and more detailed policy modelling (Bennett et al., 2002). So far, the iF has most successfully been applied for non-reactive and persistent compounds. An example of these new exposure modelling methods is presented in Loh et al. (2009) where the population-based iF distributions of benzene was analysed from local vehicular traffic in the Helsinki Metropolitan Area. Three different methods for exposure analyses were described and illustrated: firstly the use of the EXPAND-FMI model, secondly the use of data from the personal monitoring study EXPOLIS (Air Pollution Exposure Distributions of Adult Urban Populations in Europe) to estimate the intake fractions and thirdly a simple method based on a one-compartment box model that provides iF estimates within an order-of-magnitude or better for non-reactive agents in an urban area. One of the novelties of the study was that it was able to present detailed iF distributions on several different levels (spatial, individual and generic) for the same urban area. Detailed results and discussion on the pros and cons of the different methods can be seen in Loh et al. (2009).

6.3.2 Recent local scale studies: Exposure to traffic pollution in Helsinki Metropolitan Area (HMA)

Modelling of the population exposure of PM_{2.5} from traffic in HMA has been made for the year 2008. Although the traffic contribution is not directly proportional to the total PM_{2.5} concentrations and is thus not representing the total burden of PM_{2.5} concentrations to health, it is still a very crucial exposure component in HMA and also the key parameter in assessing the health outcomes of different future traffic scenarios in the area.

The population exposure, total and for five microenvironments, was calculated for HMA, based on the annual averaged traffic originated PM_{2.5} concentrations (Figure 11) computed for 18,692 receptor points. The output of the dispersion modelling consisted of hourly time series of concentrations for the calculation year. Population location data is collected annually by the municipalities of HMA. The current dataset contains data on the dwelling houses, enterprises and agencies located in the HMA based on the year 2009 (SeutuCD - Greater Helsinki database (HSY) - www.hsy.fi/sepe). The data set provides geographic information on the total number and age distribution of people living in a specific building or the total number of people working at a specific workplace. The information on the number and location of people in shops, restaurants and other recreational activities is also based on this data set. The location of people in traffic was evaluated using the computed traffic flow information; this information is available separately for buses, cars, trains, trams, metro, pedestrians, and cyclists for each street and rail section on an hourly basis.

The total exposure results, expressed in persons*µg/m³ (Figure 12), and the exposure maps in different micro-environments (Figures 13 to 15), clearly show the expected results; for traffic originated PM_{2.5} people will mostly get exposed in-traffic (Figure 15), even though the time spent in traffic is much less than the time spent at home (Figure 13) or at work (Figure 14). Looking at the different indoor microenvironments, results also show that people are more exposed to traffic PM_{2.5} at work than at home locations. This result can be explained by examining the locations of work places and home locations; work places are concentrated in the downtown area of Helsinki and the home location at the suburbs of Helsinki. The combination of higher activity at the downtown areas, and high concentration levels (Figure 11) explain the higher values of exposure at work.

To evaluate how the exposure values are affected by the input, static population exposure (only home addresses are taken into account) was modelled. Computations were also made utilising the dispersion modelling results described above and population data provided by the Statistics Finland (www.stat.fi) with a resolution of 250x250 m². The static exposure differs from the dynamic exposure by not taking the time-activity data into account. The results of the static population exposure are shown in Figure 16 (interpolated values by nearest neighbour interpolation method). Mean exposures in $\mu\text{g}/\text{m}^3$ multiplied by the number of exposed persons gives mean and maximum static PM_{2.5} exposure values of 68 and 1595 no. persons* $\mu\text{g}/\text{m}^3$, respectively. The results show a similar distribution as the home exposure (Figure 13), but with lower values. The lower values may be the result of the interpolation. The difference is accentuated by the amount of time people have spent at a certain receptor point (dynamic-exposure).

The results of these studies are used as baseline exposure estimates in the EU/TRANSPHORM project (<http://www.transphorm.eu/>) where changes in exposure are evaluated with several future city and traffic planning scenarios to find an optimal way to minimize the traffic originated exposure in HMA.

Figure 11 Annual average concentrations of PM_{2.5} in Helsinki Metropolitan Area 2008 ($\mu\text{g}/\text{m}^3$).

Figure 12 Total exposure to PM_{2.5} concentrations in Helsinki Metropolitan Area in 2008 (no. persons* $\mu\text{g}/\text{m}^3$).

Figure 13 Exposure to PM_{2.5} concentrations *at home* in Helsinki Metropolitan Area in 2008 (no. persons* $\mu\text{g}/\text{m}^3$).

Figure 14 Exposure to PM_{2.5} concentrations *at work* in Helsinki Metropolitan Area in 2008 (persons*µg/m³).

Figure 15 Exposure to PM_{2.5} concentrations *in traffic* in Helsinki Metropolitan Area in 2008 (no. persons*µg/m³).

Figure 16 Static exposure to PM_{2.5} concentrations in Helsinki Metropolitan Area in 2008 – interpolated (no. person*µg/m³).

6.3.3 Regional studies: Concentrations and exposures in Finland for 2010

The most recent exposure analyses for Finland are done for the year 2010 for the (co)carcinogens NO₂, PM_{2.5} and PM₁₀, see Appendix D. The results presented here are based on surface concentration fields from a regional-scale chemical transport model - SILAM (System for Integrated modeLing of Atmospheric cOMposition, SILAM (Sofiev et al. 2006, 2008) with a horizontal resolution of 0.2x0.2 deg = ~20x20 km², and TRANSPHORM population distribution data (reference) with a resolution of 0.3x0.2 deg = ~30x20 km². The population data is based on the 2001 JRC population density grid (Gallego, 2010) and on Eurostat data

(http://epp.eurostat.ec.europa.eu/portal/page/portal/gisco_Geographical_information_maps/geodata/reference) and scaled for the year 2005. The latter was regridded (nearest neighbor interpolation method) to a 0.2x0.2 deg resolution. These quite coarse-grid exposure figures serve well in displaying the spatial distribution of exposure to different pollutants in Finland. Note that the surface concentrations provided by SILAM model include all the emissions (traffic and non-traffic) derived from the TNO European gridded emission inventory for anthropogenic sources (van der Gon et al., 2009) and sea salt (Sofiev et al., 2012).

The regional and country-wide static population distribution exposures (Figure 17) are useful for long-term, regional or country wide health effect studies, however, they are not appropriate e.g. in more detailed city or traffic planning (see also Karppinen et al., 2010).

Figure 17 Modelled annual static exposures to all anthropogenic NO_2 , and all anthropogenic and natural (sea salt) PM_{10} and $\text{PM}_{2.5}$ for year 2010 ($\mu\text{g}/\text{m}^3 \cdot \text{persons}$) in Finland and near-by areas. Spatial resolution is approximately $20 \times 20 \text{ km}^2$.

Table 7 Exposure concentrations at regional scale 0.2×0.2 deg (no. person $\cdot \mu\text{g}/\text{m}^3$) in Finland.

Exposure	Average	Maximum
NO_2	2.33×10^4	5.18×10^6
$\text{PM}_{2.5}$	2.30×10^4	4.36×10^6
PM_{10}	2.53×10^4	5.04×10^6

7 Norway

7.1 State and trend on (co)carcinogen emissions to air

7.1.1 Sulphur dioxide (SO₂)

Manufacturing, mining and quarrying accounted for 68 % of the SO₂ emissions in 2010. The emission trend is heavily influenced by the production activity in these industries. The long-term emission trend has been a decrease. The main part of the reduction appeared before 1993, which was a consequence of lower sulphur content in oil products, as well as measures and closures within manufacturing and mining. From 1980 to 2010, total emissions decreased by 86 %.

7.1.2 Nitrogen oxide (NO_x)

The emissions of NO_x were 14 % above the level in 1980, but 3 % below the 1990 level. From 1990 to 1999, the emission increased considerably and reached a historical peak of 224 000 tonnes. After this 1999 peak, the emission dropped by 17 % to reach the 2010 level. Domestic sea transport and fishing accounted for 28 % of the total Norwegian NO_x emission in 2010. Stationary combustion in oil and gas-related activities contributed with 27 %, and road traffic with 20 % of the NO_x emissions. Manufacturing and construction made up the principal part of the remaining quarter. The last three years have shown a decline in the NO_x emissions from road traffic, due to new cleaning technologies for exhaust from heavy duty diesel vehicles and a generally newer car fleet. However, the emissions from diesel fired engines used within oil extraction increased during the latter years and curbed the overall reduction in NO_x emissions. In 2008, the emission from ships and boats dropped by as much as 13 per cent, but has remained stable since then. Increased fuel use by fishing vessels has offset the reductions due to technical efforts in this sector. Activity within manufacturing of iron, steel and ferroalloys and construction are the main causes of year-to-year fluctuations. In 2010, 1 % of NO_x emissions came from residential fuel combustion, including wood burning. Emissions of NO_x from residential fuel combustion was reduced by nearly 40 % from 1980 to the beginning of the 1990-ties, but has been relatively stable since.

The NO_x fund was established in 2008 as an instrument for financing abatement measures for NO_x in industry. According to information given by the fund, measures were implemented during the period 2006-2010 that gave verified reductions of approximately 12 000 tonnes in 2010. A total of 1 000 tonnes of reductions that have not yet been verified were also reported. Without these reductions, the NO_x emissions in 2010 would have been about 6-7 % higher.

7.1.3 Non-methane volatile organic compounds (NMVOC)

From 1980 to 2001 emissions of NMVOC more than doubled, due to increased activity in the oil industry. Improved loading technology in the oil industry was then introduced, leading to a large fall in emissions. In 2007, 86 % of the loading of oil took place at installations with NMVOC reducing equipment, and emissions of NMVOC had decreased by 70 % compared to 2001. By 2010 emissions from the oil industry had declined by another 19 % and was for the first time since the early eighties only the second largest

source. The largest source of NMVOC emissions in 2010 was use of solvents, which accounted for 30 % of the emissions in 2010. Road traffic constituted more than 50 % of NMVOC emissions in 1980. In 2010 road traffic accounted for 9 %, decreasing by almost 80 % since 1980. The decrease was a result of the requirements for exhaust gas purification which were introduced in 1989.

7.1.4 Total Suspended Particulate matter (TSP) and Particulate Matter (PM)

Sixty-two %, or about 33,000 tonnes, of the emissions of PM₁₀ in 2010 came from wood-burning. The wood consumption increased by 11 % from 2009 to 2010. At the same time, the calculated emissions of particulate matter from this source increased by only 8 %. This is due to a higher share of wood stoves with new technology. Process emissions from manufacturing and mining amounted to 14 % of the emissions of particulate matter in 2010. Motorized equipment and road dust/tyre wear were responsible for 5 and 3 % of the emissions, respectively.

7.1.5 Cadmium (Cd)

Emissions to air of cadmium totalled 590 kg in 2010. Wood-burning in private households and burning of refuse wood and waste liquor in the wood processing industry are the most important sources of emissions of cadmium to air. In 2010, these sources contributed with 25 % of the total emissions each. Cadmium emissions have been reduced by 50 % since 1990, and the main reason for this is reduced emissions from production of iron, steel and ferroalloys due to emission reduction efforts and the closing down of production plants. This source was responsible for 7 % of cadmium emissions in 2010, compared with 26 % in 1990.

7.1.6 Mercury (Hg)

Emissions of mercury were 584 kg in 2010. The most important source of mercury emissions is combustion without energy utility, which comprises cremations. The emissions have been considerably reduced since 1990, but have been relatively stable since 2000. The main reason for the decline is less process emissions from production of iron, steel and ferro alloys. Around 4 % of mercury emissions arise from residential fuel combustion, including wood burning.

7.1.7 Arsenic (As)

In 2010, 1.8 tonnes of arsenic were emitted to air. For the past few years, the variation in arsenic emissions has been due to varying arsenic content in raw materials and reducing agents used in metal production. The total arsenic emissions have been reduced by 48 % since 1990. Wood-burning in private households contributed with nearly 13 % of the emissions in 2010.

7.1.8 Lead (Pb)

Emissions of lead and copper have been relatively constant in recent years. Brake wear is now the dominating emission source and was responsible for nearly 28 % of the total lead emissions to air in 2010. Twenty-one % were process emissions from the metal industry, mainly from the production of iron, steel and ferro alloys. The emissions from the production of iron, steel and ferro alloys were reduced by 76 % from 1990 to 2010, almost half of which took place between 2008 and 2009. Use of lead-containing petrol in

domestic aviation accounted for 23 % of the 2010 lead emissions. Residential fuel combustion, including wood burning, generated 2 % of lead emissions to air in 2010.

7.1.9 Polycyclic Aromatic Hydrocarbons (PAHs)

Wood-burning in private households was the largest source in 2010, making up 39 % of total emissions of PAH. However, for emissions of the 4 most important PAHs (PAH-4), process emissions in aluminium production was the most dominant source of emissions, contributing with 51 % of these emissions to air in 2010. The PAH-4 emissions have decreased primarily because of the discontinuation of Soederberg technology in the aluminium production. The emissions from manufacture of iron, steel and ferro alloys also decreased. Road traffic contributed to 10 % of the total PAH emissions in 2010. This includes both exhaust and tyre and brake wear.

7.1.10 Dioxins and furans (PCDD/ PCDF)

In 2010, 26 grams of dioxins were emitted to air. Since 1990, the emissions have been reduced by 80 %. A large proportion of this reduction is due to the shutting down of one industrial plant and one mine. In addition, emissions from energy supply were reduced by 95 % from 1990 to 2010 as a result of more extensive cleaning measures at waste incineration plants. Since 2005, the emissions have been relatively stable. The most important emission source in 2010 was wood-burning in private households, which caused 34 % of total emissions. Combustion emissions from navigation and process emissions from metal production accounted for 17 and 10 % of the total respectively.

In Table A3 (Appendix A) Norwegian emissions for the year 2010 are shown for main source categories.

7.2 Emission inventory

Statistics Norway annually publishes figures on emissions to air of acidifying substances, ozone precursors particulate matters, heavy metals and POPs. The times series are continuous for all these substances from 1990 onwards, and they are recalculated every year. These emissions are calculated on a national level, distributed between different activities, fuel types and/or industries. In addition, emissions of a number of several hazardous substances from 2002 and onwards are calculated using data from the Norwegian Product register. However, these figures have not yet gained the status of official statistics. The result of these calculations for substances that may cause cancer or are proven cancerous is given in Table 3. The use of these substances has been relatively constant between 2002 and 2010, amounting to around 500,000 tonnes annually. Raw materials and reduction agents make up 86 % of this consumption. Most of this is used in closed systems in chemical industry. Thus, emissions are calculated to be between approximately 100 and 300 tonnes every year. Note that these figures include emissions to all recipients as a whole, not only emissions to air.

Emissions by EMEP 50 km x 50 km² grid square are reported to the UNECE and used in models of long-range air pollution. The emissions are allocated to grid squares as follows:

- Emissions from *large point sources* are allocated directly to the appropriate squares. From 2000, this also includes emissions from offshore petroleum activities.
- Emissions at sea from *national sea traffic and offshore petroleum activities* (before 2000) are allocated to squares on the basis of a detailed analysis of 1993 activity data (Flugsrud and Rypdal, 1996). The 1993 emissions are projected using national emission trends for each of the categories fishing, other sea traffic, flaring, other combustion, and other emissions in the petroleum sector.
- The *remaining emissions* in each of the 430 municipalities are allocated to squares according to the proportion of the area of the municipality in each square. This part requires emissions to be distributed among municipalities. The allocating of remaining emissions to the municipalities has been done in one of two ways:
 - When figures for the activity used to calculate emissions were available directly at municipal level, these figures were used. Examples are fuel combustion in manufacturing industries and emissions from animals.
 - When the activity at the municipal level was unknown, the national emissions were allocated indirectly using surrogate statistical data. For example, fuel combustion in service industries was allocated using employment figures. In a number of cases the activity is known directly at the intermediate level (county), but allocation within counties uses surrogate data.

The use of figures on emissions from municipalities introduces several sources of error. Firstly, updated national emission figures are no longer allocated to municipalities. In 2011 it was decided to suspend this statistics. The uncertainty in the method used to allocate national emissions to geographical units was considered to be unjustifiably large. Thus, when gridded data for 2010 were reported in 2012, the geographically distributed figures published in 2011 were scaled using the latest figures on emissions on the national level. The emissions from large point sources were treated separately.

Secondly, the method assumes that emissions are evenly distributed within municipalities. In reality, emissions often occur only in small parts of a municipality. If a municipality is large relative to the grid squares, the emissions may be allocated wrongly. However, few municipalities measure more than 50 km across. The median municipality size is 469 km². Furthermore, the larger municipalities are usually sparsely populated, with small emissions. It is therefore assumed that the level of error due to the method is acceptable. The direct allocation of large point sources also reduces the potential error.

Emission mapping of the various pollutants requires emission factors, which at present are not available. Urban scale (1 x 1 km²) emissions are available in AirQUIS for Oslo, Bergen, Trondheim, Stavanger, Drammen and Grenland. However not maps are produced at present and requires AirQUIS modelling of e.g. traffic and wood combustion in all cities.

7.3 Exposure modelling

The AirQUIS/EPISODE model (Slørdal et al., 2003) is the main model used to perform air pollution modelling at the urban and local scale, mainly of NO₂, O₃, PM₁₀ and PM_{2.5}. The model has been used to perform exposure assessment in the larger Norwegian cities (Oslo, Bergen, Trondheim), and in

several epidemiological (health) studies in Oslo in cooperation with national health authorities. The AirQUIS/EPISODE model cannot use the national reported emissions directly; rather, emissions must be given on a more disaggregated level. These are compiled in the AirQUIS emission data base, which contains a mix of bottom-up (road traffic) and top-down data (home heating and industry). For traffic, emissions are based on the National Road Authority data base, which contains detailed traffic data for individual streets and roads. For emissions from home heating (wood and oil), and industry, emissions are based on data from Statistics Norway which is a combination of top-down national emission inventory data, and a bottom-up scaling approach for distributing national emissions over specific geographical locations (municipalities). The emissions in the AirQUIS database only covers the pollutants NO_x (NO and NO_2), PM_{10} and $\text{PM}_{2.5}$ as gridded values with resolution $1 \times 1 \text{ km}^2$, and as individual road links (line sources) and point sources. Meteorology is either based on interpolation of local observations in the city, or more recently large scale synoptic data, e.g., from NCEP/NOAA or ECMWF, in combination with meteorological downscaling models such as TAPM and WRF. The grid resolution of emissions, meteorology and model output concentrations are typically $1 \times 1 \text{ km}^2$. The AirQUIS/EPISODE model can calculate the individual contribution from any source group, such as e.g., from private wood burning.

When run for an urban area, the AirQUIS/EPISODE model produces ground level hourly average concentrations in grids and individual receptor points based on emissions, meteorology and regional scale background concentrations. The latter is usually taken from nearby regional background stations. The results are usually directly applicable for epidemiological studies, except that indoor concentrations are not available. A concise description of the model can be found on the EIONET model documentation system web site: <http://acm.eionet.europa.eu/databases/MDS>.

The regional scale EMEP model is used regularly to calculate annual concentrations and dry and wet depositions in a $50 \times 50 \text{ km}^2$ grid over Norway. The data, which include the (co)carcinogens NO_2 , PM_{10} , $\text{PM}_{2.5}$ and SO_2 , have been stored for each year in the period 1995-present and can be retrieved from the main web site: <http://www.emep.int/mscw>. In principle these data should also be applicable to epidemiological studies, albeit that the grid resolution is somewhat coarse.

During each winter season (from October-April) the AirQUIS/EPISODE model is used as part of an on-line air quality forecast system in Norway called Better City Air (Bedre Byluft) (Ødegaard et al., 2011). In this forecast system, hourly prognoses of air quality (NO_2 , PM_{10} and $\text{PM}_{2.5}$) for the coming 2-3 days are made on a daily basis for several of the major Norwegian cities (including Oslo, Bergen, Trondheim, Stavanger and Drammen). The system has been operative, in cooperation with met.no, since the late 1990's, but unfortunately there has not been any systematic storage of the results, so the data are generally not retrievable.

For Oslo, for each year in the period 1992-2002, ground level hourly average concentrations of NO_2 , PM_{10} and $\text{PM}_{2.5}$ in a $22 \times 18 \text{ km}^2$ grid and individual receptor points (between 5 000 and 10 000 building points) have been calculated by the AirQUIS/EPISODE model system and stored (McInnes, 2004). These data are easily retrievable, and have been used in several epidemiological studies performed by The Norwegian Institute of Public Health. The

data consist of total concentrations from all source groups in Oslo, not from any individual source group.

In a project for the Norwegian Pollution Control Authority (Slørdal et al., 2007a and 2007b), NILU has made a set of maps of air quality for the cities Oslo, Bergen and Trondheim. The maps were produced using the AirQUIS/EPISODE model system and consist of concentration levels in grids with resolution $100 \times 100 \text{ m}^2$ and road side concentrations. Emissions from traffic (line sources) were given with this fine resolution in these calculations; all other sources, including domestic heating from wood and oil burning, were given with resolution 1 km. For Oslo, the pollutants NO_2 , PM_{10} and $\text{PM}_{2.5}$ were modeled for the year 2005, while NO_2 and PM_{10} were modeled for Bergen and Trondheim for 2003 and 2005 respectively.

The results for Oslo and Trondheim are based on model calculations performed in the project "Rikets Miljøtilstand 2005" (Slørdal et al., 2006), while the results for Bergen is based on model calculations performed in the project "Rikets Miljøtilstand 2003" (Laupsa et al., 2005). In these two projects, NILU performed dispersion and exposure calculations of NO_2 , PM_{10} and benzene (C_6H_6) for the year 2005 for Oslo (Figure 18) and Trondheim (Figure 19), and for 2003 also for Bergen. The number of inhabitants exposed to exceedances of the National Air Quality Target for these three pollutants were computed both in building points and in model grids with 1 km resolution. The calculations for 2005 for Oslo showed that 0.12 %, 44.8 %, and 6 % of the Oslo population were exposed to exceedances of the National air quality target for NO_2 , PM_{10} and benzene, respectively. The corresponding numbers for Trondheim were 0.026 %, 13.8 % and 0 %, respectively. Calculations of the percentage contribution from the various sources showed that road traffic were the dominant source to the exceedances of the national target for both NO_2 and PM_{10} . For PM_{10} , domestic wood burning was clearly the second most dominant local source. No calculations of source contributions were made for benzene, however, but earlier investigations (Laupsa et al., 2005) has shown that road traffic is by far the most dominant source for exceedances with regard to this pollutant as well.

Figure 18 Modelled annual mean benzene concentrations ($\mu\text{g}/\text{m}^3$) for Oslo in 2005.

Figure 19 Modelled annual mean benzene concentrations ($\mu\text{g}/\text{m}^3$) for Trondheim in 2005.

The calculations were repeated for Oslo and Trondheim for the year 2007 (including benzene) (Sundvor et al., 2009), again showing that road traffic is the most dominant source to exceedances of the national target for NO_2 and PM_{10} for both Oslo (93.5 % and 72.3 %, respectively) and Trondheim (98.9 % and 83.5 %), with wood burning the second most dominant source for PM_{10} with 20.7 % in Oslo and 10.1 % in Bergen. However, the percentage contributions to exceedances were not calculated for benzene in this study.

For benzene, no modelling results more recent than 2007 exists, but benzene has been observed (as monthly mean values) at a number of monitoring stations in Norway over the years 2001-present (Gjerstad et al., 2013). All observations show annual mean concentrations below the limit value of $5 \mu\text{g}/\text{m}^3$, which has been valid since 2010. However, all annual mean values before 2006 (except at 2 stations in Kristiansand in Norway) have been above the National Target in Norway which is $2 \mu\text{g}/\text{m}^3$. The concentration of benzene in Norwegian cities is quite seasonal dependent, with the highest concentrations typically during the winter period (due to reduced photochemical activity, poor dispersion and added emissions from wood burning).

Maps of annual background concentration of NO_2 , PM_{10} and $\text{PM}_{2.5}$ over Norway, with a resolution of $11 \times 6 \text{ km}^2$ have recently been made (Schneider et al., 2011). These maps have been derived using geostatistical methods,

and involve the combination of highly accurate station measurements with a range of spatially distributed auxiliary variables such as elevation, population density, incoming radiation and model output, used in order to aid the interpolation in areas of low station density. The annual maps have further been complemented with a temporal decomposition of the time series at each available measurement station into seasonal and daily cycles. This is done in order to allow for a more realistic estimate of the background value at specific dates and times.

For other pollutants such as heavy metals and POPs no modelling results exist for Norway, so for these we have only observations, mainly at the regional background stations.

In Table 8 is shown annual mean concentrations averaged over (mainly) roadside stations for some selected cities and areas in Norway in 2011. The data are based on the available annual reports which are made by local authorities for each city or area in Norway. The reports can be downloaded from the web site: <http://www.luftkvalitet.info> (reports section)).

Table 8 Observed annual mean concentrations averaged over (mainly) roadside stations for some selected cities and areas in Norway in 2011 (number of stations in parentheses). Unit: $\mu\text{g}/\text{m}^3$ (except for Benz(a)Pyrene which is in ng/m^3).

Site	NO ₂	PM ₁₀	PM _{2.5}	Benzene	Benz(a)Pyrene (ng/m^3)
Oslo	47 (6)	22 (8)	12 (7)	4.3 (2)	0.34 (2)
Bergen	36 (2)	19 (2)	(2)	2.4 (1)	0.67 (1) (for 2010)
Trondheim	35 (4)	23 (4)	9 (4)	2.8 (1)	0.25 (1)
Stavanger	44 (1)	26 (1)	-	-	-
Grenland	16 (5)	21 (3)	12 (1)	2.6 (1)	-
Mo i Rana	-	21 (1)	-	-	-

8 Sweden

8.1 State and trends of (co)carcinogen emissions to air

8.1.1 Sulphur dioxide (SO₂)

Since 1990 emissions of SO₂ have decreased significantly, about 67 %, from 105 Gg to approximately 35 Gg. The decrease is primarily due to a shift to fuels with lower sulphur content in the energy and the transport sector and the installation of desulphurisation plants. Today the most important sources of SO₂ are industrial processes (35 %) and energy production (30 %). Wood combustion in residential plants constitutes less than 5 %.

8.1.2 Nitrogen oxide (NO_x)

The total emissions of NO_x have been reduced by 40 % from 1990 (269 Gg) to 2010 (161 Gg). The greatest reduction of emissions has been performed within the energy sector. The reduction of the NO_x emissions in the energy sector is mainly due to installation of low-NO_x burners within the sector as a consequence of the introduction of the national “Nitrogen Oxide Charge” in the early 1990s. The reduction of NO_x emissions in the transport sector is mainly due to the introduction of catalytic converters on new petrol cars and the subsequent successive tightening of road transport exhaust emission standards in general. Today the largest sources of emissions of nitrogen oxides are in the energy sector: road traffic (42 %), stationary combustion (10 %) and electricity and heat production (10 %). Wood combustion in residential plants accounts for less than 5 %.

8.1.3 Non-methane volatile organic compounds (NMVOC)

Emissions of volatile organic compounds were 197 Gg in 2010. Emissions have been reduced by 45 % compared to 1990 (359 Gg). Over time road traffic has shown the greatest reduction in emissions due to new exhaust emission requirements, but environmental requirements in the installation of new wood-fired boilers have also contributed to lower emissions. Today the main sources are use of products containing solvents (40 %), road traffic (20 %), and wood combustion in the residential sector (10 %).

8.1.4 Total Suspended Particulate matter (TSP) and Particulate Matter (PM)

The emissions of TSP, PM₁₀ and PM_{2.5} were 51, 44 and 32 Gg respectively in 2010. The emissions have been reduced by about 15 % compared to 1990 for all three fractions. The largest source of emissions of TSP and PM is the energy sector, which contributed with approximately 75 % of emissions in 2010. Road transport (TSP 40 %, PM₁₀ 25 %, PM_{2.5} 20 %) and wood combustion in residential plants (TSP 15 %, PM₁₀ 20 % and PM_{2.5} 25 %) dominates the emissions within the energy sector.

8.1.5 Cadmium (Cd)

The emissions of cadmium were 0.583 Mg in 2010, which corresponds to a decrease by 74 % compared to 1990 (2.27 Mg). The reduction is mainly due to installation of efficient abatement equipment in metal smelters and steel mills. The main source today is wood combustion in residential plants (35

%), electricity and heat production and stationary combustion (30 %), since most fuels, both fossil and biofuels, naturally contain cadmium.

8.1.6 Mercury (Hg)

The mercury emissions to air have decreased by 65 % from 1990 (1.58 Mg) to 2010 (0.554 Mg). In the 1980s the emissions from waste was a major source, but this source became less important in the 1990's due to the ban on mercury in many products, improved waste sorting and improved abatement equipment in waste incinerator plants. From 1990 the emissions from crematoria (included in the waste sector) have fallen from more than 250 kg to about 115 kg in 2010. Today the main sources of mercury are electricity and heat production (35 %), industrial processes (26 %) and waste (21 %). Wood combustion in residential plants constitutes approximately 5 %.

8.1.7 Lead (Pb)

The emissions of lead have decreased by 96 % from 1990 (355 Mg) to 2012 (13 Mg). Emissions of lead to air were very large in the 1980s and earlier. Emissions of lead from traffic fell sharply when unleaded petrol was introduced in the late 1980s, when catalytic converters became mandatory in new petrol cars. Today the most important sources are passenger cars (30 %), industrial processes (25 %) and public electricity and heat production (20 %). Wood combustion in residential plants stands for approximately 10 % of the total emissions.

8.1.8 Copper (Cu)

The emissions of copper have decreased by 45 % from 1990 (98 Mg) to 2010 (54 Mg). Metal smelters were previously the largest source of emissions of copper to air, but these emissions have decreased considerably due to improved abatement equipment. Today the most important source is road transport (90 %), due to the fact that copper is used in the brakes of the automobiles.

8.1.9 Chromium (Cr)

The emissions of chromium were 5.11 Mg in 2010, which corresponds to a decrease by 78 % compared to 1990 (23.3 Mg). The reduction is mainly due to installation of efficient abatement equipment in metal smelters and steel mills. The main source of chromium, both historically and today, is process emissions from the iron and steel industry, which contributed with approximately 70 % of the total emissions in 2010. During 2009 and 2010 the emissions of chromium dropped. This is mainly due to reduced production of ferrochrome in the ferroalloy industry.

8.1.10 Polycyclic Aromatic Hydrocarbons (PAHs)

The emissions of PAH (sum-4) have decreased by 2 % from 1990 (16.8 Mg) to 2010 (16.4 Mg). Emissions from wood boilers, stoves and other small-scale burning of wood approximately account for two-thirds of the Swedish emissions of PAH. Emissions from burning wood are largely unchanged since 1990 despite an increase in the burning of wood fuel. The increase in emissions has been offset by newer wood and pellet boilers having more efficient combustion. The aluminium plant outside Sundsvall was previously the largest single source, but from 2009, emissions decreased close to zero due to exchange of production process.

8.1.11 Dioxins and furans (PCDD/ PCDF)

Emissions of dioxins and furans have decreased by 29 % from 1990 (60 g) to 2010 (43 g). The main source of PCDD/PCDF in Sweden is the energy sector (mainly public electricity and heat production) which contributed with approximately 90 % in 2010. As a result of an increased use of biofuels (wood chips, bark) within this sector the emissions of dioxins have increased by approximately 50 % between 1990 and 2010. On the other hand industrial processes emissions within the metal industry (secondary steel production) have decreased by approximately 90 % during the same period. Wood combustion in residential plants stands for approximately 10 % of the total emissions.

8.1.12 Hexachlorobenzene (HCB)

The Swedish reporting of HCB according to CLRTAP is considered incomplete, due to the fact that only emissions originating from national navigation (shipping) and national fishing are included in the inventory.

In Table A4 (Appendix A) Swedish emissions for the year 2010 are shown for main source categories.

8.2 Emission inventory

SMHI, IVL, SCB and SLU work together in the SMED (Swedish Methodology for Environmental Data) consortium on behalf of the Swedish EPA, and maintain a spatially distributed national emission inventory. The inventory is updated every year and is, as far as possible, consistent with the international reporting under UNFCCC and CLRTAP. Also, since the results are used for trend analysis on regional level, a retroactive update of all years is done on a yearly basis. This way, the tendencies seen in the data can be regarded as real and not just a consequence of changes in methodology. So far, the inventory includes the years 1990, 2000 and 2005-2010. The inventory is a mixture between a top-down and a bottom-up approach. As far as possible, emissions are allocated to individual emission sources with rather exact locations. Only the part of the total emissions that cannot be explained by known individual sources is distributed using proxy data, such as population density. All gridded information in the inventory has a spatial resolution of 1 x 1km². The inventory covers the pollutants required by CLRTAP (and greenhouse gases), with exception of HCB, PCB and individual PAHs (benz(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene and indeno (1,2,3-cd) pyrene), which because of incomplete information and large uncertainties have not been included in the spatially distributed inventory.

Emissions are stored in a dedicated emission database and can be exported routinely to any geographic coordinate system or projection. All emissions are coded on the finest code level available, which in most cases corresponds to NFR (as in CLRTAP) or CRF (as in UNFCCC). Emissions can automatically be aggregated to any compatible categorization. Emissions can be gridded and exported in standard GIS-formats. The parts of the inventory described by individual sources can be exported as point-sources, area-sources and line-sources. Together with the inventory, documentation is maintained that in more detail describes the methodology for the different sectors. The data is also published online at:

<http://utslappisiffror.naturvardsverket.se/en/Emissions-to-Air/>

Emissions have been aggregated into the prescribed sectors. Emission categories not covered by these sectors (such as waste management and aviation) have been added in separate sectors. The emission grids have been exported as ESRI ascii grids in the geographical coordinate system WGS-84. In addition, emission data has been aggregated as national totals and as share of national total for private wood burning and traffic. The grids have been delivered as shape files in orthogonal grids in the geographical coordinate system WGS-84. All grids have a resolution of 0.01 degrees (which is close to 1x1km²).

Benzene is not included specifically in the inventory. It can however be estimated from the emission of NMVOC. For this purpose the factor: 0.03448 (mass Benzene/mass NMVOC) can be used, which has given reasonable results in verifications using dispersion models.

8.3 Exposure modelling

In SMED SMHI is data host for Swedish emissions and responsible for the geographical distribution on a 1x1 km² grid. Using EMEP expert emissions and the SMED 1x1 km² emissions over Sweden, the national modelling system SIMAIR (Gidhagen et al., 2009) produces 44x44 km² regional European scale concentrations grids over the entire country and 1x1 km² urban grids over about 130 Swedish cities, down to approximately 5000 inhabitants see Figure 20, for three (co)carcinogens: PM₁₀, NO₂ and benzene. Additionally local (address) scale grids have been produced for selected sites to study the influence of traffic and private wood burning.

Figure 20 Areas for which urban contributions are calculated on a 1x1 km² resolution in Sweden.

The SIMAIR model system, used for exposure calculations all over Sweden, is based on simulated hourly concentration fields during a historic year (Gidhagen et al 2009). The most recent SIMAIR year available, 2010, was selected as a base for the exposure assessment. This means that meteorological conditions, long-range contributions and emissions reflect the conditions of that particular year.

The European contribution to the regional scale PM₁₀, NO₂ and benzene was calculated on a 44x44 km² grid, using the photochemical transport model MATCH (Robertson et al., 1999). Model output for PM₁₀ has been improved by a 2D variational data assimilation scheme (Robertson and Kahnert, 2007), which adjusts the simulated levels towards monitored PM10 data at three regional stations in southern, middle and northern Sweden, maintaining the spatial distribution given by the model. As for benzene emissions SMHI delivers benzene regional background concentrations to SIMAIR, but the emissions to that simulation is not based on a specific benzene inventory, instead an assumption has been made of benzene as a percentage of VOC emissions.

Urban contributions were calculated for the approximately 130 cities or villages identified as urban environments in SIMAIR, using 1x1 km² grids that covered the urbanized areas (Figure 20). PM emissions from road traffic were separated in a combustion part, based on the ARTEMIS emission model (André and Rapone, 2009), and a road-wear and re-suspended dust contribution (Omstedt et al., 2005). The road wear emission model takes into account time-varying conditions of studded tire use, road wetness and road maintenance (sanding, salting). The model is able to account for the main features in the day-to-day and seasonal mean PM₁₀ variability, especially the peaks in PM₁₀ emissions at both ends of the winter season, when studded tires are used on dry road surface.

The non-traffic emissions used on the urban scale are from the National Swedish Inventory (SMED) but spatially distributed over a nationwide 1x1 km² grid and aggregated to three sectors: residential wood combustion, sea traffic and others. The last (other) sector covers principally energy production and industrial sources, thought of as elevated point sources. Traffic emissions have a temporal variation according to typical daily patterns found in traffic volume, different for weekdays and weekend days. Emissions from residential wood combustion were set to vary over time according to 3-hourly ambient air temperature and with the requirement that temperatures should be below 10 °C for emissions to occur. Sea traffic and other emissions were kept constant over time.

The numerical simulation of urban background contributions was made on the same 1x1 km² grid as the emission inventory, using two different model approaches. For ground-level sources, i.e. traffic emissions and residential wood combustion, the simple approach of determining the influence area upwind the receptor point was used, an adjoint approach suggested firstly by Berkowicz (2000). The dispersion of stack emissions was treated in a separate Gaussian point source model, forming part of the DISPERSION model (Omstedt, 1988). The two urban models were fed with meteorological input data that are relevant for dispersion models – e.g. wind speed and direction, temperature, cloudiness, global radiation, sensible heat flux, friction velocity, humidity and precipitation (Häggmark et al., 2000).

8.3.1 Exposure grids for Sweden delivered in this project

SIMAIR exposure simulations are available from 2004 to 2010. The first grid deliveries are for 2010. For 2011 and following years there is a plan to deliver also PM_{2.5} and BaP concentrations on the same grids, thus with a possibility to extend the gridded output in future phases of the project. The first year with this extended (6) pollutant list will likely be 2011.

The first export of gridded concentration does not have any division into different sector contributions, but there are possibilities for later separation of some contributions on the urban scale, e.g. traffic and residential wood burning. Normally the SIMAIR system does not separate source-specific impacts from the existing regional or urban scale exposure concentrations, however over urban areas, where the urban model is applied on the 1x1 km² scale, there are emission relations that can be used to scale source-specific contributions. For example the $\text{Emission}_{\text{traffic}}/\text{Emission}_{\text{total}}$ and $\text{Emission}_{\text{wood}}/\text{Emission}_{\text{total}}$ ratios are available. Using these ratios as factors, the contributions of traffic and wood combustion concentrations in each urban background concentrations can be estimated and mapped in a similar way

as total concentrations. However, for most of the area there are only regional contributions, where no emission ratios are available at the moment.

The concentrations are thus delivered as annually averaged 1x1 km² grids covering all of Sweden. The regional contribution and the urban contribution (which will have high resolution information only over the 130 urban areas, see Figure 20) will be delivered as two separate grids which added together form total urban background concentrations. Note that concentration data should only be used over Swedish territory; outside Sweden they do not have good quality.

To summarize: Year 2010 annual averages of the following pollutants have been facilitated to this NMR project, all covering the Swedish territory and simulated on SWEREF99TM projection:

- NO₂: Regional contributions (output European scale model)
- NO₂: Urban contribution (output urban model over some 130 cities)
- NO₂: Total urban background (regional contribution plus urban contribution)
- Benzene: Regional contributions (output European scale model)
- Benzene: Urban contributions (output urban model over some 130 cities)
- Benzene: Total urban background (regional contribution plus urban contribution)
- PM₁₀: Regional contributions (output European scale model)
- PM₁₀: Urban contributions (output urban model over some 130 cities)
- PM₁₀: Total urban background (regional contribution plus urban contribution)

The total urban background concentrations will thus, over rural areas, be identical to the sole regional contribution. These grids can be seen in Appendix D.

In Table 9 simulated maximum average concentrations in Sweden are shown. For PM₁₀ the rural background, reflecting sources outside Sweden, in southern and central Sweden are higher than the urban contribution, even in the larger cities. The contribution is from primary and secondary PM from sources in Europe, including minor contribution from sea spray. Only on a local scale are the contributions from residential wood burning and road dust larger than the rural contribution. A similar pattern is seen for benzene.

Table 9 Maximum annual average concentrations in Sweden as simulated by SIMAIR on the regional and urban scale for the year of 2010, together with observed maximum concentration in traffic environments (based on IVL: Datavårdskap för luftkvalitet, <http://www.ivl.se/tjanster/datavardskap/luftkvalitet.4.7df4c4e812d2da6a41680004804.htm>)

Maximum concentration		PM ₁₀	NO ₂	Benzene
(annual average, $\mu\text{g}/\text{m}^3$)	simulated rural background	13.5	11.9	2.54
	simulated urban contribution	11.0	33.3	1.40
	simulated total urban background	21.5	34.8	3.53
	observed total (incl. local)	26.2	48.9	3.6
		PM ₁₀	NO ₂	Benzene
Location for maximum concentration	simulated rural	Malmö	Malmö	Stockholm
	simulated urban contribution	Göteborg	Göteborg	Stockholm
	simulated total urban background	Göteborg	Göteborg	Stockholm
	observed total (incl. local)	Stockholm	Göteborg	Göteborg

8.3.2 Local scale and hot-spot concentrations

While comparing urban scale maps of NO₂, PM₁₀ and benzene with threshold values it can be concluded that on the urban background scale (1x1 km²) the limit values, see Table 4 and 8, are not exceeded. However, this may be the case closer to traffic or residential wood combustion where local scale concentrations at the address level can be considerably higher than at larger scales and thus represent hot-spot concentrations. In order allow assessments of hot-spot concentrations, the SIMAIR model system also couples the regional and urban scales to the local (street) scale. There are three local model applications in the SIMAIR system:

- SIMAIR road: local model is focusing traffic, either the street canyon model OSPM or a infinitive line source model for roads without surrounding buildings (Gidhagen et al., 2009),
- SIMAIR crossing: local model is focusing traffic with a finite line source model that simulates the integral impact of traffic on various road links and junctions (Omstedt et al., 2011),
- SIMAIR rwc: local mode focusing residential wood combustion with a point source model.

The full SIMAIR system with regional, urban and local contributions has been extensively used for assessments of Swedish pollution levels, e.g. by Omstedt et al. (2012), see Table 10. As can be seen the local contribution is highly significant.

Table 10 Modeled air concentrations in 48 Swedish street environments in 2008 (Omstedt et al., 2012) calculated from values in Appendix 1 in the report (available measurements at a few locations have been used to correct model output).

2008	Annual mean ($\mu\text{g}/\text{m}^3$)			Daily	Hourly
	Total	Urban background	Local (traffic) contribution	98-percentile ($\mu\text{g}/\text{m}^3$)	98-percentile ($\mu\text{g}/\text{m}^3$)
PM _{2.5}	9.61 ±1.94	7.88 ±1.76	1.74 ±0.75	-	-
PM ₁₀	21.7 ±5.36	12.7 ±2.92	9.03 ±3.86	39.8 ±12.9 (90-percentile)	-
NO ₂	27.9 ±8.00	13.2 ±5.71	14.7 ±5.08	53.3 ±14.2	71.9 ±19.3
Benzene	1.54 ±0.57	1.04 ±0.44	0.49 ±0.21	-	-

- Not calculated.

Receptor point concentrations can be generated to include local impact from local road traffic and residential wood burning. SMHI is currently doing an epi study considering BaP and PM₁₀ from residential wood combustion and traffic. This is, however, only done for two cities in Västerbotten, a county in Northern Sweden; Umeå and Vännäs, the latter being very small but dominated by residential wood combustion. The study comprises the local scale, and here SMHI can simulate specific source contributions to the air concentrations (such as traffic or wood combustion). It is possible to simulate NO₂ and benzene but they are not currently in focus of the project.

The SIMAIR results of 2004 has been used for a health study where PM exposure was related to self-reported health outcomes given by a National Health Survey from 2007 (Gidhagen et al., 2013). The study, titled ROADSIDE, separated the impact of combustion particles (PM₁) and coarser, mostly road-wear particles (PM₁₀-PM₁) at about 26,000 home addresses all over Sweden. It was found that long-range transport dominates average Swedish residential PM₁ and PM₁₀ levels, but for urban populations the contributions from urban and local traffic sources are important and may even dominate for residences close to heavily trafficked roads. Total PM₁₀ was ranging from 3.5 (northernmost) to 13.5 µg/m³ (southernmost). Local traffic and urban sources contributed nationally on average to 16 % of total PM₁₀, but for urban populations this contribution was larger (for Stockholm around 30 %). Generalized to the Swedish adult population the average residential exposure contributions from regional, urban, and local traffic PM₁₀ were 10.2, 1.3 and 0.2 µg/m³, respectively. Corresponding exposure to PM₁ was 5.1, 0.5 and 0.03 µg/m³ (for further details of ROADSIDE results, see Gidhagen et al. 2013).

The on-going Västerbotten study, funded by the Swedish EPA, focuses the impact of residential wood combustion in the northern parts of Sweden. A first simulation with the SIMAIR rwc applications in Lycksele shows the impact of residential heating in 642 houses. In SIMAIR rwc the local contribution comes both from traffic and wood combustion, however the traffic impact is insignificant in comparison with wood combustion (Figure 21 and, Table 11). Clearly the residential wood combustion is a very important source for PM pollution in cities in the northern Sweden.

Figure 21 Simulated annual averages of PM₁₀ at 642 home addresses, ordered on the x axis according to PM₁₀ level, (that all uses wood combustion for heating their houses). The regional contribution is around 6 µg/m³ (greyish blue), the urban contribution is yellow, the traffic contribution is not visible and the impact of local wood combustion is shown.

Table 11 Simulated annually averaged contributions to and concentrations of PM₁₀ in Lycksele. Units are µg/m³.

	Regional contribution	Urban contribution	Local contributions from wood combustion	Local contributions from road traffic	Total concentrations
Median	6.21	0.30	1.57	0.01	8.13
Max	6.23	0.46	5.64	0.74	12.20

8.3.3 Trends in Swedish exposure levels

SMHI has, commissioned by the Swedish EPA, assessed the expected changes in PM₁₀, NO₂ and benzene levels in 48 different traffic environments (Omstedt et al., 2012). A baseline was calculated for 2008 by the SIMAIR system (see Table 3 above). With relevant changes in traffic, ship traffic and other pollutions sources expected to 2020, the SIMAIR system was run for the same 48 environments.

For PM₁₀ the future levels were rather similar to the present ones, as the effect of some technical improvements with lower combustion emissions was opposed by an increased traffic volume. For PM₁₀ the future use of studded tyres is important and commonly suggested measures will not be able to considerably lower the PM₁₀ levels in Swedish cities. The situation is better for PM_{2.5} where almost all 48 traffic environments will have slightly lower levels in 2020.

For NO₂ there are generally lower values to be expected in 2020, although there are a few examples where levels will increase. The different trends depend to a large extent on the size of the heavy duty vehicle fraction (the latter will have significantly reduced emission in 2020). The increasing amount of diesel fuelled personal cars make the general NO₂ trend to 2020 more difficult to estimate. An increase in NO₂ levels to about 2015 is expected in some traffic environments, thereafter the NO₂ concentrations probably will decrease.

For benzene lower concentrations are reported at all 48 sites in 2020, as compared to 2008.

Another assessment of trends in future urban background levels has been reported by Gidhagen et al., 2012. There the 2010 concentrations of NO₂ in the Stockholm-Uppsala area were compared to projected concentrations in 2030, here also taking into account climate change effects. The simulations were made with two different climate scenaria, however the effect of climate change on future air quality NO₂ levels were rather small. Taking into account the expected reductions in air pollution emissions in Europe, a clear trend for decreasing NO₂ was determined for air coming into the Stockholm area. Combining this lower long-range contribution with reductions in NO_x emissions in the Stockholm-Uppsala area, a clear trend with lower future NO₂ values can be seen.

9 Epidemiological studies and future work

It is generally accepted that exposure to air pollution may be linked to severe adverse health effects in the population. Such health effects have been demonstrated in various epidemiological studies as well as in animal models and cell studies. The effects of air pollution on human health have been shown for short-term as well as from long-term exposure. Long-term exposure to air pollution has been linked to an increased risk of chronic respiratory illness (Folinsbee, 1993), cardiopulmonary mortality (Hoek et al., 2002), and of developing various types of cancer (Pope et al., 2002). Higher prevalence of bronchitis (Karakatsani et al., 2002), acute cardiovascular disease (Urch et al., 2005), asthma and other symptoms (Sunyer et al., 1997) has on the other hand been associated with short-term exposure during episodes of enhanced air pollution concentrations. In recent years there has been a significant focus on the health effects of particulate air pollution (Davidson et al., 2005), but also gaseous pollutants like e.g. ozone, nitrogen dioxide, sulphur dioxide and various hydrocarbons have significant negative health effects (Maynard, 2004). In addition, the presence of pollutant mixtures may have synergistic effects.

A central issue in environmental epidemiology is to relate chemical exposure to occurrence of health effects, or rather to relate changes in exposure to changes in health effect. In doing so one has to define the study boundaries in terms of sources, pollutants, health effects and spatial and tidal scales. In reality there is an abundance of pressures that influence the state of health of humans, ranging from life-style factors such as smoking, diet and exercise and residence to pressures from the surroundings, such as workplace and domestic conditions and chemical exposure from environment and exposure from consumer products. Also noise, UV and radon radiation and the state of stress are factors to be considered. Even genetic characteristics may need to be considered because they may affect selection of health habits or modify effect of external factors. Finally parental exposures may have an effect on health of offspring, and family structures may lead to clustering of exposures and ill-health. It is important to perform an adequate adjustment for potential confounding factors, such as smoking etc., in order to obtain a reliable correlation between sources and effects (e.g. Pukkala, 1991).

An epidemiological study requires backtracking of air exposures at address level over, e.g., a 30 year period, and applying statistical methods for investigating correlation patterns in terms of incidence rate ratios. On a less ambitious level health data with the most predominant cancer forms, distributed over age and year, on a national level or regional level, could also be used together with annual air concentrations. This pilot clarifying study presents a snapshot of the air exposure concentrations at a given time (2010), and to assess this exposure a comparison with threshold values that represent certain risk when exceeded, can be performed. This is not as accurate as a long terms study but gives indications of potential critical sites, sources and pollutants, which can be studied more extensively in a detailed epidemiological study. The maps and data presented in this report thus present an important integral part and can directly be used to evaluate cancer air quality threshold values.

9.1 New approach (residential wood combustion)

Lung cancer is one of the most frequent cancers and the typical primary exposure route of carcinogens to humans in urban environments is the ambient air quality (Danaei et al. 2005). Cancer development is complex and per definition multi-causal and often displays delayed and non-linear responses, which needs to be reflected in the risk appraisal (Hanahan & Weinberg, 2000). Hence, ascribing causal relationships is often very difficult. However, previous studies have shown significant correlation between chemical exposures via environment and cancers; e.g. air pollution from traffic in Denmark (Raaschou-Nielsen et al., 2010), Urban air pollution in Stockholm (Nyberg et al., 2000) and Lung cancer and ambient air pollution in Helsinki (Pönkä et al., 1993).

Most recently, in the ESCAPE project the incidence of lung cancer related to air pollution was investigated; the 312 944 cohort members contributed 4,013,131 person-years at risk. During follow-up (mean 12.8 years), 2095 incident lung cancer cases were diagnosed. The meta analyses showed a statistically significant association between risk for lung cancer and PM₁₀ (hazard ratio (HR) 1.22 [95 %-confidence interval 1.03-1.45] per 10 µg/m³). For PM_{2.5} the HR was 1.18 (0.96-1.46) per 5 µg/m³. The same increments of PM₁₀ and PM_{2.5} were associated with HRs for adenocarcinomas of the lung of 1.51 (1.10-2.08) and 1.55 (1.05-2.29), respectively. An increase in road traffic of 4000 vehicle-km per day within 100 m of the residence was associated with an HR for lung cancer of 1.09 (0.99-1.21). The results showed no association between lung cancer and nitrogen oxides concentration (HR 1.01 (0.95-1.07) per 20 µg/m³) or traffic intensity on the nearest street (HR 1.00 (0.97-1.04) per 5000 vehicles per day) (Raaschou-Nielsen et al., 2013).

Most health effects studies have focussed on impact of traffic air pollution, and it would thus be highly relevant to focus on health effects of air pollution from a variety of other sources:

- Smoke from domestic wood stoves,
- Allergenic pollen from trees, grasses and new invasive species,
- Airborne allergenic material (skin tissue, hair etc) released from livestock,
- Fungal spores from agriculture and other sectors,
- Airborne particles from natural sources (sea spray, soil dust)
- Long-range transported organic and inorganic particles incl. agricultural emissions
- Pesticides applied in farming

Wood combustion is one of the largest sources of particles emissions and constitutes a large, and still increasing, fraction of the emissions of (co)carcinogenic pollutants, e.g. NMVOC, NO_x, PAH and heavy metal, in the Nordic countries. Emissions are potentially critical because they are relatively close to the ground as opposed to point sources, such as industries and power plants, which typically have emissions from chimneys at heights leading to dilution with reduced impact on a local scale. Furthermore the emissions usually take place where the population reside and therefore contribute significantly to human exposure of air pollution.

Health effects from wood combustion have been studied in chamber experiments (Riddervold et al., 2011), but an estimate of the total air concentration and epidemiological studies of critical pollutants has not yet been made. Often proxies in the form of a single pollutant have been used, such as NO_x as

a proxy for air pollution from traffic (Raaschou-Nielsen et al., 2010). However, when a total risk, expressed as Margin of Exposure, is required, it is necessary to consider all, or at least the predominant, pollutants, as their cancer threshold levels may vary considerable. From this perspective an important aim of the present study is to present and assess the most complete exposure profile, based on the available emissions and air exposure concentrations in the Nordic countries. This will enable a more accurate and transparent estimate of critical pollutants and sources and give support to decision makers to initiate regulatory action on a source and pollutant scale.

To consider common methodologies for other Nordic countries for gridding of residential wood combustion, the key question is availability of spatial (GIS) data. An optimal situation would be to have a building register with house coordinates and information about wood heating devices and their use. However, such data are rarely available. If there is a building register with information about main building types that are relevant for the wood use in the country (e.g. residential/other, apartment/detached/semi-detached), and an estimation about urban/rural differences in wood use, a relatively good approximation for large area average can be achieved. In case of absence of building register, population data in spatial format is often available. However, using population data as a proxy leads to emission allocation to apartment houses and city centres that are not typically heated with wood. In this case, one idea could be to use population density information to rule out apartment houses. For Sweden, where a considerable share of residential wood combustion is by small-scale wood boilers, might benefit from gridding boiler emissions separately from stove emissions. Different heating needs in different parts of the country because of climate conditions might be relevant for at least for Norway and Sweden, and can be taken into account with degree-day values.

9.2 Denmark

Health effects of air pollution in the Danish population have been studied in a variety of projects over the past 20 years and a review of these is currently under preparation (Hertel et al., 2013, in preparation). In this context it should be noted that Danish air pollutant levels are generally moderate due to windy climate and moderate emissions (Hertel and Goodsite, 2009). Despite this, Danish epidemiological studies points at severe negative adverse health effects: stroke (Andersen et al., 2009; Andersen et al., 2010), lung cancer (Raaschou-Nielsen et al., 2010), COPD (Andersen et al., 2011), asthma in adults (Andersen et al., 2012a), wheeze in infants (Andersen et al., 2005; Andersen et al., 2008), asthma hospital admission in children (Iskandar et al., 2011), oxidative stress in blood DNA (Bräuner et al., 2007), vascular function in elderly (Brauner et al., 2008), air pollution enhances effects of radon on childhood leukaemia (Brauner et al., 2010), and most recently diabetes (Andersen et al., 2012b). These findings have been possible due to access to precise health data and advanced exposure assessment methods. Unique population and health registries in the Nordic countries allow detailed health impact assessments to be carried out. Measurements from the Danish air pollution monitoring programmes and AirGIS calculations on address level have been used as exposure proxies in a series of publications to evaluate various negative health outcomes. The later were based either on data from health registers or biomarker measurements.

There are different planned activities in Denmark regarding the exposure from residential wood burning and the influence on both chronic and acute

diseases. From the recent update of the EMEP emission inventory guidebook the emission factors have been improved, and the big challenge is obtaining a more accurate and reliable spatial distribution of wood burners. Planned monitoring campaigns of particle fractions and e.g. PAHs in rural, urban and local sites with high occurrence of wood burners, will supply input to source apportionment and estimates on the influence of wood burners on certain cancers and acute illnesses such as bronchitis and asthma and other symptoms.

9.3 Finland

Residential wood combustion, i.e. heating of residential houses using stoves, fireplaces or small-scale house boilers, takes place in majority (more than 80 %) of Finnish detached houses. Most of the houses use some other heating media as a primary heat source and wood combustion is only supplementing the primary heating. This means that wood combustion amounts per household varies widely. Based on questionnaires made by Finnish forest research institute METLA, the main factors are the primary heating media and the level of “urbanization”. The FRES model uses for gridding:

- Digital building and dwelling register that includes information about primary heating media and coordinates of all the Finnish residential houses.
- SYKE GIS system with categorization of all inhabited areas to metropolitan/urban/semi-urban/rural classes.
- Average wood use estimates categorized for different primary heating media and metropolitan/urban/semi-urban/rural classes based on METLA questionnaire and separate questionnaire by HSY for Helsinki metropolitan area.
- Degree-day weighting to take into account different heating needs at different parts of Finland.

9.4 Norway

An epidemiological study with residential wood burning in a Norwegian city, e.g., Oslo, for a longer period will generally require a recalculation of air quality with AirQUIS/EPISODE for the time period selected (months, years), since such data are not directly retrievable from earlier studies; or in the case of Oslo, since the existing 1992-2002 data only consists of total concentrations from all source groups. Even though wood burning emissions exists in the AirQUIS data base for recent years, it may be difficult to define such data accurately if the epidemiological study extends further back in time.

If cohort data (cancer incidents) connected with geographical location (e.g., home addresses) exist and are available in Oslo for the years 1992-2002, an epidemiological study can be performed relatively easy, using existing air quality data (total concentrations) in grids and receptor points. A more comprehensive backtrack tidal study with 20-30 years of exposure on address level in Oslo may be feasible, but will require a considerable amount of effort, especially to obtain good emission data. Doing this for other cities in Norway is perhaps feasible, but again problematic over long time spans mainly due to the difficulty of obtaining good emission data.

Defining an epidemiological study for Norway based on annual mean exposure for regions, or for the whole country, using aggregated regional cancer incidents will generally require the use of regional scale modelling results

for Norway for the whole study period, which exists in the form of EMEP model results for the period 1995-present, but will perhaps be too coarse and inaccurate, especially close to major cities and urban areas.

9.5 Sweden

As the effect of an exposure to particle matter will depend on its origin and composition, i.e. which source gives the dominating contribution there is thus a need for splitting up the total exposure into contributions from separate sources with special focus on local emissions from traffic and residential wood combustion.

For Swedish exposure there could be two ways for the next phase:

Analysing and discussing the results of the Västerbotten study, in which the SIMAIR rwc model will assess the separated impact of local traffic and local wood combustion. The model results will be evaluated against measurements of $PM_{2.5}$, soot and BaP in four locations. The model will then be used over the entire county of Västerbotten.

Perform a special analysis of the regional + urban concentrations (the urban background $1 \times 1 \text{ km}^2$) all over Sweden, where we distinguish the impact of within city traffic and residential wood combustion on $PM_{2.5}$, PM_{10} and benzene (if possible also BaP, this depending on the timing with other ongoing projects). The method to separate traffic and wood combustion impact in Swedish cities is through the emission ratios of the two sectors to total emissions (as described above under the section "Exposure grids for Sweden delivered in this project". The impact separation will only be possible in the 130 cities/villages.

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Appendix A Pollutant emissions in main sectors

Table A1 Emissions for different pollutants per sector for year 2010 for Denmark.

Sector	NM VOC (tonnes)	NO _x (tonnes)	SO ₂ (tonnes)	PM _{2.5} (tonnes)	PM ₁₀ (tonnes)	TSP (tonnes)	Pb (kg)	Cd (kg)	Hg (kg)	As (kg)	Cr (kg)	Cu (kg)	Ni (kg)	PCDD/F (mg I- Teq)	B(a)P (kg)	B(b)F (kg)	B(k)F (kg)	Indeno (kg)
Public Power	2,380	18,670	3,660	473	592	761	361	27	217	85	184	162	349	1,180	9	36	21	7
Industrial Combustion	364	13,800	3,630	239	311	396	396	42	77	100	92	134	1,720	50	17	63	14	4
Small Combustion	15,610	8,140	2,800	18,730	19,020	19,970	1,810	47	36	37	109	363	340	18,150	4,650	4,710	2,660	3,230
Industrial Processes	4,970	21	11	97	149	222	472	13	14	13	47	59	86	53	0	1	1	0
Fugitive emissions	9,850	173	1,270	33	280	691	1	3	1	1	4	2	6	0	0	0	0	0
Solvents	26,780	41	40	215	247	355	43	4	1	9	205	2,410	164	89	1	0	0	0
Road Rail	12,700	46,980	78	2,480	3,210	4,090	6,330	43	26	7	184	46,740	142	111	59	73	82	63
Shipping	1,380	20,250	1,800	469	471	474	41	5	15	44	24	45	1,820	178	2	9	4	14
Off-Road Mobile	9,600	19,390	92	1,570	1,570	1,570	117	7	5	0	22	18	7	42	8	16	15	9
Aviation	183	1,170	80	5	5	5	939	0	0	0	0	0	0	0	0	0	0	0
Other Waste Disp	174	38	558	6	6	6	130	0	0	0	1	4	0	6,010	56	68	55	83
Waste Incineration	3	20	4	4	4	5	0	0	47	0	0	0	0	54	0	0	0	0
Agriculture Livestock	0	0	0	1,190	5,650	11,350	0	0	0	0	0	0	0	0	0	0	0	0
Agriculture Other	1,930	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Agriculture Waste	232	89	11	203	214	214	32	2	0	2	8	0	7	22	103	101	40	38
Total	86,160	128,800	14,040	25,700	31,720	40,110	10,670	192	440	299	880	49,940	4,650	25,930	4,900	5,070	2,890	3,450
Total per inhabitant	15.6	23.3	2.54	4.64	5.73	7.25	1.93	0.035	0.080	0.054	0.159	9.02	0.840	4.69	0.885	0.916	0.522	0.623
	kg/person kg/person kg/person kg/person kg/person kg/person g/person g/person g/person g/person g/person g/person g/person g/person µg/person g/person g/person g/person g/person																	

Table A2 Emissions for different pollutants per sector for year 2010 for Finland.

Sector	NM VOC (tonnes)	NO _x (as NO ₂) (tonnes)	SO _x (as SO ₂) (tonnes)	PM _{2.5} (tonnes)	PM ₁₀ (tonnes)	TSP (tonnes)	As (kg)	Cd (kg)	Cr (kg)	Hg (kg)	Pb (kg)	PCDD/ PCDF (mg I-Teq)	PAH-4 (kg)
Energy production	3,330	84,840	56,600	2,870	5,600	9,510	3,110	663	7,930	412	14,560	5,370	575
Aviation	120	761	48	27	27	27	0	0	0	0	435	0	0
Road transport	22,150	43,420	72	6,610	10,750	18,450	0.23	4	75	21	665	2,710	1,059
Railways	122	2,230	0.64	37	43	44	0	0	0	0	0	0	0
Shipping	5,220	9,590	1,050	437	447	451	0	0	0	0	0	0	0
Other transport	46,740	25,100	6,600	23,320	25,220	29,430	219	347	2,630	39	3,640	1,480	14,760
Fugitive emissions	8,650	0	0	4,790	6,820	10,430	9.6	0	46	0	57	190	1,240
Industrial processes	8,640	205	2,310	1,920	4,020	7,330	349	400	11,200	382	3,900	4,730	8.8
Solvents	20,990	0.015	45	361	364	382	0.002	0	0	0.0012	0.00062	1.5	21
Agriculture	0	11	0	337	1,490	3,710	0.28	0.24	1.1	0.039	4.2	2.4	0.00052
Waste management	579	293	70	4.7	7.6	10	0.87	0	10	5.1	48	178	74
Total	116,540	166,450	66,790	40,700	54,770	79,770	3,680	1,410	21,890	854	23,310	14,660	17,730
Total per inhabitant	21.8 kg/person	31.1 kg/person	12.5 kg/person	7.60 kg/person	10.2 kg/person	14.9 kg/person	0.687 g/person	0.264 g/person	4.09 g/person	0.159 g/person	4.35 g/person	2.74 µg/person	3.31 g/person

Table A3 Emissions for different pollutants per sector for year 2010 for Norway.

Sector	NM VOC (tonnes)	NO _x (tonnes)	SO ₂ (tonnes)	PM _{2,5} (tonnes)	PM ₁₀ (tonnes)	TSP (tonnes)	As (kg)	Cd (kg)	Cr (kg)	Hg (kg)	Pb (kg)	PCDD/PCDF (mg)	PAH-4 (kg)	PAH (kg)
Oil and gas extraction – stationary combustion	2,420	50,640	443	1,450	1,710	1,770	35	12	119	21	33	1,660	16	755
Oil and gas extraction – process emissions	37,900	-	5	-	-	-	-	-	-	-	-	-	-	-
Metal industry – process emissions	1,190	10,750	7,780	2,490	3,020	3,070	755	85	196	60	1,190	2,570	4,600	32,130
Other manufacturing industries and mining	10,180	10,190	5,540	2,000	5,470	15,320	378	204	1,810	116	820	2,920	107	1,160
Energy supply	1,150	2,960	1,410	687	796	935	71	37	83	24	60	612	17	777
Heating in other industries, incl. waste incineration	674	1,180	403	615	655	701	29	13	106	18	128	1,410	234	2,340
Heating in households	10,240	1,890	487	33,060	33,070	33,100	243	148	231	25	94	8,590	2,170	38,390
Road traffic	12,360	37,270	44	1,220	1,280	1,280	165	33	165	108	250	330	1,090	9,890
Railways	50	584	0	47	47	47	1	0	1	1	1	1	1	41
Domestic aviation	1,200	4,130	112	5	5	5	19	4	19	11	1,270	26	2	129
Coastal navigation	2,380	51,370	3,220	2,310	2,430	2,430	88	17	128	65	167	4,420	44	1,770
Motorized equipment etc.	14,630	14,910	12	2,990	3,120	3,120	31	6	31	28	58	63	50	1,980
Agriculture - fertilizer and other	-	-	-	1	1	1	-	-	-	-	-	-	-	-
Road, tyre and brake wear and abrasion of railway contact wires	-	-	-	545	1,820	8,470	0	30	55	3	1,580	-	240	484
Products containing solvents etc.	41,820	13	-	153	154	154	1	0	1	27	0	5	1	8,430
Fires, cremations etc.	1	1	0	282	282	282	3	2	2	75	1	2,990	7	503
Petrol distribution	4,570	-	-	-	-	-	-	-	-	-	-	-	-	-
Other	0	-	-	-	1	1	-	0	0	1	4	7	0	65
Total	140,740	185,890	19,450	47,850	53,860	70,680	1,820	590	2,950	584	5,650	25,610	8,580	98,840
Total per inhabitant	28.6 kg/person	37.7 kg/person	3.95 kg/person	9.71 kg/person	10.9 kg/person	14.3 kg/person	0.369 g/person	0.120 g/person	0.597 g/person	0.119 g/person	1.15 g/person	5.19 µg/person	1.74 g/person	20.05 g/person

Table A4 Emissions for different pollutants per sector for year 2010 for Sweden.

Sector	NMVOC (tonnes)	NO _x (tonnes)	SO _x (tonnes)	PM _{2.5} (tonnes)	PM ₁₀ (tonnes)	As (kg)	Cd (kg)	Cr (kg)	Hg (kg)	Ni (kg)	Pb (kg)	PCDD/ PCDF (mg I-Teq)	PAH-4 (kg)
Energy production	8,850	34,970	15,900	7,980	9,850	390	300	1,170	260	13,020	4,830	33,340	3,570
Fugitive emissions	101,340	440	970	510	770	0	0	0	0	0	0	240	1,130
Households	12,430	5,140	790	6,900	6,900	30	180	180	30	160	910	4,160	11,530
Industrial processes	14,440	14,390	12,370	5,850	7,790	400	90	3,560	140	1,280	3,390	3,690	20
Traffic	35,420	67,490	80	6,650	11,070	0	0	0	0	0	3,910	590	100
Other transports	16,350	18,020	4,470	1,140	1,160	140	0	200	0	5,420	40	10	10
Machinery	7,750	20,720	10	1,410	1,490	0	0	0	0	0	0	0	0
Household machinery	660	1,500	0	70	70	0	0	0	0	0	0	0	0
Agriculture	0	0	0	800	4,450	0	0	0	0	0	0	0	0
Waste management	0	90	0	220	220	0	0	0	120	10	0	870	30
International aviation (cruise)	330	9,130	630	30	30	0	0	0	0	0	0	0	0
International shipping on swedish waters	5,600	245,060	158,950	14,380	15,070	0	0	0	0	0	0	0	0
Total	203,170	416,950	194,170	45,940	58,870	960	570	5,110	550	19,890	13,080	42,900	16,390
Total per inhabitant	21.7 kg/person	44.5 kg/person	20.7 kg/person	4.91 kg/person	6.29 kg/person	0.103 g/person	0.0609 g/person	0.546 g/person	0.0588 g/person	2.12 g/person	1.40 g/person	4.58 µg/person	1.75 g/person

Appendix B Emission and exposure grids in Nordic countries

Table B1 Summary of atmospheric emissions and exposure modelling of (co-)carcinogens performed in Denmark, Finland, Norway and Sweden. Spatial scales of grids are stated. Compared to complete pollutant list in Table 1, only pollutants where emission and/or exposure data are available, are included. Model grids cover entire respective country unless otherwise stated.

	Denmark		Finland		Norway		Sweden	
Pollutant	Emissions (kmxkm)	Exposure modelling (kmxkm)	Emissions (kmxkm)	Exposure modelling (kmxkm)	Emissions (kmxkm)	Exposure modelling (kmxkm)	Emissions (kmxkm)	Exposure modelling (kmxkm)
Chlorine and its inorganic compounds		5.6x5.6 ¹⁾						
<i>TSP</i>	1x1	5.6x5.6	1x1				1x1	
<i>PM_{2.5}</i>	1x1	5.6x5.6	1x1	~20x20	1x1	1x1	1x1	1x1 ⁴⁾
<i>PM₁₀</i>	1x1	5.6x5.6	1x1	~20x20	1x1	1x1	1x1	44x44 and 1x1 ⁴⁾ 1x1 ⁵⁾ and 1x1 ⁶⁾
<i>Arsenic</i>	1x1	5.6x5.6 ²⁾					1x1	
<i>Cadmium</i>	1x1	5.6x5.6 ²⁾					1x1	
<i>Chromium</i>	1x1	5.6x5.6 ²⁾					1x1	
<i>Lead</i>	1x1	150x150 ²⁾					1x1	
<i>Mercury</i>	1x1	150x150 ³⁾					1x1	
<i>Nickel</i>	1x1	5.6x5.6 ²⁾					1x1	
<i>NM VOC (not specified)</i>	1x1	5.6x5.6	1x1				1x1	
Acetaldehyde		5.6x5.6						
Benzene		5.6x5.6			1x1	1x1		44x44 and 1x1 ⁴⁾ 1x1 ⁵⁾
Ethylbenzene		5.6x5.6						
Formaldehyde		5.6x5.6						
<i>Hexachlorobenzene</i>	1x1							
Hexachlorocyclohexane (Lindane)		150x150						
<i>PCDD/F (dioxins not specified)</i>	1x1							
PCB		150x150						
<i>SO_x (SO₂ for PRTR data)</i>	1x1	5.6x5.6	1x1		50x50	50x50	1x1	
<i>NO_x (NO₂ for PRTR data)</i>	1x1	5.6x5.6	1x1	~20x20	1x1	1x1	1x1	44x44 and 1x1 ⁴⁾ 1x1 ⁵⁾
<i>PAHs (not specified)</i>		5.6x5.6 ²⁾					1x1	
<i>Benzo(a)pyrene</i>	1x1							44x44 and 1x1 ⁷⁾
<i>Benzo(b)fluoranthene</i>	1x1							
<i>Indeno(1,2,3-cd)pyrene</i>	1x1							

¹⁾ Only Cl, as a part of sea salt

²⁾ As a part of PM_{2.5}. Totals taken from the national emission reporting and distributed by a proxy, (not taken in 1x1 km resolution from emission inventory) however the re-emissions from soil-dust is not modelled explicitly, could be very important today since the emissions from chimneys are reduced significantly

³⁾ RGM=Reactive Gaseous Mercury= the sum of HgCl₂ and HgO; TPM=Total Particulate Mercury; See: Christensen et al. (2004)

⁴⁾ Urban and local scale 48 street level environments

⁵⁾ Regional scale and 130 urban scale agglomerations

⁶⁾ Urban and local scale two cities Västerbotten

⁷⁾ Regional, urban and local scale two cities Västerbotten

Appendix C Nordic emission maps

All sources

The following figures show annual mean emissions to air in 2010 from all sources (national total). Pollutant and units are stated in legend.

Residential wood combustion

The following figures show annual mean emissions to air in 2010 from residential wood combustion. Pollutant and units are stated in legend.

Traffic

The following figures show annual mean emissions to air in 2010 from traffic. Pollutant and units are stated in legend.

Appendix D Nordic air exposure maps

All sources

The following figures show modeled annual mean air concentrations (exposures) from all main sources (national total). Pollutant and units are stated in legend.

Appendix E Example: Human exposure from ships in Finland

Finland: Contribution of the ships to air pollution and population exposure

In general the exposure modelling methodology is always adjusted to meet the requirements of the health effects studies, and depending on the final application, also the methods for estimating the exposure vary. If a very detailed resolution and detailed information on population activity is needed, FMI-EXPAND model, combined with urban/local scale dispersion modelling developed and evaluated in Helsinki Metropolitan area is used. If exposure modelling is needed for a wide region/ country, a different approach is used. Much coarser spatial resolution and normally just static information on population distribution is required for these types of studies. The SILAM-FMI regional scale model combined with static population data has proved to be a very useful tool in many long-term regional or country-wide health effect studies.

As an example for Finland, a more detailed description of methods used for estimating and linking the exposure and health effect from shipping, is shown below. This illustrates the complete chain of modelling approaches starting from emission and exposure modelling to health effect estimations. In this example the contribution of shipping to air quality and exposure is shown to illustrate the process of concentration calculations and exposure analyses until the final health effect studies.

Emission scenarios

The assessment considered four cases: (i) the baseline case representing the ship traffic and in-land emission of 2007, (ii) the estimated ship traffic emission following the Tier-II scenario, (iii) the estimated ship traffic emission following the Tier III scenario, and (iv) the zero-ship-emission case. In all scenarios the in-land emission rates were taken constant at the present level and EMEP emissions inventories were used for the description of ship traffic. The EMEP ship emissions were scaled according to Tier II and III requirements allowing for 2 % annual growth in ship emissions. The emission for the Tier scenarios were estimated for the year 2045 when the ship traffic is assumed to double while the emission per traffic-unit would decrease by 20% in Tier-II and by 80% by Tier-III. As a result, the total emissions would increase by a factor of 1.6 for Tier II and decrease by 60% for Tier III, in comparison with 2007 ship emission. Finally, a separate case was considered for the ship emission estimated on the basis of new methodology of Jalkanen et al., (2009), which provided the indication of the emission uncertainty and its impact on the results of the assessment. It also enabled a quantitative estimation of the impact of emission reduction measures in major harbours, the part of emissions which is missing from the official EMEP Baltic Sea inventory.

Dispersion modelling

The dispersion modelling tool used in this study is the System for Integrated ModelIng of Atmospheric coMposition, SILAM (<http://silam.fmi.fi>) developed by the Finnish Meteorological Institute (FMI). For the current assessment, the chemical transformation scheme of the DMAT model (Sofiev, 2000) was refined. The scheme considers both gas-phase and heterogeneous SO_x-NO_x-NH_x transformations, thus covering the whole set of conversions important for the study. Meteorological information and land cover maps were taken from the High Resolution Limited Area Model (HIRLAM, Uden (2002)) and the European Centre on Medium-Range Weather Forecast (ECMWF, www.ecmwf.int) meteorological models. The emission serving as input for SILAM, including NO_x, SO_x, CO, and NH₃, was derived from a European gridded emission inventory by TNO Built Environment and Geosciences (van der Gon et al., 2009).

The dispersion results were obtained from the model runs for the whole year of 2007, repeated for each emission scenario. The year was selected almost arbitrarily, with the only requirement to be free from extreme meteorological events, such as heat waves (2003, 2006, and 2010) or exceptionally cold winters (e.g. 2009).

Health effect evaluation

To evaluate the short- or long-term health effects it is important to choose averaging periods accordingly with the effects to be evaluated. The current WHO guideline values for NO₂ are 200 µgN m⁻³ for 1-hour level and 40 µgN m⁻³, an annual average. This guideline has been established as such in the European Legislation on ambient air quality, Directive 2008/50/EC

(<http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:32008L0050:en:NOT>). According to the latter Directive, 1 hour-level should not be exceeded more than 18 times during a calendar year. This aims to avoid, prevent or reduce harmful effects on human health and/or the environment. For the short-term health effect evaluation, this study will analyze the exceedances of this limit by computing the 99th percentile of concentration and depositions. Annual averages will be used to assess the long-term health effect.

To estimate the exposure of the population to certain pollutants, the exposure is modelled by combining concentration and population in a certain period of time. The following equation shows how the exposure (E) is computed for this study:

$$E = n * t * C [\mu\text{g}\cdot\text{s}/\text{m}^3]$$

Where n is the number of people per grid cell, t is the time period [s] and C is concentration [$\mu\text{g}/\text{m}^3$] for the pollutant. The number of people per grid cell (n) is retrieved from Gridded Population of the World Database (<http://sedac.ciesin.columbia.edu/gpw>), the SILAM computations provide the concentrations in a 20x20 km² grid and time period is 1 hour. The final results will be shown in terms of annual exposure for the above-specified emission scenarios.

Impact of TIER scenarios on human exposure

The impact on human health is evaluated in terms of short- and long-term effects, i.e. we considered the impact of both episodic high-concentration events and mean NO_x levels. According to the current European Legislation, within a year, the limit of 200 $\mu\text{gN m}^{-3}$ for 1-hour can be exceeded not more than 18 times (~99.8th percentile). To increase the statistical significance of the computations, we evaluated the 99th percentile (Figure E1). From Figure E1 one can see that the main sources contributing to the highest concentrations are situated inland, where traffic, in particular, is the most important source. Over these areas the probability to exceed the above threshold is not negligible but cannot be accurately estimated within the scope of the present study due to its strongly local nature and due to neglecting harbour emission estimates.

99% pctl_cnc_NOx, $\mu\text{gN}/\text{m}^3$

Figure E1. Annual 99th percentile for NO_x concentrations for the different emission scenarios: 2007 emissions (left-hand panel), Tier II (without NECA, middle) and Tier III (with NECA, right).

Nevertheless, it is visible that enforcing the Tier III scenario, the Baltic Sea and the coastal areas will benefit from such reduction, in particular the western side of the Baltic Sea and Helsinki area (Figure 11). The outcome is to be considered as the lower estimate of the Tier-III benefits: the reduction of the harbour emissions will bring about further improvements in the air quality in the main Baltic Sea cities.

Figure E2 Difference between the 99th percentile for NO_x concentrations for emission scenarios Tier II (NECA) and Tier III (without NECA), Unit: $\mu\text{g.h/m}^3$.

Long-term effect can be evaluated using the annual limit value set by the European Legislation: $40 \mu\text{gN m}^{-3}$ (annual average). As seen from the Figure 10, this is not a serious concern at present. The exposure to NO_x has been calculated taking into account the population distribution in the BSA and the annual average concentrations computed by SILAM. The overall impact on annual human exposure is shown in Figure 12 and it is computed as a product of average concentration, human population and time.

Figure E3 Annual average exposure to NO_x in the Baltic Sea area for the different emission scenarios: 2007 emissions, Tier II (without NECA) and Tier III (with NECA). Unit: $\mu\text{g.h/m}^3$.

On average, the human exposure to NO_x is reduced by a factor of ~ 1.5 when Tier III scenario is enforced instead of Tier II. The geographical distribution of exposure changes between Tier II and Tier III scenarios is seen from Figure E4. Application of Tier II only will lead to an increase of human exposure whereas Tier III reductions will decrease the exposure, especially in the coastal areas of the Baltic Sea.

Figure E4 Difference between human health exposure for Tier II (without NECA) and TIER III (with NECA). Exposure difference is given in %.