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Nordic Council of Ministers

Ved Stranden 18
DK-1061 Copenhagen K
Phone (+45) 3396 0200

www.norden.org

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Foreword

This report “Risk of Air Pollution in Relation to Cancer in the Nordic Countries” together with the report “Emissions and air exposure of carcinogens and co-carcinogens in four Nordic countries” constitutes the results from the projects KoL 14–07 and KoL 12–08 that were finalized in 2016 and 2013, respectively, for the Nordic Climate and Air Pollution Group (Klima- og Luftgruppen, KoL), Nordic Council of Ministers.

Seventeen carcinogenic and co-carcinogenic pollutants (particles, heavy metals, inorganic gases and organic compounds) are for the first time analyzed with respect to their emissions and air concentrations in a screening of the carcinogenic risk at very high resolution and large scale in ambient air in the Nordic countries.

The pollutants are included in the national emission inventories, which act as the primary data source, holding information on emission quantities from societal activities such as energy production, industrial activities, road transport, navigation, agriculture, waste management, residential heating, solvent and product use.

The risk towards humans was calculated for each pollutant by employing threshold values for air quality and additional lifetime cancer risk of 1:100,000. The term threshold value is in this report defined as a representation of the EU air quality limit and target value, WHO Air Quality Guidelines and US-EPA 1:100,000 risk concentrations. The risk is expressed relatively to area and population that are exposed to air concentrations exceeding threshold values.

In the KoL 12–08 report pollutant emission levels for 2010 and trends for 1990 to 2010 were compiled and discussed. In this KoL 14–07 report the emissions were used to model the annual mean atmospheric concentrations for 2010 for each pollutant as well as the total concentrations, which represent the exposure to humans in the Nordic countries. Measured concentrations in air for rural and urban sites were compiled and used for evaluation of the modelled concentrations and to estimate the contribution from national sources relative to background long range transport.

The work was carried out by atmospheric emission and air concentration modellers from Denmark, Finland, Norway and Sweden:

- Lars Gidhagen and Gunnar Omstedt, Swedish Meteorological and Hydrological Institute (SMHI).
- Tina Skårman, Swedish Environmental Research Institute (IVL).
- Alena Bartonova, Norwegian Institute for Air Research (NILU).
- Per Schwarze, Norwegian Institute of Public Health (FHI).
- Niko Karvosenoja, Ville-Veikko Paunu, Finnish Environment Institute (SYKE).
- Jaakko Kukkonen and Ari Karppinen, Finnish Meteorological Institute (FMI).
- Patrik Fauser, Matthias Ketzel, Marlene Plejdrup, Thomas Becker and Jørgen Brandt, Department of Environmental Science, Aarhus University (AU/ENVS).

Summary

Exposure to air pollution is linked to severe adverse health effects in the population. 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 have significant negative health effects (Maynard, 2004). The presence of pollutant mixtures may have synergistic effects and therefore it is important to consider all, or at least the predominant, pollutants.

From this perspective an important aim of the present study is to quantify and present the most complete mapping of air concentrations of carcinogenic pollutants, based on the available emissions in the Nordic countries, and evaluate these with respect to pollutant EU air quality values and carcinogenic 1:100,000 threshold values.

Methods

Danish, Finnish, Norwegian and Swedish 2010 emission data from all national sources and the regional contribution are included in an analysis of the carcinogens: arsenic (As), cadmium (Cd), chromium(VI) (Cr(VI)), lead (Pb), mercury (Hg), nickel (Ni), dioxins (PCDD/F) and poly aromatic hydrocarbons (PAHs): benzo(a)pyrene (B(a)P), benzo(b)fluoranthene (B(b)F), benzo(k)fluoranthene (B(k)F) and indeno(cd)pyrene (I(cd)P), and co-carcinogens: Sulphur dioxide (SO₂), nitrogen oxides (NO_x), fine particles (PM_{2.5}) and coarse particles (PM₁₀). The data sources are the national emission reporting to United Nations Economic Commission for Europe (UNECE), Convention on Long Range Transboundary Air Pollution (CLRTAP), and various scientific studies.

The requirements for including pollutants and sources in this study are that emissions must be available on a 1x1 km² grid covering the entire country. This yields 14 pollutants (11 carcinogens, 3 co-carcinogens) for Denmark, 4 pollutants (co-carcinogens) for Finland, and 11 pollutants (7 carcinogens, 4 co-carcinogens) for Sweden. Norway has no gridded emission data ready for this type of analysis. To reduce the modelling time only the areas with more than 20 persons living per square km are included for Sweden and Finland. For Sweden this implies

that 93.9% of the population of 9,528,000 persons corresponding to 27.5% of the populated area is included. For Finland 90.5% of the total population of 5,334,000 that lives on 19.8% of the populated area is included.

Gridded (1x1 km² resolution) annual mean air concentrations of PM_{2.5}, PM₁₀, SO₂ and NO_x are derived from modelling of national sources and long-range transport (background). Air concentrations of As, Cd, Cr(VI), Hg, Ni, Pb, PCDD/F, B(a)P, B(b)F, B(k)F and I(cd)P are derived from modelling of national sources and accounting for long-range transport from outside the country by adding a background concentration, which is the difference between a measured concentration at a rural site and a modelled concentration from national sources at the same site. The background concentration is applied to the entire country.

Considering annual mean air concentrations for a single year will give a screening of the air concentrations at a given time, which indicates critical sites in the 1x1 km² grid, sources (e.g. residential wood combustion) and pollutants (e.g. B(a)P).

The risk of exceedances of EU air quality values is expressed through the Margin of Exposure (MoE), and additionally for carcinogens the increased risk of developing cancer due to a lifetime exposure to the pollutant is calculated. The latter is expressed as a 0.00001 in 1 chance (or 1: 100,000 meaning one chance in a hundred thousand) of developing cancer.

Furthermore the populated area and number of persons exposed to risk concentrations are calculated and the contributions to the air concentration from national sources and regional long-range transport (background) are quantified. All results can be found in the respective country chapters and are summarized in Table 4 (Denmark), Table 6 (Finland) and Table 9 (Sweden).

Results

In summary, the calculated annual mean air concentrations of carcinogens and co-carcinogens comply with their respective EU air quality values and 1:100,000 cancer risk values in all countries. The only exception is B(a)P in Denmark, which has an annual mean concentration that exceeds the 1:100,000 cancer risk value of 0.12 ng/m³ (US-EPA IRIS, 2015) for approximately 80% of the Danish population living in 60% of the populated area. Note that the annual mean EU target value

threshold for B(a)P of 1 ng/m³ is not exceeded in any parts of Denmark. Residential wood combustion comprises the most influential source accounting for approximately 70% of the B(a)P concentration in ambient Danish air. Regional sources (long-range transport) comprise the second most influential sources for B(a)P.

The contribution from residential wood combustion to the ambient Swedish air varies between 0.1% for Ni and 7% for Cd. In this project there is no observed cancer risk explicitly from residential wood combustion in Sweden or Finland, a probable reason being that the PAHs, which are the most critical pollutants for this source, are not included in the Swedish or Finnish data.

Long-range transport is significant to the ambient air in all countries for all pollutants except the PAHs, which are mainly associated with residential wood combustion. NO_x, SO₂, Cd, Cr and Pb have significant contributions from national sources, especially in urban air, due to primarily road traffic, residential wood combustion and energy production and industrial point sources.

Uncertainties

There are various uncertainties associated with the calculated risk values. The largest uncertainties are related to the emission inventories, especially regarding the spatial distribution and emission factors of residential wood combustion. This implies that pollutant concentrations in urban areas in Denmark are overestimated in the calculations due to imprecise allocation of residential wood burning, see chapter 3.1.2. An improved allocation of residential wood burners in Denmark is currently being implemented, but is not yet available for the 2010 emissions used in this project. It is not known how the improved inventory will change the calculated exposure of the Danish population. But probably the values in the cities will be reduced and in rural areas the concentrations will increase.

The concentrations represent annual means for one year where in reality pollutants associated with e.g. residential wood combustion show a seasonal pattern. The population is therefore not exposed to the same concentration continuously, but rather higher concentrations in winter and lower in summer.

The data for the carcinogens (metals, PAHs and dioxins), concerning the background concentrations from long-range transport from outside the country, are very limited. In this project the background

concentrations are applied uniformly in the country. However, metals, except for gaseous mercury, are typically associated with coarse and/or fine particles (Malandrino *et al.*, 2016; Allen *et al.*, 2001; WHO, 2007) for which there are characteristic concentration gradients across the countries. This may overestimate the background concentrations in some parts and underestimate it in others.

The results presented here show the air concentrations on a 1x1 km² scale, which is typical for background pollution exposure estimates. However modelling on a 100x100 m² scale would show concentration peaks on a local scale, e.g. from traffic in street canyons and from residential wood combustion, that may not be discernible on a 1x1 km² scale and may cause further exceedances of threshold values.

The used air pollution model considers only O₃-NO_x chemistry while other substances are treated as inert pollutants, which may give additional uncertainties for reactive or volatile pollutants especially VOCs and PAHs.

Furthermore only primary particles are included in the urban scale dispersion modelling; these are assumed to be the most relevant in relation to cancer risk.

The applied models have been validated for urban background and rural locations for the pollutants NO_x, PM₁₀ and PM_{2.5} and show bias in the annual mean concentrations of typically 10–35%. Due to a lack of appropriate measurements the uncertainties for the other pollutants could not been quantified in detail in this study and therefore it is not possible to give probability ranges of pollutant concentrations or risk estimates.

1. Project aims and outlook

The projects seek to qualify the scientific and political answers to the questions:

- What is the exposure of carcinogens and co-carcinogens from inhalation of ambient air in the Nordic countries?
- What are the exceedances of the EU air quality values with respect to population number and location?
- What is the risk of atmospheric exposure with respect to cancer for the population?
- Which pollutants are the most critical with respect to exceeding their respective cancer risk values?
- Are the environmental cancer risks related to specific activities (e.g. residential wood burning) or locations (hot spots)?
- What are the most important needs in terms of emission and air concentration data to give a more accurate and complete understanding of the cancer risk from exposure of atmospheric pollutants?

To answer these questions the following results for Denmark, Finland, Norway and Sweden, are presented in the two reports:

- A comprehensive list of carcinogenic and co-carcinogenic pollutants (particles, heavy metals, inorganic gases and organic compounds) that are potentially emitted to the atmosphere from energy production, industrial activities, road transport, navigation, agriculture, waste, residential heating and product use.
- List of cancer risk values for pollutants with available emission data in the Nordic emission inventories.
- Description of emission inventory methods and air concentration models.
- Presentation and discussion of state (2010) and trends (1990 to 2010) of emissions, for sources and (co)carcinogens in main sectors, as reported to UNECE-CLRTAP.

- Tables with annual mean 2010 emissions of all (co)carcinogens in main sectors and total pr. inhabitant.
- Tables with measured atmospheric concentration data with focus on 2010 and 2011 on rural, urban and street environment.
- Tables with modelled annual mean 2010 atmospheric concentration data on rural and urban scales.
- Tables with calculated cancer risk data (Margin of Exposure, MoE) for all sources and pollutants.
- Prioritized list of sources and pollutants with highest potential contribution to atmospheric cancer risk on local and national scale.
- 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.
- Nordic maps of 2010 air concentrations covering the populated area of the countries on 1x1 km² scale for PM_{2.5}, PM₁₀, NO_x, SO₂, PAHs, dioxin, arsenic, cadmium, chromium, lead, mercury and nickel for all aggregated main sectors.
- Tables with modelled and/or measured annual mean 2010 air concentrations. Selected data are shown for the heavy metals, PAHs (BaP), benzene, PM_{2.5}, PM₁₀, NO_x (NO₂) and dioxin.
- Overview of local studies on air concentrations for cities or communities with emphasis on wood combustion and traffic.
- Discussion of existing epidemiological studies on cancer and environment. Application of available data in environmental health studies.
- Future application of available data in environmental health studies.

The results from this study can serve as input to (epidemiological) studies where correlation between environmental concentrations 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 air concentration levels at grid scale (1x1 km²), which is the most differentiated available spatial scale. From maps it is possible to identify localities with critically high air concentrations, and identify sources and pollutants that have high contribution to the air pollution.

The scope of a future project is to investigate and quantify the influence and effect of exposure of carcinogenic pollutants via the environment on the occurrence of childhood cancer as compiled and described in The Danish National Birth Cohort (DNBC). An ongoing project between Department of Environmental Science (ENVS) and the US National Institute of Health (NIH) "Estimating Agricultural Pesticide Near Residences in the Danish National Birth Cohort" considers the influence of pesticide use on childhood cancer. DNBC is a large prospective cohort study that followed over 100,000 children and their parents. The overall aim is thus to integrate the results generated within this Nordic project covering 17 (co-)carcinogens with the pesticide studies and furthermore to include the Norwegian children cohort to possibly reveal effects of differences in pesticide use and chemical emission practices on the occurrence of childhood cancer between the two countries.

In August 2015, a new project, NordicWelfAir, funded by NordForsk was initiated. The project will develop the air pollution modelling further by calculating air pollution levels at the 1x1 km² resolution over a period of 25 years (1990–2014) for the Nordic countries, Denmark, Sweden, Finland, Norway and Iceland. Linking the detailed information of the spatio-temporal distribution of air pollution levels and the chemical composition of the atmospheric particles with register data for mortality and morbidity, we have a unique opportunity in the Nordic countries to gain new understanding of the various health impacts from different kinds of air pollution from different kind of sources. This will provide the basic understanding needed for policy making of strategies to optimally reduce the air pollution challenge and to assess the related impacts on the distribution of health impacts and related societal costs and welfare, based on the integrated health impact assessment system EVA (Brandt *et al.*, 2013 a, b).

The results from the project can be used in both a Nordic as well as global perspective to improve the health and welfare by finding the optimal solutions to societal and public health challenges from air pollution through high quality research. The results from the proposed research have the potential to act as new international standards in our understanding of health impacts from air pollution for different population groups due to the possibility to integrate the unique data and knowledge of air pollution, register, health, socio-economics, and welfare research in the Nordic countries in a highly interdisciplinary project. The study will provide a Nordic contribution to international research on the topics of environmental equality and justice within the area of air quality related risks, amenities and wellbeing.

2. Introduction:

Air concentrations and cancer risk

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. Previous studies on the influence of chemicals via the environment have 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 exposure and risk of several inorganic and organic pollutants, inorganic gases and heavy metals based on existing data. This will give more realistic estimates, 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.

This report is a continuation of the work and results of project KoL 12–08. The following topics are assessed and presented in the report “Emissions and air exposure of carcinogens and co-carcinogens in four Nordic countries” and will not be repeated here unless there are important corrections:

- Cancer incidences showing that 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 28 EU member states.
- List of carcinogenic and co-carcinogenic pollutants and their sources that is relevant to exposure via the environment with focus on the atmosphere.

- Emission reporting obligations by EU member state countries for Persistent Organic Pollutants (POP) and heavy metal emissions under the Convention on Long-Range Transboundary Air Pollution (CLRTAP).
- Methodologies and structure of the national emission inventories with emphasis on residential wood burning.
- Mean 2010 emissions from main source categories for the carcinogens and co-carcinogens; Sulphur dioxide (SO₂), Nitrogen oxide (NO_x), Non-methane volatile organic compounds (NMVOC), Total Suspended Particulate matter (TSP) and Particulate Matter (PM), Cadmium (Cd), Mercury (Hg), Lead (Pb), Polycyclic Aromatic Hydrocarbons (PAHs), Dioxins and furans (PCDD/ PCDF).
- State and trend of emissions to air of above pollutants.
- Measured air concentrations of some pollutants.
- Air concentration modelling methodologies in the Nordic countries.
- Epidemiological principles and previous work done in this field by the Nordic countries.

2.1 Air concentration modelling

The pollutants with available emission data, covering the respective countries on a 1x1 km² resolution grid and where atmospheric concentrations are calculated within this project, are stated in Table 1. Model grid cells cover populated areas of the respective countries. This means that all 1x1 km² grid cells with any residents are included in the analysis.

Table 1: Gridded emission data available and air concentrations modelled in this project for carcinogens (italic) and co-carcinogens

Pollutant	Denmark		Finland		Norway		Sweden	
	Emissions (km ²)	Exposure modelling (km ²)	Emissions (km ²)	Exposure modelling (km ²)	Emissions (km ²)	Exposure modelling (km ²)	Emissions (km ²)	Exposure modelling (km ²)
TSP (total particles)	1x1		1x1				1x1	
PM _{2.5}	1x1	1x1	1x1	1x1	1x1		1x1	1x1
PM ₁₀	1x1	1x1	1x1	1x1	1x1		1x1	1x1
<i>Arsenic</i>	1x1	1x1					1x1	1x1
<i>Cadmium</i>	1x1	1x1					1x1	1x1
<i>Chromium</i>	1x1	1x1					1x1	1x1
<i>Lead</i>	1x1	1x1					1x1	1x1
<i>Mercury</i>	1x1	1x1					1x1	1x1
<i>Nickel</i>	1x1	1x1					1x1	1x1
NMVOC (unspecified)	1x1		1x1				1x1	
<i>Benzene</i>					1x1			
<i>PCDD/F (dioxin unspecified)</i>	1x1	1x1						1x1
SO ₂ (SO _x)	1x1		1x1	1x1	50x50		1x1	1x1
NO _x (NO ₂)	1x1	1x1	1x1	1x1	1x1		1x1	1x1
PAHs (unspecified)							1x1	
<i>Benzo(a) pyrene</i>	1x1	1x1						
<i>Benzo(b) fluoranthene</i>	1x1	1x1						
<i>Benzo(k) fluoranthene</i>	1x1	1x1						
<i>Indeno (1,2,3-cd)pyrene</i>	1x1	1x1						

Air concentration calculations are done for area sources and point sources as a national total (sum) for all categories, due to the extensive time being used for model calculations. It can be seen that the analysis comprises 14 pollutants (11 carcinogens, three co-carcinogens) for Denmark, four pollutants (co-carcinogens) for Finland, Norway has no emission data ready for this type of analysis, and 11 pollutants (seven carcinogens, four co-carcinogens) for Sweden.

Regional background modelling is done with the Danish Eulerian Hemispheric Model (DEHM) on a 5.6x5.6 km² resolution grid and is based on emissions on the Northern Hemisphere of all major sources and at present covers the pollutants SO₂, NO_x, NMVOC, TSP, PM_{2.5} and PM₁₀ (Christensen 1997; Brandt *et al.*, 2012 and <http://envs.au.dk/videnudveksling/luft/model/dehm/>). The contribution from national sources to the air concentration is calculated with the Urban Background Model (UBM) on a 1x1 km² scale (Brandt *et al.*, 2001a, b, c, 2003). The UBM model also calculates concentrations from point sources, such as industrial facilities, on a 1x1 km² resolution, and includes the dispersion up to 30 km from the point source.

The exposure for a person thus consists of three parts, the regional/continental scale background or long-range transport, the urban background from national emissions, and variations caused by

local conditions such as street canyons. Street canyons are, however, not included in this study.

For the heavy metals, PAHs and dioxin where no background concentrations from long-range transport are available, typically due to missing reliable emission estimates from other countries, the background concentrations can be found by comparing the modelled concentrations from national emissions with measured concentrations, at rural sites:

$$\text{Background concentration} = (\text{measured} - \text{modelled from national sources})_{\text{rural}}$$

Equation 1.

When measurements and modelled concentrations are available for several rural sites an average is used. The background concentration is assumed to be constant and distributed evenly on the whole country. Metals, except for gaseous mercury, are typically associated with coarse and/or fine particles (Malandrino *et al.*, 2016; Allen *et al.*, 2001; WHO, 2007). For fine particles there is a decreasing southeastern – northwestern concentration gradient, and a constant distribution of metal background concentrations may therefore overestimate the background concentrations in some parts and underestimate it in others. In this project the total ambient air concentration at any given point is found by adding the modelled concentration from national emissions with the constant background concentration.

The urban scale modelling of national sources are in this project performed assuming inert pollutants, i.e. degradation, volatilization, transformation etc., are not taken into account, except for NO_x/O₃ chemistry. This will in most cases lead to an overestimation, or conservative estimate, of the calculated air concentrations.

The model results are validated against measurements for NO_x, NO₂, PM₁₀, PM_{2.5}. The contribution from secondary organic particles is included in the regional calculations (using DEHM). For the local urban contribution (using UBM) (about 20 km up-wind from the receptor) only the primary particles are included in the calculations. Primary particles are assumed to dominate the local concentrations while secondary particle formation can be neglected at this scale. Furthermore primary particles are assumed to be the most relevant in relation to cancer risk, but it is not apparent how to distinguish the risk relative to secondary particles.

The air concentrations maps are shown in a 1x1 km² resolution grid for the entire country. The concentration intervals are coloured from light blue to dark brown representing no exceedances, whereas red represents

exceedance of pollutant threshold values. Since this project considers human health, only grid cells where there is any population are included for Denmark and grid cells with more than 20 persons for Sweden and Finland. This is done to save time on the comprehensive modelling especially for Sweden and Finland. This implies that approximately 96% of the area in Denmark is covered, typically omitting some coastal areas, which are hardly discernible. For Sweden 94% of the population of 9,527,546 persons corresponding to 27.5% of the populated area is included in the analysis. For Finland 91% of the total population of 5,334,016 that lives on 19.8% of the populated area is included.

2.2 Risk assessment

The risk quotient quantified by the Margin of Exposure (MoE), which is well established in the human health risk screening, is simply the EU air quality value (EEA, 2014) (threshold concentration) divided by the calculated air concentration:

$$\text{MoE} = \frac{\text{EU air quality value}}{\text{air (exposure) concentration}}$$

Equation 2.

When no EU air quality value is available the WHO Air Quality Guidelines (AQG) (WHO, 2000 update 2005) are used. In addition we calculate a MoE for carcinogens:

$$\text{MoE} = \frac{\text{acceptable risk level (1 : 100,000 cancer risk threshold value)}}{\text{air (exposure) concentration}}$$

Equation 3.

Where the numerator is the 1:100,000 risk concentrations from US-EPA IRIS (2015). These are mechanistic estimates since, in most cases, the shape of the risk curve is not known.

Classification groups are defined according to the IARC Monographs, Volumes 1–113 (<http://monographs.iarc.fr/ENG/Classification/>):

- Group 1: Carcinogenic to humans.
- Group 2A: Probably carcinogenic to humans.
- Group 2B: Possibly carcinogenic to humans.
- Group 3: Not classifiable as to its carcinogenicity to humans.
- Group 4: Probably not carcinogenic to humans.

The MoE approach is both a prioritization tool and a risk assessment tool, and a MoE below unity indicates risk. For carcinogens a 1:100,000 risk is equivalent to an increased lifetime chance of 0.00001 in 1 (or one chance in a hundred thousand) of developing cancer due to lifetime exposure to the pollutant concentration. A MoE of 10 express a safety factor of 10, or highest tolerable effect concentration (RfC), according to a 1:100,000 risk value, or a safety factor of 1 according to a 1:1,000,000 risk value. A MoE less than 10 could suggest further assessment is needed.

In Table 2 pollutant threshold values are shown, i.e. EU air quality limit and target values (EEA, 2014), WHO Air Quality Guidelines (AQG) (WHO, 2000 update 2005) and US-EPA IRIS (2015) 1:100,000 risk concentrations. In the last column the values used in this study are stated.

A total MoE (added risk from several pollutants) can in principle be calculated for pollutants with similar mode or mechanism of action, i.e. the mechanism by which a pollutant produces an effect on the human body. This implies that carcinogens and co-carcinogens cannot be aggregated. Co-carcinogens are chemical substances that cannot induce cancer when they are administered alone, but can enhance the carcinogenic effect of other substances. In general, co-carcinogens act as promoters in tissues in which the initiation stage has appeared (Haverkos, 2004). As can be seen from Table 2 the carcinogens are mainly related to lung cancer. A total MoE is obviously dependant on the number of pollutants being included. Accordingly the country with the most pollutants in the emission inventory has the potential for reaching the highest risk. However, as caution should be taken in using the MoE as a quantitative measure (Constable *et al.*, 2009) and in order not to cause unrealistic concern due to the uncertainty of assuming additive risks, a total MoE will not be calculated here.

Table 2: Threshold values for carcinogens (*italic*) and co-carcinogens covered by CLRTAP

Pollutant	CAS no	Threshold values (TV)	Values used
Particles			
Particles (unspecified)		NA	
PM _{2.5}		EEA (2014): 25 µg/m ³ annual mean, exposure concentration obligation of 20 µg/m ³ . WHO (2005): 10 µg/m ³ annual mean; 25 µg/m ³ 24 hr mean – lung	EU: 20 µg/m ³
PM ₁₀		EEA (2014): 40 µg/m ³ annual mean. WHO (2005): 20 µg/m ³ annual mean; 50 µg/m ³ 24 hr mean – lung	EU: 40 µg/m ³
Heavy metals			
<i>Arsenic</i>	7440–38–2	EEA (2014): 6 ng/m ³ . US-EPA IRIS (2015): 1 in 100,000 = 2 ng/m ³ (IARC group 1) – lung	EU: 6 ng/m ³ 1:100,000: 2 ng/m ³
<i>Cadmium</i>	231–152–8	WHO (2005) and EEA (2014): 5 ng/m ³ . US-EPA IRIS (2015): 1 in 100,000 = 6 ng/m ³ (IARC group 1) – lung	EU: 5 ng/m ³ 1:100,000: 6 ng/m ³
<i>Chromium</i>	7440–47–3	US-EPA IRIS (2015): 1 in 100,000 = 0.8 ng/m ³ for Cr(VI) WHO (2000): 1 in 100,000 = 0.25 ng/m ³ for Cr(VI) (IARC group 1 for Cr(VI) and group 3 for others) – lung	1:100,000: 0.8 ng/m ³ for Cr(VI)
<i>Lead</i>	7439–92–1	WHO (2005) and EEA (2014): 0.5 µg/m ³ (IARC group 2A–3) – lung	EU: 500 ng/m ³
<i>Mercury</i>	7439–92–1	WHO (2005): 1 µg/m ³ (inorganic mercury) (IARC group 3 for mercury and inorganic mercury compounds)	WHO: 1000 ng/m ³
<i>Nickel</i>	1295–35–8	EEA (2014): 20 ng/m ³ . US-EPA IRIS (2015): 1 in 100,000 = 20 ng/m ³ (Ni subsulfide) (IARC group 1) – lung	EU: 20 ng/m ³ 1:100,000: 20 ng/m ³
Organic compounds			
NM VOC (unspecified)		IARC group 3	
<i>Benzene</i>	71–43–2	EEA (2014): 5 µg/m ³ . US-EPA IRIS (2015): 1 in 100,000 = 1.3 µg/m ³ (IARC group 1) – leukaemia	
<i>PCDD/F (dioxins unspecified)</i>	9014–42–0	WHO (2000): An air quality guideline for PCDD/F is not proposed because direct inhalation exposures constitute only a small proportion of the total exposure, generally less than 5% of the daily intake from food. (IARC group 1)	NA
Inorganic compounds			
SO _x and SO ₂	7446–09–5	WHO (2005): 78 µg/m ³ annual mean (US); 20 µg/m ³ 24 hr; 500 µg/m ³ 10 min mean (for SO ₂) (IARC group 3 for SO ₂)	WHO US: 78 µg/m ³ for SO ₂
NO _x and NO ₂	10102–44–0	WHO (2005) and EEA (2014): 40 µg/m ³ annual mean; 200 µg/m ³ 1 hr mean (for NO ₂) (no IARC group)	NA for NO _x

Pollutant	CAS no	Threshold values (TV)	Values used
Polyaromatic Hydrocarbons (PAHs)			
PAH (unspecified)		NA	
<i>Benzo(a)pyrene</i>	50–32–8	EEA (2014): 1 ng/m ³ US-EPA IRIS (2015): 1 in 100,000 = 0.12 ng/m ³ (IARC group 1) – lung	EU: 1 ng/m ³ 1:100,000: 0.12 ng/m ³
<i>Benzo(b)fluoranthene</i>	205–99–2	TEF relative to B(a)P of 0,1	1:100,000: 1.2 ng/m ³
<i>Benzo(k)fluoranthene</i>	207–08–9	TEF relative to B(a)P of 0,1	1:100,000: 1.2 ng/m ³
<i>Indeno (1,2,3-cd)pyrene</i>	193–93–5	TEF relative to B(a)P of 0,1	1:100,000: 1.2 ng/m ³

Note: Threshold values: EU air quality limit and target values (EEA, 2014) and WHO Air Quality Guidelines (AQG) (WHO, 2000 update 2005) where EU values are given preference in this study. 1:100,000 risk concentrations for carcinogens (US-EPA IRIS, 2015).
NA: not applicable.
TEF: Toxicity Equivalency Factor (Cal-EPA, 2005).

3. Denmark

3.1 Air concentrations

3.1.1 Air measurements

Gridded (1x1 km² resolution) Danish air concentrations of PM_{2.5}, PM₁₀, *As*, *Cd*, *Cr*, *Hg*, *Ni*, *Pb*, NMVOC, *PCDD/F*, NO_x, *B(a)P*, *B(b)F*, *B(k)F* and *Indeno(cd)pyrene* are derived from UBM modelling of national sources and accounting for long-range transport from outside Denmark by adding a background concentration (Eq. 1) for the pollutants in italics, and from UBM (national sources) and DEHM (long-range transport) modelling for the other pollutants.

Measured air concentrations of POPs and heavy metals are found from campaigns at various locations e.g. in urban (Ellermann *et al.* 2012a&b) or rural regions (Wählin *et al.* 2010; Ellermann *et al.* 2011). In KoL 12-08 figures and tables can be found and in Table 3 the concentrations are summarized.

Table 3: Measured annual mean pollutant concentrations in Denmark

Pollutant	Rural	Sub-urban	Urban background	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(total)	0.5 0.52		1.2	7.0 3.7 3.7
Ni	2.0 2.47		3.4	2.9 2.8 5.7
Cu	2.2 1.41		13	97 42 37
As	0.7 0.46		0.8	0.9 1.0 0.7
Cd	0.1 0.08		0.1	0.2 0.2 0.1
Pb	4.1 3.15		4.5	5.9 5.8 3.9
Hg	1.5			
B(a)P		0.61		0.21
Dioxin	0.02		0.02	
Benzene			0.9–1.4	

Note: 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 (Hertel, 2009 and Ellermann, 2012a&b). Heavy metals: Annual statistics for Chromium (Cr(total)), 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). Risø and Anholt (rural) 2011 annual mean heavy metal concentrations are measured in total suspended particulate matter (TSP) (ng/m³). 0.61 ng B(a)P/m³ is a measured annual mean concentration in a wood stove area. The Hg concentrations are DEHM model calculations as average for the Northern Hemisphere (Christensen *et al.*, 2004). Dioxin is in pg I-TEQ/m³.

3.1.2 Model calculations

Recalculations are continuously being made in the emission inventories, and the latest recalculation in 2015 for small-scale combustion, which is essentially residential combustion with wood, include updated emission factors for metals and PAHs in connection with an update of the EMEP Guidebook (2012) and recalculations made by Danish Energy Agency (DEA, Energistyrelsen) on the used amount of wood. Most significantly updated 2010 emission amounts in kg/year are for As: 37 to 13.9, Cd: 47 to 483, Cr: 109 to 861, Pb: 1810 to 1117, Ni: 340 to 89.2, B(a)P: 4650 to 2318, B(b)F: 4710 to 2265, B(k)F: 2660 to 842, Indeno: 3230 to 1318.

The allocation of wood consumption made by the DEA was on municipality level and based on the national building and dwelling register and population density, thus concentrating a high fraction of the emissions in relatively small areas with high population density, resulting in an overestimation of emissions and concentrations in e.g. Copenhagen municipality. The updated method is based on location of wood stoves according to the national building and dwelling register and

including weightings between stoves and boilers, different building types, and primary and supplementary heating with residential wood combustion (Jensen *et al.*, 2015). As population density is not included in the new methodology, emissions are no longer accumulated in densely populated areas. Furthermore the previous methodology allocated a very large wood consumption to Bornholm and Lolland, which has been reduced significantly in the new methodology. The improved allocation of residential wood burners in Denmark, which will give a different distribution of burners but not a change in total emissions, is currently being implemented but is not available to the 2010 recalculated emissions in this project. It is not known if more or less of the Danish population will be exposed to pollutant levels exceeding the air quality levels or not. But probably the maximum values in the cities will be reduced and in rural areas the concentrations will increase.

To get an overview of the influence of the recalculations a simple adjustment of the calculated air concentrations covering all of Denmark is made for the PAHs, where residential wood combustion is the primary source (95% of total 2010 emissions). The metals are also influenced by the recalculations but residential wood combustion is not the most significant source of most metals. The maps for all pollutants other than B(a)P are based on emission data before recalculation.

Figure 1: PM_{2.5} (fine particles) air concentrations in a 1x1 km² resolution grid covering populated area of Denmark. There is no exceedance of the EU air quality value of 20 µg/m³. Highest national emission source categories are small-scale combustion, i.e. residential combustion, followed by road traffic and livestock in agriculture

Calculated annual mean PM_{2.5} concentrations of primary particles from national sources are $0.79 \pm 0.40 \mu\text{g}/\text{m}^3$ and the calculated annual mean PM_{2.5} concentrations including regional sources are $9.5 \pm 0.25 \mu\text{g}/\text{m}^3$. The difference between national and regional sources varies throughout the country, where the influence from regional sources decreases from the southeastern to the northwestern Denmark. Calculated concentrations in urban areas (Copenhagen) are 1.86 and $10.3 \mu\text{g}/\text{m}^3$ from national sources and all sources including long-range transport, respectively. Secondary organic particles are included as described in section 2.1.

Figure 2: PM₁₀ (coarse particles) air concentrations in a 1x1 km² resolution grid covering populated area of Denmark. There is no exceedance of the EU air quality value of 40 µg/m³. Highest national emission source categories are small-scale combustion, i.e. residential combustion, followed by livestock in agriculture and road traffic

National and regional sources are mainly residential combustion, agriculture and road traffic and furthermore a significant regional source along the western shores of Jutland and Zealand is sea salt. Calculated annual mean PM₁₀ concentrations of primary particles from national sources are 1.1 ± 0.54 µg/m³ and the calculated annual mean PM₁₀ concentrations including regional sources are 12.5 ± 1.1 µg/m³. Calculated urban background concentrations (Copenhagen) are 2.1 and 12 µg/m³ from national sources and all sources including long-range transport, respectively.

Figure 3: Arsenic air concentrations in a 1x1 km² resolution grid covering populated area of Denmark. There is no exceedance of the EU air quality value of 6 ng/m³ or the 1:100,000 cancer risk value of 2 ng/m³. Highest national emission source categories are industrial combustion followed by public power, shipping and small-scale combustion, i.e. residential combustion

For arsenic the measured rural annual mean concentrations are 0.7 and 0.46 ng/m³ for Risø and Anholt, respectively (Table 3). The modelled rural annual mean concentration (Risø) from national sources is 0.008 ng/m³, and the modelled urban background annual mean concentration (Copenhagen) from national sources is 0.012 ng/m³. To derive the total air concentration from modelling the national sources a calculated background concentration (Eq. 1) of 0.57 ng/m³ is added. The regional background contributions are predominant both in rural (99%) and urban (85%) sites for arsenic.

Figure 4: Cadmium air concentrations in a 1x1 km² resolution grid covering populated area of Denmark. There is no exceedance of the EU air quality value of 5 ng/m³ or the 1:100,000 cancer risk value of 6 ng/m³. Highest national emission source categories are small-scale combustion, i.e. residential combustion, followed by road traffic and industrial combustion

For cadmium the measured rural annual mean concentrations are 0.1 and 0.08 ng/m³ (Table 3) for Risø and Anholt, respectively. The modelled rural annual mean concentration (Risø) from national sources is 0.008 ng/m³, and the modelled urban background annual mean concentration (Copenhagen) from national sources is 0.015 ng/m³. To derive the total air concentration from modelling the national sources a calculated background concentration (Eq. 1) of 0.08 ng/m³ is added. The regional background contributions to cadmium in ambient air in Denmark are significant in rural sites (91%) followed by national residential combustion. In urban areas the regional background contributes with 83%.

Figure 5: Chromium(VI) air concentrations in a 1x1 km² resolution grid covering populated area of Denmark. There is no exceedance of the US-EPA IRIS 1:100,000 risk concentration of 0.8 ng/m³. Highest national emission source categories are small-scale combustion, i.e. residential combustion, use of solvents and other products, road and rail traffic and public power

For chromium(total) the measured rural annual mean concentrations are 0.5 and 0.52 ng/m³ (Table 3) for Risø and Anholt, respectively. The modelled rural annual mean concentration (Risø) from national sources is 0.06 ng/m³, and the modelled urban background annual mean concentration (Copenhagen) from national sources is 0.13 ng/m³. To derive the total air concentration from modelling the national sources a calculated background concentration for total Cr (Eq. 1) of 0.45 ng/m³ is added. This indicates that the regional background contributions to total Cr in ambient air in Denmark is significant in rural sites (88%) followed by national residential combustion. In urban areas the regional background contributes with 63% of the total Cr in air.

When inhaled, only Cr(VI) is carcinogenic in humans and the 1:100,000 value is therefore valid for Cr(VI) only (WHO, 2000). ATSDR (2008) reports that Cr(VI) account for approximately one third of the total chromium emitted to the atmosphere annually in the USA. However, the characteristics of the sources and the environmental conditions govern the distribution between Cr(VI) and the non-carcinogenic Cr(III) in the ambient air. The data quantifying this

distribution are scarce. Yu *et al.* (2014) found that Cr(VI) concentrations were more significantly impacted by meteorological factors than traffic and that total Cr, were not significantly correlated with ambient Cr(VI) concentrations. In order to derive mean annual Cr(VI) concentrations from the calculated mean annual Cr(total) concentrations in this project, we will use the factor: $\text{mean Cr(VI)}/\text{mean Cr(total)} = 0.035$ derived from the measured values in Yu *et al.* (2014). This factor indicates that approximately 3.5% of the Cr(total) concentrations in ambient air is Cr(VI), all other things being equal. This value corresponds with an estimate by the State of California Air Resources Board (ARB, 1986) stating that 3 to 8% of total ambient chromium is in the hexavalent state.

The background concentration of Cr(VI) in Denmark is thus 0.016 ng/m³ and the relative contribution from long-range transport is similar to that of total Cr.

Figure 6: Lead air concentrations in a 1x1 km² resolution grid covering populated area of Denmark. There are no exceedances of the EU air quality value of 500 ng/m³. Highest national emission source categories are road and rail traffic, small-scale combustion, i.e. residential combustion and aviation

For lead the measured rural annual mean concentrations are 4.1 and 3.15 ng/m³ for Risø and Anholt, respectively (Table 3). The modelled rural annual mean concentration (Risø) from national sources is 0.77 ng/m³, and the modelled urban background annual mean concentration

(Copenhagen) from national sources is 2.0 ng/m³. To derive the total air concentration from modelling the national sources a calculated background concentration (Eq. 1) of 2.9 ng/m³ is added. The regional background contributions are significant both in rural (79%) and urban background (60%) sites for lead.

For mercury there are no exceedances of the WHO Air Quality Guideline value of 1,000 ng/m³. Highest national emission source categories are public power, industrial combustion, waste incineration and small-scale combustion, i.e. residential combustion.

DEHM model calculations of Hg mean concentration in the Northern Hemisphere gives a background concentration of 1.5 ng Hg/m³. This background concentration applies for Denmark and the other Nordic countries. Hg sources have been reduced significantly in the EU and the major part of Hg emissions on the Northern Hemisphere is in Asia (Christensen *et al.*, 2004). Regional sources are most significant (99%).

Figure 7: Nickel air concentrations in a 1x1 km² resolution grid covering populated area of Denmark. There is no exceedance of the EU air quality value of 20 ng/m³ or the 1:100,000 cancer risk value of 20 ng/m³. Highest national emission source categories are industrial combustion, shipping and public power

For nickel the measured rural annual mean concentrations are 2.0 and 2.47 ng/m³ for Risø and Anholt, respectively (Table 3). The modelled rural annual mean concentration (Risø) from national sources is 0.12

ng/m³, and the modelled urban background annual mean concentration (Copenhagen) from national sources is 0.17 ng/m³. To derive the total air concentration from modelling the national sources a calculated background concentration (Eq. 1) of 2.1 ng/m³ is added. The regional background contributions are significant both in rural (95%) and urban background (79%) sites for nickel. Maximum nickel concentrations occur in the vicinity of industrial combustion sites and power plants and at these localities the contribution from national sources are predominant.

Figure 8: PCDD/F (dioxins) air concentrations in a 1x1 km² resolution covering populated area of Denmark. No EU air quality value is given for dioxins. Highest national emission source categories are small-scale combustion, i.e. residential combustion, followed by waste disposal

The physical/chemical properties of dioxins allow the compounds to be present in the gasphase as well as sorbed to particles in the ambient air. The distribution is temperature dependant, with higher occurrence in the gasphase at higher temperatures. Atmospheric transport and deposition are different for the two phases; however, transport modelling in this study assumes inert compounds with no chemical transformation. Annual mean measured dioxin concentrations are 0.02 pg I-TEQ/m³ both at rural and urban sites, and with modelled air concentrations of 0.002 and 0.003 pg I-TEQ/m³ at rural and urban sites, respectively, a calculated background concentration (Eq. 1) of

0.02 pg/m³ is found. Regional sources are predominant to dioxins in rural (92%) and urban background sites (88%).

Figure 9: NO_x air concentrations in a 1x1 km² resolution grid covering populated area of Denmark. No EU air quality value is given for NO_x. Highest national emission source categories are road and off-road traffic, shipping, public power and industrial combustion

Modelled annual mean NO_x concentration from national sources is 5.9 ±5.2 µg/m³ and the modelled annual mean NO_x concentration including regional sources is 11 ±5.5 µg/m³ where the standard deviations reflect the variation throughout the country. Modelled urban background concentrations (Copenhagen) are 18 and 24 µg/m³ from national sources and all sources including long-range transport, respectively.

There are modelled high values around six industrial and power production point sources. The positioning of sources and receptor points may not be exactly overlapping in the 1x1 km² grid and the height of sources may not be exact, which introduces errors in the model results for point sources. Therefore these modelled concentrations are assigned with >40 µg/m³ in the map.

Figure 10: Benzo(a)pyrene air concentrations in a 1x1 km² resolution grid covering populated area of Denmark. There are no exceedances of the EU target value of 1 ng/m³. There are exceedances of the 1:100,000 cancer risk value of 0.12 ng/m³ in approximately 60% of the populated area. Highest national emitter is small-scale combustion, i.e. residential combustion

For benzo(a)pyrene the adjusted (from recalculations) modelled annual mean air concentration (busy traffic locations in Copenhagen) from national sources is: $0.292 \text{ ng/m}^3 * (2318/4900 + (4900 - 4650)/4900) = 0.15 \text{ ng/m}^3$, where 4,900 kg/y is the total B(a)P emission. The measured annual mean air concentration in Copenhagen (H.C. Andersens Boulevard) is 0.21 ng/m^3 (Table 3). The measured annual mean concentration in a wood stove area is 0.61 ng/m^3 (Table 3). The latter cannot be reproduced by the urban scale modelling as there are conditions in a hot spot area that need to be modelled on local scale. The calculation of the regional background concentration is based on the measurement in Copenhagen as there are no measured annual means of benzo(a)pyrene at rural sites for 2010 or 2011. The total ambient air concentrations are then found from adding the modelled concentrations derived from national sources with a background concentration (Eq. 1) of 0.06 ng/m^3 . This indicates that the benzo(a)pyrene air concentration in urban background has a significant contribution from national sources (73%), where the main emissions are from residential combustion.

For the other three PAHs (benzo(b)fluoranthene, benzo(k)fluoranthene and indeno(1,2,3-cd)pyrene) the concentrations are found from the benzo(a)pyrene concentrations multiplied with the ratio of annual emissions. The highest national emitter and the predominant emitter overall is small-scale combustion, i.e. residential combustion, so the concentration profiles in Denmark are similar to benzo(a)pyrene. There are no exceedances of the 1:100,000 cancer risk value of 1.2 ng/m³ for the three PAHs. In EU as an average, the measured annual mean benzo(a)pyrene concentration has shown an increase of 0.2% per year over the last 6 years (EEA, 2014). Emissions of benzo(a)pyrene increased by 21% from 2003 to 2012, driven by the increase (24%) from residential wood combustion (EEA, 2014).

Long-range transport is significant to the ambient Danish air for all pollutants except the PAHs, which are mainly associated with residential wood combustion. NO_x, chromium and lead have significant contributions from national sources, especially in urban sites, due to primarily road traffic, residential wood combustion and energy production and industrial point sources.

In Table 4 the measured and modelled annual mean rural and urban background air concentrations in Denmark are compiled for each pollutant. Contribution from Danish and regional sources to ambient air concentration and the most significant sources are stated. Percentage (%) populated area and population with exceedance of EU air quality values and 1:100,000 cancer risk for carcinogens are also shown.

Table 4: Rural, urban and regional background air concentrations in Denmark. Contribution from Danish sources to ambient air concentration, modelled with UBM. Most significant sources are stated in descending order. % populated area and population with exceedance of EU air quality values and 1:100,000 cancer risk for carcinogens (italic)

Denmark	PM _{2.5} µg/m ³	PM ₁₀ µg/m ³	As ng/m ³	Cd ng/m ³	Cr(total) ng/m ³	Pb ng/m ³	Hg ng/m ³	Ni ng/m ³	PCDD/F pg/m ³	NO _x µg/m ³	B(a)P ng/m ³	B(b)F ng/m ³	B(k)F ng/m ³	Indeno ng/m ³
<i>Rural air conc</i>	Total DK	Total DK								Total DK				
DK sources	0.79±0.40	1.1±0.54	0.008	0.008	0.06	0.77	0.008	0.12	0.002	5.9±5.2	0.12	0.1	0.03	0.07
Total (REG and DK)	9.5±0.25 ²⁾	12.5±1.1 ²⁾	0.58 ³⁾	0.09 ³⁾	0.51 ³⁾	3.6 ³⁾	1.5 ²⁾	2.2 ³⁾	0.02 ³⁾	11±5.5 ²⁾	–	–	–	–
<i>Urban background air conc</i>	Total DK	Total DK								Total DK				
DK sources	0.79±0.40	1.1±0.54	0.012	0.015	0.13	2.0	0.016	0.17	0.003	5.9±5.2	0.15	0.2	0.04	0.09
Total (REG and DK)	9.5±0.25 ²⁾	12.5±1.1 ²⁾	0.8 ³⁾	0.1 ³⁾	1.2 ³⁾	4.5 ³⁾	–	3.4 ³⁾	0.02 ³⁾	11±5.5 ²⁾	0.21 ³⁾	–	–	–
Regional background air conc	8.7±0.41 ²⁾	11±1.4 ²⁾	0.57	0.08	0.45	2.9	1.5	2.1	0.02	5.0±0.74 ²⁾	0.06	–	–	–
<i>Contrib of DK sources to air conc</i>														
Rural air	7.8% (mean rur & urb)	8.3% (mean rur & urb)	1.4%	9%	12%	21%	0.5%	5%	8%	47% (mean rur & urb)	–	–	–	–
Urban air			15%	17%	37%	40%	–	21%	12%		73%	73% ¹⁾	73% ¹⁾	73% ¹⁾
Most significant sources (in decending order)	REG (southern and eastern DK) DK res com DK road	REG (western shores) DK res com DK agri DK road	REG DK ind com DK pub power	REG DK res com	REG DK res com DK pub power DK road and rail DK solvents	REG DK road and rail DK res com DK aviation	REG DK pub power	REG DK ind com DK ship	REG DK res com DK waste	REG (mean 53%) DK road DK shipping DK pub power DK ind com	DK res com REG	DK res com REG	DK res com REG	DK res com REG
% of populated area > EU qual	NE	NE	NE	NE	Cr(VI): NTV	NE	NE	NE	NTV	NTV	NE	NE	NE	NE
Population > EU qual (% of total population)														
% of populated area > 1:100,000	NTV	NTV	NE	NE	Cr(VI): NE	NTV	NTV	NE	NTV	NTV	60%	NE	NE	NE
Population > 1:100,000 (% of total population)											4,500,000 (80%)			

Note: ¹⁾ Assuming same source distrubution as B(a)P.

²⁾ Modelled with DEHM and UBM, including national and regional sources.

³⁾ Measurements at rural (Risø and Anholt) and urban background (Copenhagen) sites. Regional background concentrations are based on measurements and modelling of rural air concentrations, except for particles and NO_x where they are calculated for entire populated area.

NTV: No threshold value (TV). NE: No exceedance of TV. -: No value.

Sources: REG: Regional sources, res com: residential combustion, pub power: public power.

3.2 Risk towards the Danish population

Benzo(a)pyrene has an annual mean concentration that exceeds the 1:100,000 cancer risk values for approximately 80% of the Danish population. Note that the EU target value threshold for benzo(a)pyrene of 1 ng/m³ is *not* exceeded. Residential wood combustion comprises the most influential source for benzo(a)pyrene.

All other investigated pollutants comply with their respective EU air quality values and 1:100,000 cancer risk values.

Recalculations show significant increases of cadmium and chromium emissions from residential wood combustion. Adjustment of the calculated concentrations at a site influenced mainly by residential wood combustion (Jyllinge north), show concentrations that do not exceed the EU target values or 1:100,00 cancer risk values.

4. Finland

4.1 Source category emissions

There have been some corrections in the Finnish emission values compared to the KoL 12–08 report. The updated emission values used in the modelling in this report are shown in Table 5.

Table 5: Finnish 2010 emissions in t per year for different pollutants and aggregated sectors

	PM _{2.5}	PM ₁₀	SO ₂	NO _x (as NO ₂)	NMVOC
Traffic and machinery exhaust (excl. Air and marine)	3,266	3,651	196	62,767	32,435
Other area sources (incl. Road traffic non-exhaust)	21,446	41,348	13,687	24,074	31,380
Point sources	4,086	6,935	53,598	64,311	0
Total	28,799	51,934	67,480	151,153	63,815

4.2 Air concentrations

4.2.1 Model calculations

Gridded (1x1 km² resolution) Finnish air concentrations of PM_{2.5}, PM₁₀, SO₂ and NO_x are derived from UBM modelling of national sources and accounting for regional background concentrations with the DEHM model.

To reduce the calculation time for the time demanding models only the part of Finland with a population more than 20 persons per grid cell (1x1 km²) is included. This implies that 90.5% of the population of 5,334,016 persons corresponding to 19.8% of the populated area is included in the analysis.

The calculated annual mean PM_{2.5} concentration from national sources is 0.81 ±0.70 µg/m³ and the calculated annual mean PM_{2.5} concentration including regional sources is 6.2 ±1.6 µg/m³. National sources contribute an average of 13% to the total PM_{2.5} concentration in the populated (>20 persons) part of Finland, the predominant contribution is from long-range transport.

Figure 11: PM_{2.5} (fine particles) air concentrations in a 1x1 km² resolution grid covering 19.8% of populated area and 90.5% of population in Finland. There is no exceedance of the EU air quality value of 20 µg/m³. The main national emission source categories are various area sources other than traffic

Figure 12: PM₁₀ (coarse particles) air concentrations in a 1x1 km² resolution grid covering 19.8% of populated area and 90.5% of population in Finland. There is no exceedance of the EU air quality value of 40 µg/m³. The main national emission source categories are various area sources other than traffic

The calculated annual mean PM₁₀ concentration from national sources is $1.8 \pm 1.9 \mu\text{g}/\text{m}^3$ and the calculated annual mean PM₁₀ concentration including regional sources is $7.8 \pm 2.7 \mu\text{g}/\text{m}^3$. National sources contribute an average of 23% to the total PM₁₀ concentration in the populated (>20 persons) part of Finland. The predominant contribution is from long-range transport.

Figure 13: SO₂ air concentrations in a 1x1 km² resolution grid covering 19.8% of populated area and 90.5% of population in Finland. There is no exceedance of the US air quality value 78 $\mu\text{g}/\text{m}^3$. The highest national emission source categories are point sources

The calculated annual mean SO₂ concentrations from national sources only and all sources including regional are 2.1 ±2.8 µg/m³ and 3.1 ±2.9 µg/m³, respectively. National sources contribute an average of 67% to the total SO₂ concentration in the populated (>20 persons) parts of Finland.

There are modelled high values around some point sources. The positioning of sources and receptor points may not be exactly overlapping in the 1x1 km² grid and the height of sources may not be exact, which introduces errors in the UBM results for point sources. Therefore these modelled concentrations are assigned with >78 µg/m³ in the map.

Figure 14: NO_x air concentrations in a 1x1 km² resolution grid covering 19.8% of populated area and 90.5% of population in Finland. No EU air quality value is given for NO_x. Highest national emission source categories are traffic and machinery exhaust (excl. air and marine), other area sources and point sources

The calculated annual mean NO_x concentrations from national sources only and all sources including regional are 3.7 ±4.1 µg/m³ and 5.9 ±4.8 µg/m³, respectively. National sources contribute an average of 64% to the total NO_x concentration in the populated (>20 persons) part of Finland.

In Table 6 the modelled ambient air concentrations in Finland are compiled for each pollutant. Contribution from Finnish and regional sources and the most significant sources are stated. Percentage populated area and population with exceedance of threshold values for the co-carcinogens are also shown.

Long-range transport is significant for all pollutants, and furthermore for SO₂ and NO_x national point sources are significant.

Table 6: Ambient air concentrations in Finland for co-carcinogens, and contribution from Finnish sources modelled with UBM and DEHM. Most significant sources are stated in descending order. % populated area with > 20 persons pr grid (91% of population), and population with exceedance of EU air quality values

Finland	PM _{2.5} µg/m ³	PM ₁₀ µg/m ³	SO ₂ µg/m ³	NO _x µg/m ³
<i>Air conc</i>				
FI sources	0.81 ±0.70	1.8 ±1.9	2.1 ±2.8	3.7 ±4.1
Total (REG and FI)	6.2 ±1.6	7.8 ±2.7	3.1 ±2.9	5.9 ±4.8
Regional background air conc	5.4±1.2	6.0±1.2	1.0±0.2	2.2±1.0
Contrib of FI sources to air conc	13% (country mean)	23% (country mean)	67% (country mean)	64% (country mean)
Most significant sources (in descending order)	Regional sources FIN area sources	Regional sources FIN area sources	FIN point sources Regional sources	FIN point sources FIN traffic exhaust Regional sources
% of populated area > EU qual	No exceedance	No exceedance	No exceedance	No limit value
Population > EU qual (% of total population)	No exceedance	No exceedance	No exceedance	No limit value

4.3 Risk towards the Finnish population

All calculations are for the part of Finland where there are more than 20 persons living per 1x1 km². This covers 90.5% of the total population of 5,334,016 living on 19.8% of the populated area. All investigated pollutants comply with their respective EU air quality values.

There is no observed cancer risk explicitly from residential wood combustion in Finland, a probable reason being that the PAHs, which are the most critical pollutants for this source, are not included in the Finnish data.

5. Sweden

5.1 Source category emissions

There have been some corrections in the Swedish emission values compared to the KoL 12-08 report, and the updated emission values used in the modelling in this report are stated in Table 7.

5.2 Air concentrations

Gridded (1x1 km² resolution) Swedish air concentrations of PM_{2.5}, PM₁₀, *As*, *Cd*, *Cr*, *Hg*, *Ni*, *Pb*, *NMVOC*, *PCDD/F*, SO₂, NO_x and *PAH-4* are derived from UBM modelling of national sources and adding background concentrations from long-range transport (Eq. 1) for the pollutants in italics. For the other pollutants the total ambient air concentrations are found from UBM (national sources) and DEHM (long-range transport) modelling. Concentrations that are measured and modelled in previous studies for rural, urban background and street canyon conditions are compiled in Table 8, and are used to evaluate and adjust the calculated urban background concentration levels.

To reduce the calculation time for the time demanding models only the part of Sweden with a population more than 20 persons per grid cell (1x1 km² resolution) is included. This implies that 94% of the population of 9,527,546 persons corresponding to 27.5% of the populated area is included in the analysis.

Table 7: Emissions for different pollutants per sector for year 2010 for Sweden

Sector	NMVOC (t/y)	NO _x (t/y)	SO ₂ /SO _x (t/y)	PM _{2.5} (t/y)	PM ₁₀ (t/y)	As (kg/y)	Cd (kg/y)	Cr(total) (kg/y)	Hg (kg/y)	Ni (kg/y)	Pb (kg/y)	PCDD/ PCDF (mg I- teq/y)	PAH-4 (kg/y)
Combustion in energy and transformation industries	3,916	17,426	9,918	4,292	5,650	273	180	748	192	5,203	2,948	23,673	751
Agriculture	0	0	0	803	4,476	0	0	0	0	0	0	0	0
Other sources and sinks	4,058	206,643	47,411	7,717	7,717	0	0	0	0	0	1,552	0	0
Non-industrial combustion plants	12,058	5,558	952	6,563	6,565	28	168	172	29	268	864	3,847	10,858
Combustion in manufacturing industry	1,708	14,256	5,288	1,856	2,318	110	90	386	63	7,358	1,683	8,979	237
Production processes	4,581	79	0	197	1,537	9	5	3	0	7	4	0	0
Extraction and distribution of fossil fuels and geothermal energy	5,194	0	0	83	317	0	0	0	0	0	0	0	0
Solvent use and other product use	86,788	0	0	319	319	0	0	0	0	0	0	0	0
Road transport	32,223	63,470	80	3,118	9,906	0	3	2	0	4	4,621	551	91
Other mobile sources and machinery	26,337	32,538	19,84	2,243	2,329	77	2	112	0	3,058	490	56	5
Waste treatment and disposal	0	0	0	219	219	0	0	0	51	0	0	855	30
Point sources	18,768	16,278	137,61	5,232	6,471	403	96	3,551	146	1,610	3,410	3,913	1,152
Total	195,631	356,249	793,94	32,642	47,824	899	542	4,974	481	17,508	15,572	41,873	13,123

5.2.1 Air measurements

Table 8: Measured annual mean pollutant concentrations in Sweden

Pollutant	Rural	Urban background	Street
PM _{2.5}	5–12	7–13	7–20
PM ₁₀	6–18	7–25	12–45
Cr(total)	0.30 0.09 0.08 0.11 0.476 0.07 0.11 2.74	2.3 (±2.1) 3.0 (±1.1)	6.1 (±5.1)
Ni	1.47 0.44 0.38 0.56 2.358 2.00 1.83 4.88	2.3 (±1.9) 2.7 (±1.8)	2.9 (±2.7)
As	0.22 0.07 0.06 0.08 0.35 0.09 0.11 0.02	0.88 (±0.92) 0.88 (±0.36)	1.04 (±1.0)
Cd	0.06 0.02 0.02 0.02 0.0956 – – –	0.11 (±0.09) 0.31 (±0.36)	0.12 (±0.11)
Pb	0.00 0.64 0.55 0.82 3.44 2.15 1.77 0.28	3.4 (±3.1) 9.1 (±13)	7.2 (±5.5)
Hg (particulate)		11.7 (±6.2) 8.5 (±6.5)	19.5 (±8.1)
Hg (gaseous)	1.5	1.5 (±0.9) 1.7 (±0.35)	2.2 (±1.4)
Dioxin	0.02	0.02	
B(a)P	0.05–0.1	0.14 0.20	

Note: Metal concentrations (mean ±st.dev.) measured at a roof-top site (urban background) in central Stockholm September 2003–September 2004, August 1995–September 1996 and in Hornsgatan (street canyon) in central Stockholm September 2003–September 2004 (Johansson *et al.*, 2009). Mean is calculated assuming concentration is equal to detection level if concentration is below detection level. Long-range concentrations in four cities in the Swedish Urban Air Quality Network, and from Hornsgatan in central Stockholm (three model results for PM₁₀ and one for PM_{2.5}) (Sjödén *et al.*, 2010). Background Hg mean concentration applies for the Northern Hemisphere. PM₁₀ and PM_{2.5} concentrations are from Omstedt *et al.* (2010). Units are in ng/m³ for metals except Hg (particulate) in pg/m³ and dioxin in pg I-TEQ/m³, and µg/m³ for the remaining pollutants. – indicates no model result.

5.2.2 Model calculations

Figure 15: PM_{2.5} (fine particles) air concentrations in a 1x1 km² resolution grid covering 27.5% of populated area and 94% of population in Sweden. There are no exceedances of the EU air quality value of 20 µg/m³. There are many equally significant national emission source categories including road transport, non-industrial combustion plants, point sources and combustion in energy production

Measured annual mean concentrations of PM_{2.5} vary between 5–12 µg/m³ (regional background), 7–13 µg/m³ (urban background) and 7–20 µg/m³ (street) (Omstedt *et al.*, 2010; Omstedt *et al.*, 2014), see Table 8. The modelled annual mean PM_{2.5} concentration of primary particles from national sources is 0.55 ± 0.70 µg/m³ and the modelled annual mean PM_{2.5} concentration including regional sources is 6.9 ± 2.3 µg/m³. This factor 12 difference in mean concentrations from national sources and total varies throughout the country as there is a north-south gradient in the regional background concentrations. For urban and street sites the concentrations are influenced by the population size, local emissions and meteorology. National sources contribute an average of 8% to the total PM_{2.5} concentration in the populated (>20 persons) part of Sweden, the predominant contribution is from long-range transport. The contribution to PM_{2.5} in ambient air (mean value for Sweden) from residential wood combustion is 2%. Secondary organic particles are included as described in section 2.1.

There are modelled high values around some point sources. The positioning of sources and receptor points may not be exactly overlapping in the 1x1 km² grid and the height of sources may not be exact, which introduces errors in the UBM results for point sources. Therefore these modelled concentrations are assigned with >20 µg/m³ in the map.

Figure 16: PM₁₀ (coarse particles) air concentrations in a 1x1 km² resolution grid covering 27.5% of populated area and 94% of population in Sweden. There are no exceedances of the EU air quality value of 40 µg/m³. The sources are similar to PM_{2.5} sources with an added contribution from road transport and agriculture

Measured concentrations of PM₁₀ at regional background stations vary in the country, with the highest concentrations in the southern part near the densely populated Öresundsregion and on the west coast where the influence from sea spray is relatively high. Regional background annual mean concentrations vary between 13–18 µg/m³ in southern and western Götaland, 9–15 µg/m³ in eastern Svealand and 6–8 µg/m³ in Västerbotten (Omstedt *et al.*, 2010). For urban background typically measured on roof tops in the centre of villages/cities, e.g. in Stockholm, Göteborg and Malmö the PM₁₀ concentrations vary depending on population density and location. Annual mean concentrations are 13–25 µg/m³ in southern Götaland, 10–23 µg/m³ in northern Götaland, 11–20 µg/m³ in Svealand and 7–18 µg/m³ in Norrland, see Table 8 (Omstedt *et al.*, 2010).

The calculated annual mean PM₁₀ concentration of primary particles from national sources is 0.94 ±1.2 µg/m³ and the calculated annual mean PM₁₀ concentration including regional sources is 8.2 ±3.0 µg/m³. National sources contribute an average of 11% to the total PM₁₀ concentration in the populated (>20 persons) part of Sweden. The predominant contribution is from long-range transport where the contribution is from primary and secondary particulate matter 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 regional contribution. The contribution to PM₁₀ in ambient air (mean value for Sweden) from residential wood combustion is 1.6%.

There are modelled high values around some point sources. The positioning of sources and receptor points may not be exactly overlapping in the 1x1 km² grid and the height of sources may not be exact, which introduces errors in the UBM results for point sources. Therefore these modelled concentrations are assigned with >40 µg/m³ in the map.

Measured air concentrations of heavy metals are found from campaigns in central Stockholm at a roof-top site (urban background) in 1995–1996 and 2003–2004 and in Hornsgatan (street canyon) in 2003–2004 (Johansson *et al.*, 2009), see Table 8. Calculated (COPREM model) long-range (background) contributions to measured metal concentration in PM₁₀ in ng/m³ in four cities in the Swedish Urban Air Quality Network, and from Hornsgatan in central Stockholm (three model results for PM₁₀ and one for PM_{2.5}) (Sjödin *et al.*, 2010) are shown in Table 8.

Figure 17: Arsenic air concentrations in a 1x1 km² resolution grid covering 27.5% of populated area and 94% of population in Sweden. There are no exceedances of the EU air quality value of 6 ng/m³ or the 1:100,000 cancer risk value of 2 ng/m³. Highest national emission source categories are point sources, energy production and combustion in manufacturing industry

In rural areas the measured arsenic annual mean concentration is 0.125 ng/m³ (Table 8), and the modelled rural annual mean concentration from national sources is 0.002 ng/m³. In central Stockholm (urban background) the measured annual mean concentration is 0.88 ng/m³ (Table 8), and the modelled urban background concentration from national sources is 0.10 ng/m³. To derive the total air concentration from modelling the national sources a calculated background concentration (Eq. 1) of 0.12 ng/m³ is added. The regional background contributions are predominant both in rural (99%) and urban (51%) sites for arsenic. The contribution to arsenic in ambient air (mean value for Sweden) from residential wood combustion is 0.4%.

There are modelled high values around some point sources. The positioning of sources and receptor points may not be exactly overlapping in the 1x1 km² grid and the height of sources may not be exact, which introduces errors in the UBM results for point sources. Therefore these modelled concentrations are assigned with >2 ng/m³ in the map.

Figure 18: Cadmium air concentrations in a 1x1 km² resolution grid covering 27.5% of populated area and 94% of population in Sweden. There is no exceedance of the EU air quality value of 5 ng/m³ or the 1:100,000 cancer risk value of 6 ng/m³. Highest national emission source categories are small-scale energy production, non-industrial combustion and point sources

In rural areas the measured cadmium annual mean concentration is 0.043 ng/m³ (Table 8), and the modelled rural annual mean concentration from national sources is 0.0037 ng/m³. In central Stockholm (urban background) the measured annual mean concentration is 0.21 ng/m³ (Table 8), and the modelled urban concentration from national sources is 0.08 ng/m³. To derive the total air concentration from modelling the national sources a calculated background concentration (Eq. 1) of 0.04 ng/m³ is added. This indicates that regional background contribution to cadmium to ambient air in Sweden is significant both in rural (91%) and urban (40%) ambient air. The contribution to cadmium in ambient air (mean value for Sweden) from residential wood combustion is 7%.

Figure 19: Chromium(VI) air concentrations in a 1x1 km² resolution grid covering 27.5% of populated area and 94% of population in Sweden. There is no exceedance of the US-EPA IRIS 1:100,000 risk concentration of 0.8 ng/m³. Highest national emission source categories are point sources, energy production and combustion in manufacturing industries

In rural areas the measured total chromium annual mean concentration is 0.50 ng/m³ (Table 8), and the modelled total chromium rural annual mean concentration from national sources is 0.008 ng/m³. In central Stockholm (urban background) the measured total chromium annual mean concentration is 2.65 ng/m³ (Table 8), and the modelled urban total chromium concentration from national sources is 0.35 ng/m³. To derive the total chromium air concentration from modelling the national sources a calculated background concentration (Eq. 1) of 0.49 ng/m³ is added. The regional background contributions are significant both in rural (99%) and urban (53%) sites for chromium. The contribution to total chromium in ambient air (mean value for Sweden) from residential wood combustion is 0.7%. As outlined in section 2.1 the fraction of total chromium that is chromium(VI) is assumed to be 3.5%, which gives a regional background chromium(VI) concentration of 0.017 ng/m³. The percentages of national and regional contribution of chromium(VI) to the rural and urban background air and contribution of residential wood combustion are as for total chromium.

Figure 20: Lead air concentrations in a 1x1 km² resolution grid covering 27.5% of populated area and 94% of population in Sweden. There are no exceedances of the EU air quality value of 500 ng/m³. Highest national emission source categories are road traffic, energy production, point sources and combustion in manufacturing industries

In rural areas the measured lead annual mean concentration is 1.21 ng/m³ (Table 8), and the modelled rural annual mean concentration from national sources is 0.09 ng/m³. In central Stockholm (urban background) the measured annual mean concentration is 6.25 ng/m³ (Table 8), and the modelled urban background concentration from national sources is 2.4 ng/m³. To derive the total air concentration from modelling the national sources a calculated background concentration (Eq. 1) of 1.1 ng/m³ is added. The regional background contributions are significant both in rural (92%) and urban (40%) sites for lead. The contribution to lead in ambient air (mean value for Sweden) from residential wood combustion is 1%.

For mercury there are no exceedances of the WHO Air Quality Guideline value of 1,000 ng/m³. Highest national emission source categories are energy production and point sources. The DEHM model calculations of the Hg mean background concentration in the northern hemisphere are 1.5 ng Hg/m³ (Table 8), and the measured urban background annual mean concentration is 1.6 ng/m³ (Table 8). The modelled rural and urban (Stockholm) annual mean mercury concentrations from national sources are 0.001 and 0.07 ng/m³, respectively. 1.5 ng/m³ is approximately the total ambient air concentration in all of Sweden. The regional background contributions are predominant both in rural (99.9%) and urban (95%) sites. The contribution to mercury in ambient air (mean value for Sweden) from residential wood combustion is 0.04%.

Figure 21: Nickel air concentrations in a 1x1 km² resolution grid covering 27.5% of populated area and 94% of population in Sweden. There are no exceedances of the EU air quality value of 20 ng/m³ or the 1:100,000 cancer risk value of 20 ng/m³. Highest national emission source categories are combustion in manufacturing industries, energy production and other mobile sources and machinery

In rural areas the measured nickel annual mean concentration is 1.74 ng/m³ (Table 8), and the modelled rural annual mean concentration from national sources is 0.04 ng/m³. In central Stockholm (urban background) the measured annual mean concentration is 2.5 ng/m³ (Table 8), and the modelled urban background concentration from national sources is 0.4 ng/m³. To derive the total air concentration from modelling the national sources a calculated background concentration (Eq. 1) of 1.7 ng/m³ is added. The regional background contributions are significant in rural (98%) and urban sites (76%). The contribution to nickel in ambient air (mean value for Sweden) from residential wood combustion is 0.1%.

There are modelled high values around some point sources. The positioning of sources and receptor points may not be exactly overlapping in the 1x1 km² grid and the height of sources may not be exact, which introduces errors in the UBM results for point sources. Therefore these modelled concentrations are assigned with >20 ng/m³ in the map.

Figure 22: PCDD/F (dioxins) air concentrations in a 1x1 km² resolution grid covering 27.5% of populated area and 94% of population in Sweden. No EU air quality value is given for dioxins. Highest national emission source categories are energy production, non-industrial combustion, combustion in manufacturing industries and point sources

Similar Swedish annual mean dioxin concentrations of 0.02 pg I-TEQ/m³ at rural and urban background sites as in Denmark are assumed. With modelled air concentrations from national sources of 0.0002 and 0.008 pg I-TEQ/m³ at rural and urban background (Stockholm) sites, respectively, a calculated background concentration (Eq. 1) of 0.02 pg I-TEQ/m³ is added. The regional background contributions are predominant both in rural (99%) and urban (79%) sites. The contribution to dioxin in ambient air (mean value for Sweden) from residential wood combustion is 0.4%.

There are modelled high values around some point sources. The positioning of sources and receptor points may not be exactly overlapping in the 1x1 km² grid and the height of sources may not be exact, which introduces errors in the UBM results for point sources. Therefore these modelled concentrations are assigned with >0.04 pg/m³ in the map.

Figure 23: SO₂ air concentrations in a 1x1 km² resolution grid covering 27.5% of populated area and 94% of population in Sweden. There is no exceedance of the US air quality value of 78 µg/m³. Highest national emission source categories are point sources, other sources and sinks and energy production

The calculated annual mean SO₂ concentrations in this project from national sources only and all sources including regional are 1.7 ±4.4 µg/m³ and 2.4 ±4.5 µg/m³, respectively. National sources contribute an average of 51% to the total SO₂ concentration in the populated (>20 persons) part of Sweden. The contribution to SO₂/SO_x in ambient air (mean value for Sweden) from residential wood combustion is 0.7%.

There are a few high modelled values around point sources that are assigned with >78 µg/m³ in the map. The positioning of sources and receptor points may not be exactly overlapping in the 1x1 km² grid and the height of sources may not be exact, which introduces errors in the concentration estimates for point sources.

Figure 24: NO_x air concentrations in a 1x1 km² resolution grid covering 27.5% of populated area and 94% of population in Sweden. No EU air quality value is given for NO_x. Highest national emission source categories are other sources and sinks and road and other mobile sources

The calculated annual mean NO_x concentrations in this project from national sources only and all sources including regional are 4.1 ±5.5 µg/m³ and 6.5 ±6.1 µg/m³, respectively. National sources contribute an average of 54% to the total NO_x concentration in the populated (>20 persons) part of Sweden. The contribution to NO_x in ambient air (mean value for Sweden) from residential wood combustion is 1.7%.

There are modelled high values around some point sources. The positioning of sources and receptor points may not be exactly overlapping in the 1x1 km² grid and the height of sources may not be exact, which introduces errors in the UBM results for point sources. Therefore these modelled concentrations are assigned with >40 µg/m³ in the map.

In urban air at various sites across the world the B(a)P concentration is in the range of 0.3–2 ng/m³, which constitutes approximately 1.4% of the total PAH concentrations of 20–150 ng/m³ (Hoff and Chan, 1987; Broman *et al.*, 1991; Coleman *et al.*, 1997; EC, 2001; US-EPA, 2010). B(a)P is the most monitored PAH species, due to its relatively high abundance and toxicity, and in the absence of measured PAH-4 concentrations the assessment will be made for B(a)P.

In the Swedish emission inventory no single PAH species are represented, instead PAH-4 comprising all four PAHs is used. For sources where PAH-4 is estimated, usually B(a)P is estimated separately but not always the other three species, due to missing information. As a consequence national totals for the 4 specified PAHs species in relation to PAH-4 may be misleading (Swe IIR, 2015). There is therefore no possibility to derive the B(a)P or PAH-4 concentrations or spatial distribution based on the reported national emissions. Instead estimates based on measurements in rural sites, representing regional sources, and in villages with residential wood burning are assessed.

Highest national emission source categories are non-industrial combustion plants, i.e. residential wood combustion where emissions from wood boilers, stoves and other small-scale burning of wood approximately account for two-thirds of the Swedish PAH emissions. Studies of health effects of PAHs in air have been done for various sites in Sweden (e.g. Boström *et al.*, 2002). Villages are representing high PAH concentration areas and in Andersson *et al.* (2015) it is stated that annual mean regional background concentrations for B(a)P vary from 0.1 ng/m³ in the south (Skåne) to 0.05 ng/m³ in the north (Västerbotten). The influence from residential wood combustion is smaller in the southern parts. The B(a)P mean value in the period (November 2012 – October

2013) was 0.20 ng/m³ in Vännäsby and 0.14 ng/m³ in Sävar, both villages with significant residential wood combustion.

In Table 9 the measured and modelled annual mean rural and urban background (Hornsgatan in central Stockholm) air concentrations in Sweden are compiled for each pollutant. Contribution from Swedish and regional sources and national residential wood combustion to ambient air concentration, and the most significant sources are stated. % populated area and population with exceedance of EU air quality values and 1:100,000 cancer risk values for carcinogens are also shown.

Long-range transport is significant to the ambient Swedish air for all pollutants. NO_x, SO₂, cadmium and lead have significant contributions from national sources, especially in urban sites, due to primarily road traffic, residential wood combustion and energy production and industrial point sources. National sources are more influential in urban air compared to rural air, and the contribution from residential wood combustion to the total ambient national mean air concentration varies between 0.1% (nickel) and 7% (cadmium). It is not possible to evaluate the influence of national vs. regional contribution for PAHs, due to missing national emission data for PAH-4 or specific species.

5.3 Risk towards the Swedish population

All the figures are calculated for the part of Sweden where there are more than 20 persons living per 1x1 km², which covers 93.9% of the total population and 27.5% of the populated area.

When the highest modelled concentrations around point sources have been omitted, due to modelling uncertainties for PM_{2.5}, PM₁₀, NO_x, SO₂, arsenic and nickel, all investigated pollutants comply with their respective EU air quality values and 1:100,000 cancer risk values.

From the available emission data there is no calculated 1:100,000 cancer risk from residential wood combustion in Sweden. A probable reason being, that the PAHs, which are the most critical pollutants for this source, are not included in the Swedish data.

Table 9: Rural, urban and regional background air concentrations in Sweden. Contribution from Swedish sources to ambient air concentration modelled with UBM. Most significant sources are stated in descending order. Contribution from residential wood combustion. % populated area with > 20 persons pr grid (94% of population), and population with exceedance of EU air quality values and 1:100,000 cancer risk for carcinogens (italic)

Sweden	PM _{2.5} µg/m ³	PM ₁₀ µg/m ³	As ng/m ³	Cd ng/m ³	Cr(total) ng/m ³	Pb ng/m ³	Hg ng/m ³	Ni ng/m ³	NMVOC µg/m ³	PCDD/F pg/m ³	SO ₂ µg/m ³	NO _x µg/m ³	B(a)P ng/m ³
<i>Rural air conc</i>											Total SE	Total SE	
SE sources	0.55±0.70	0.94±1.2	0.002	0.004	0.008	0.09	0.001	0.04	2.4	0.0002	1.7±4.4	4.1±5.5	–
Total (REG and SE)	6.9±2.3 ¹⁾ 5–12 ²⁾	8.2±3.0 ¹⁾ 6–18 ²⁾	0.13 ²⁾	0.043 ²⁾	0.5 ²⁾	1.2 ²⁾	1.5 ¹⁾	1.7 ²⁾	–	0.02 ²⁾	2.4±4.5 ¹⁾	6.5±6.1 ¹⁾	0.05–0.1 ²⁾
<i>Urban background air conc</i>											Total SE	Total SE	
SE sources	0.55±0.70	0.94±1.2	0.10	0.08	0.35	2.4	0.07	0.4	38	0.008	1.7±4.4	4.1±5.5	–
Total (REG and SE)	6.9±2.3 ¹⁾ 7–13 ²⁾	8.2±3.0 ¹⁾ 7–25 ²⁾	0.88 ²⁾	0.21 ²⁾	2.7 ²⁾	6.3 ²⁾	1.6 ²⁾	2.5 ²⁾	–	0.02 ²⁾	2.4±4.5 ¹⁾	6.5±6.1 ¹⁾	0.17 ²⁾
Regional background air conc	6.3±2.1 ¹⁾	7.3±2.5 ¹⁾	0.12	0.04	0.49	1.1	1.5	1.7	–	0.02	0.70±0.23 ¹⁾	2.4±1.3 ¹⁾	–
<i>Contrib of SE sources to air conc</i>													
Rural air	8% (mean SE)	11% (mean SE)	1.4%	9%	1.5%	8%	0.1%	2.4%	–	1.0%	51% (mean SE)	54% (mean SE)	–
Urban air			49%	60%	47%	60%	5%	24%	–	21%			–
Contrib of SE wood comb to air conc (country mean)	2%	1.6%	0.4%	7%	0.7%	1.3%	0.04%	0.1%	5% (of SE sources)	0.4%	0.7%	1.7%	–
Most significant sources (in descending order)	REG SE road SE non-ind comb SE point sourc SE energy prod	REG SE road SE non-ind comb SE point sourc SE energy prod SE agriculture	REG SE point sourc SE energy prod SE comb man ind	REG SE energy prod SE non-ind comb SE point sourc	REG SE point sourc SE energy prod SE comb man ind Cr(VI): NTV	REG SE road SE energy prod SE point sourc SE comb man ind	REG SE energy prod SE point sourc	REG SE comb man ind SE energy prod SE other mob sourc	Only SE sources: SE solvent use SE road SE other mob sourc SE point sourc	REG SE energy prod SE non-ind comb SE comb man ind SE point sourc	SE point sourc SE oth sourc and sinks SE road and oth mob sourc REG	SE non-ind comb	SE non-ind comb
% of populated area > EU qual Population > EU qual (% of total population)	NE	NE	NE	NE	NE	NE	NE	NE	NTV	NTV	NE	NTV	–
% of populated area > 1:100,000 Population > 1:100,000 (% of total population)	NTV	NTV	NE	NE	Cr(VI): NE	NTV	NTV	NE	NTV	NTV	NTV	NTV	–

Note:

¹⁾ Modelled with DEHM and UBM, including national and regional sources.

²⁾ Measurements at rural and urban background sites.

Regional background concentrations are based on measurements and modelling of rural air concentrations, except for particles, SO₂ and NO_x where they are calculated for entire populated area.

NTV: No threshold value (TV).

NE: No exceedance of TV.

–: No value.

Sources:

REG: Regional sources (long-range transport), non-ind comb: non-industrial combustion, comb man ind: combustion in manufacturing industries, oth mob source: other mobile sources.

6. Conclusions and recommendations

Seventeen pollutants (particles, heavy metals, inorganic gases and organic compounds) are for the first time analyzed in a screening of the carcinogenic risk at very high resolution and large scale in ambient air in the Nordic countries. Modelled 2010 annual mean air concentrations show no exceedances of their respective EU air quality values. With respect to US-EPA 1:100,000 cancer risk concentrations the only exceedance occurs for the PAH B(a)P (0.12 ng/m³, US-EPA IRIS, 2015) in Denmark for approximately 80% of the Danish population. However, the EU target value threshold of 1 ng/m³ for B(a)P is not exceeded in any parts of Denmark. No emission data for this type of analysis for B(a)P are available for the other countries and important uncertainties are still related to the Danish emission estimates.

Long-range transport is significant to the ambient air for all pollutants except the PAHs, which are mainly associated with residential wood combustion. NO_x, SO₂, Cd, Cr and Pb also have significant contributions from national sources; up to approximately 60% for NO_x and SO₂, and for the metals up to approximately 60% in urban areas and up to 20% in rural areas. High national contributions occur especially in urban air, due to primarily road traffic, residential wood combustion and energy production and industrial point sources.

1. It is recommended to perform modelling at high resolution for longer time trends including most recent years for the metals and PAHs, which are significant in the estimation of the cancer risk from long term human exposure.
2. Furthermore to perform more extensive monitoring and higher resolution modelling in urban and rural areas for the PAHs and Cr(VI) to establish more precisely the influence from residential wood burning and to quantify concentration peaks on local scale.
3. The speciation of Cr(VI) and Cr(III) in the ambient air as a function of adjacent sources and season needs to be understood more fully.

4. The additivity of quantitative risk measures for carcinogens also needs to be investigated as the combined risk from exposure of many carcinogens exceeds risks from single pollutants and has the potential to exceed risk limit values.
5. The background concentrations of metals need to be quantified more precisely as long-range transport is significant.
6. To obtain a more complete picture of the human exposure and to increase the accuracy of the risk estimates, generating high resolution emission data and model air concentrations for all the reported pollutants in each country is recommended.

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Sammenfatning

Luftforurening er forbundet med mange alvorlige helbredsmæssige effekter blandt befolkningen. I de senere år har der været fokus på partikulær luftforurening (Davidson *et al.*, 2005), men der er imidlertid også betydelige negative helbredseffekter fra forurenende stoffer der optræder på gasform (Maynard *et al.*, 2005). Påvirkning af flere skadelige stoffer på én gang kan have en synergistisk effekt og det er derfor vigtigt at tage højde for alle, eller i det mindste de mest signifikante stoffer.

På denne baggrund, er et vigtigt mål med dette studie at kvantificere og præsentere den mest fuldstændige kortlægning af luftforureningen af kræftfremkaldende stoffer, baseret på de emissionsestimater der er tilgængelige i de nordiske lande, samt at evaluere forureningsniveauet med hensyn til EU's luftkvalitetskrav og 1:100,000 tærskelværdier for kræftfremkaldende stoffer.

Metoder

Danske, finske, norske og svenske emissionsdata fra 2010 fra alle nationale kilder samt den langtransporterede forurening fra regionale kilder er analyseret for de kræftfremkaldende stoffer: arsenik (As), cadmium (Cd), chrom(VI) (Cr(VI)), bly (Pb), kviksølv (Hg), nikkel (Ni), dioxin (PCDD/F) og poly aromatiske hydrokarboner (PAH'er): benzo(a)pyrene (B(a)P), benzo(b)fluoranthene (B(b)F), benzo(k)fluoranthene (B(k)F) og indeno(cd)pyrene (I(cd)P), og for co-carcinogenerne: svovldioxid (SO₂), nitrogen oxider (NO_x), fine partikler (PM_{2.5}) og grove partikler (PM₁₀). Datakilderne er de nationale årlige emissionsrapporteringer til United Nations Economic Commission for Europe (UNECE), Convention on Long Range Transboundary Air Pollution (CLRTAP), samt forskellige videnskabelige studier.

Kravet til at stoffer og kilder kan inkluderes i dette studie er, at emissionerne skal være tilgængelige i et 1x1 km² net, dækkende hele landet. Dette indebærer at der er inkluderet 14 stoffer (11 kræftfremkaldende, tre co-carcinogener) for Danmark, fire stoffer (co-carcinogener) for Finland og 11 stoffer (syv kræftfremkaldende, fire co-

carcinogener) for Sverige. Norge har i øjeblikket ingen data tilgængelige til denne type analyse. For at reducere tiden til de omfattende modelberegninger, er der kun medregnet arealer med mere end 20 indbyggere bosat per kvadratkilometer i Sverige og Finland. For Sverige betyder det, at 93.9% af indbyggertallet på 9,528,000, svarende til 27.5% af det befolkede areal er inkluderet. For Finland er 90.5% af den totale befolkning på 5,334,000 bosat på 19.8% af det befolkede areal inkluderet.

Årsmiddelluftkoncentrationer i et 1x1 km² net for PM_{2.5}, PM₁₀, SO₂ and NO_x er fundet ved at modellere nationale kilder samt langtransporteret forurening (baggrund) fra omkringliggende lande. Årsmiddelluftkoncentrationer for As, Cd, Cr(VI), Hg, Ni, Pb, PCDD/F, B(a)P, B(b)F, B(k)F og I(cd)P er fundet ved at modellere nationale kilder og ved at tage højde for langtransporteret forurening, ved at addere en baggrunds koncentration. Denne beregnes som differencen mellem en eller flere målte koncentrationer i landområder og modellerede koncentrationer fra nationale kilder de samme steder. Baggrundskoncentrationerne anvendes i hele landet.

Ved at anvende årsmiddelluftkoncentrationer for et enkelt år kan luftforureningsniveauet og -fordelingen vurderes til et givet tidspunkt, hvilket vil indikere kritiske steder i landet, samt de pågældende kilder (f.eks. brændeovne) og forurenende stoffer (f.eks. B(a)P).

Risiko for overskridelse af EU's luftkvalitetskrav beregnes ved Margin of Exposure (MoE); og for de kræftfremkaldende stoffer beregnes også den øgede risiko for at udvikle kræft, som følge af en livstidseksponering af det pågældende stof. Sidstnævnte risiko udtrykkes som 0.00001 til 1 (eller 1:100,000, dvs. en til ethundrede tusind) for at udvikle kræft.

Herudover beregnes det befolkede areal og antallet af personer der er eksponeret for koncentrationer der overstiger risiko grænseværdierne, samt bidrag fra de nationale kilder og fra langtransport (baggrund). Alle resultater kan ses i de respektive landes kapitler og i de opsummerende tabeller; tabel 4 (Danmark), tabel 6 (Finland) og tabel 9 (Sverige).

Resultater

De beregnede årsmiddelkoncentrationer for kræftfremkaldende stoffer og co-carcinogener overholder deres respektive EU-luftkvalitetskrav samt 1:100,000 tærskelværdier for kræftfremkaldende stoffer i alle lande. Den eneste undtagelse er B(a)P i Danmark, der har en årsmiddelkoncentration

der overstiger 1:100,000-tærskelværdien på 0.12 ng/m³ (US-EPA IRIS, 2015) for omtrent 80% af den danske befolkning, svarende til ca. 60% af det befolkede areal. Bemærk, at EU's luftkvalitetskrav på 1 ng/m³ for B(a)P er overholdt i hele Danmark. Brændeovne udgør den væsentligste kilde og bidrager med ca. 70% af B(a)P luftkoncentrationen i Danmark. Langtransport fra omkringliggende lande udgør den næstvæsentligste kilde til B(a)P i luften i Danmark.

Bidraget fra brændeovne til luftkoncentrationerne i Sverige varierer mellem 0.1% for Ni og 7% for Cd. I dette projekt er der ikke observeret risiko for de kræftfremkaldende stoffer fra brændeovne i Sverige og Finland, hvilket sandsynligvis skyldes at PAH'erne, som udgør den mest kritiske gruppe af stoffer, ikke er inkluderet i de svenske eller finske data.

Langtransport er en væsentlig kilde til luftkoncentrationerne i alle lande og for alle de betragtede stoffer på nær B(a)P, som hovedsagelig kommer fra brændeovne i de respektive lande. NO_x, SO₂, Cd, Cr og Pb har væsentlige bidrag fra nationale kilder, specielt i byområder, hvilket primært skyldes vejtrafik, brændeovne, energiproduktion og industrielle punktkilder.

Usikkerheder

Der er flere usikkerheder knyttet til de beregnede luftkoncentrationer og risikoværdier. Den største usikkerhed er relateret til emissions-estimerterne, specielt med hensyn til den rummelige fordeling af brændeovne og deres emissionsfaktorer. Dette indebærer, at de beregnede koncentrationer i byområder i Danmark er overestimerede på grund af upræcis placering af brændeovne, se kapitel 2.1 for nærmere forklaring. En forbedret fordeling er under udarbejdelse, men den er endnu ikke tilgængelig for 2010-emissionerne, som anvendes i dette projekt. Det vides ikke hvorledes den forbedrede placering vil influere de beregnede luftkoncentrationer i Danmark. Sandsynligvis vil værdierne reduceres i byerne og stige i landområderne.

De beregnede luftkoncentrationer repræsenterer årsmiddelværdier, hvorimod forurenende stoffer fra specielt brændeovne udviser et mønster der følger årstiderne. Befolkningen er derfor ikke påvirket af den samme koncentration hele året, men snarere højere koncentrationer om vinteren og lavere om sommeren.

Emissionsdata for de kræftfremkaldende stoffer (metaller, PAH'er og dioxin) for den langtransporterede (baggrunds-)forurening udenfor landenes grænser, er meget begrænsede. I dette projekt er baggrunds-

koncentrationerne tilføjet med et konstant bidrag over hele landet. Imidlertid er metallerne, med undtagelse af gasformig kviksølv, typisk forbundet med de grove og/eller fine partikler (Malandrino *et al.*, 2016; Allen *et al.*, 2001; WHO, 2007), for hvilke der er karakteristiske koncentrationsgradienter hen over landene. Dette vil overestimere baggrundskoncentrationerne i visse dele og underestimere dem i andre.

Den anvendte model til beregning af luftkoncentrationerne inkluderer kun O₃-NO_x kemi, mens andre luftforureningskomponenter håndteres som inerte stoffer, hvilket vil give yderligere usikkerheder for reaktive eller flygtige stoffer som VOC'er og PAH'er.

Kun primærpartikler er inkluderet i modelleringen af spredningen i by-baggrund; disse antages at være mest relevante i forbindelse med kræftisiko.

De anvendte modeller er valideret for by-baggrund og landområder for stofferne NO_x, PM₁₀ og PM_{2.5}, og viser bias i årsmiddel-koncentrationerne på typisk 10–35%. På grund af manglende målinger for nogle af de øvrige stoffer, kunne usikkerhederne ikke kvantificeres i detaljer i dette studie, og det er derfor ikke muligt at give sandsynlighedsintervaller for modellerede koncentrationer og risiko-estimer.

Risk of Air Pollution in Relation to Cancer in the Nordic Countries

Seventeen pollutants (particles, heavy metals, inorganic gases and organic compounds) are for the first time analyzed in a screening of the carcinogenic risk at very high resolution and large scale in ambient air in the Nordic countries. Modelled 2010 annual mean air concentrations show no exceedances of the EU air quality values. The only exceedance of US-EPA 1:100,000 cancer risk concentrations occurs for the PAH BaP in Denmark. However, the EU target value threshold for BaP is not exceeded. No emission data for BaP are available for the other countries and important uncertainties are still related to the Danish emissions. Long-range transport is significant except for BaP that originates mostly from residential wood combustion. It is recommended to monitor the influence from residential wood combustion more extensively, and to analyze longer time trends for long-term human exposure.

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