# The influence of residential wood combustion on the concentrations of $PM_{2.5}$ in four Nordic cities

Kukkonen Jaakko[1], Susana López-Aparicio[2], David Segersson[3], Camilla Geels[4], Leena Kangas[1], Mari Kauhaniemi[1], Androniki Maragkidou[1], Anne Jensen[4], Timo Assmuth[5], Ari Karppinen[1], Mikhail Sofiev[1], Heidi Hellén[1], Kari Riikonen[1], Juha Nikmo[1], Anu Kousa[6], Jarkko V. Niemi[6], Niko Karvosenoja[5], Gabriela Sousa Santos[2], Ingrid Sundvor[7], Ulas Im[4], Jesper H. Christensen[4], Ole-Kenneth Nielsen[4], Marlene S. Plejdrup[4], Jacob Klenø Nøjgaard[4], Gunnar Omstedt[3], Camilla Andersson[3], Bertil Forsberg[8], Jørgen Brandt[4]

[1] Finnish Meteorological Institute, Erik Palmenin aukio 1, P.O. Box 503, 00101, Helsinki, Finland
[2] Norwegian Institute for Air Research, Instituttveien 18, P.O. Box 100, 2027 Kjeller, Norway
[3] Swedish Meteorological and Hydrological Institute, SE-60176 Norrköping, Sweden
[4] Aarhus University, Frederiksborgvej 399, 4000 Roskilde, Denmark
[5] Finnish Environment Institute, Latokartanonkaari 11, FI-00790 Helsinki
[6] Helsinki Region Environmental Services Authority, Ilmalantori 1, FI-00240 Helsinki
[7] Institute of Transport Economics, Gaustadalléen 21, 0349 Oslo, Norway
[8] Umeå University, 901 87 Umeå, Sweden

Correspondence: Jaakko Kukkonen (jaakko.kukkonen@fmi.fi)

**Abstract.** Residential wood combustion (RWC) is an important contributor to air quality in numerous regions worldwide. This study is the first extensive evaluation of the influence of RWC on ambient air quality in several Nordic cities. We have analyzed the emissions and concentrations of $PM_{2.5}$ in cities within four Nordic countries: in the metropolitan areas of Copenhagen, Oslo and Helsinki, and in the city of Umeå. We have evaluated the emissions for the relevant urban source categories and modelled atmospheric dispersion on regional and urban scales. The emission inventories for RWC were based on local surveys, the amount of wood combusted, combustion technologies and other relevant factors. The accuracy of the predicted concentrations was evaluated based on urban concentration measurements. The predicted annual average concentrations ranged spatially from 4 to 7 µg/m$^3$ (2011), from 6 to 10 µg/m$^3$ (2013), from 4 to more than 13 µg/m$^3$ (2013) and from 9 to more than 13 µg/m$^3$ (2014), in Umeå, Helsinki, Oslo and Copenhagen, respectively. The higher concentrations in Copenhagen were mainly caused by the relatively higher regionally and continentally transported background contributions. The annual average fractions of $PM_{2.5}$ concentrations attributed to RWC within the considered urban regions ranged spatially from 0 to 15 %, from 0 to 20 %, from 8 to 22 % and from 0 to 60 % in Helsinki, Copenhagen, Umeå and Oslo, respectively. In particular, the contributions of RWC in central Oslo were larger than 40 % as annual averages. In Oslo, wood combustion was used mainly for the heating of larger blocks of flats. On the contrary, in Helsinki, RWC was solely used in smaller detached houses. In Copenhagen and Helsinki, the highest fractions occurred outside the city centre in the suburban areas. In Umeå, the highest fractions occurred both in the city centre and its surroundings.

# 1. Introduction

The combustion of wood or other kinds of biomass for residential heating and cooking is a significant source of atmospheric pollution, both in developed and developing countries (e.g., Patel et al., 2013; Sigsgaard et al., 2015; Butt et al., 2016). Biomass combustion and the combustion of residential solid fuels (RSF), such as wood crop residues, animal waste, coal and charcoal (Butt et al., 2016; Capistrano et al., 2017) have been found to contribute significantly to particulate matter emissions in numerous countries worldwide (e.g., Karagulian et al., 2015; Butt et al., 2016; Vicente and Alves, 2018; Im et al., 2019). In addition, such a combustion results in emissions of harmful or toxic gaseous pollutants, such as CO, CO2, NOx, heavy metals (i.e. Pb, Cu, Fe, Zn, and Hg, etc), polycyclic aromatic hydrocarbons (PAHs) and other toxic compounds (Patel et al., 2013; Capistrano et al., 2017).

Epidemiological studies have documented that both short- and long-term exposure to smoke from biomass and RSF combustion are responsible for chronic obstructive pulmonary disease (COPD), acute lower respiratory and cardiovascular disease, pneumonia, tuberculosis, asthma, and even lung cancer (Patel et al., 2013; Sigsgaard et al., 2015; Capistrano et al., 2017). Several studies have pointed out the strong relationship between particulate matter from biomass burning and severe consequences in health, including hospitalizations, cardiovascular, respiratory and premature mortality (McGowan et al., 2002; Pope III and Dockery, 2006; Sanhueza et al., 2009; Brook et al., 2010). According to WHO (2011; 2014), approximately 4 million deaths were attributed to RSF combustion every year worldwide. Butt et al. (2016) evaluated that the global annual excess adult premature mortality attributed to residential emissions was 308 000. In Europe and North America, 29 000 premature deaths have been estimated to be ascribed annually to residential biomass burning (Chafe et al., 2015).

For simplicity, we mainly use in this article the term residential wood combustion (RWC), which includes the combustion of various wood products. The concept of RWC refers here to either detached residential houses, row (terraced) houses, or moderately-sized blocks of flats. The term ‘small-scale combustion’ (SSC) has also been used in the literature to refer to combustion in stationary small-scale appliances. Such appliances can be used, e.g., at homes, in small and medium-scale industry and in heat and energy production. However, this definition does not include small-scale combustion in traffic. Clearly, the concept SSC is more comprehensive, and includes more fuels and sources compared with RWC.

With respect to RWC globally, Vicente and Alves (2018) evaluated residential fuel burning to be responsible for substantial shares of particulate matter concentrations in Africa (34 %), in Central and Eastern Europe (32 %), Northwestern Europe (22 %), the Southern China region (21 %), South Eastern Asia (19 %), and India (16 %). According to Karagulian et al.’s (2015) review, 25 % of urban ambient air pollution from $PM_{2.5}$ was attributed to traffic, 15% to industrial activities, 20 % to domestic fuel burning, 22% to unspecified anthropogenic sources, and 18% to natural dust and salt. Regarding Northwestern, Western, Central and Eastern, and Southwestern Europe, they reported that domestic wood burning was responsible for 22 %, 15 %, 32 % and 12 % of the concentrations, respectively. In another study conducted by Butt et al. (2016), their computations showed that the largest residential emissions of $PM_{2.5}$ occurred in East and South Asia, and Eastern Europe.

Regarding RWC findings in Europe, Brandt et al. (2013), based on emissions for 2000 and the Economic Valuation of Air pollution (EVA) system, estimated that non-industrial combustion (dominated by RWC) contributed to approximately 10 % of the total health costs due to air pollution in Europe. Two studies for major cities in the UK indicated that the contributions of RWC to particulate matter were clearly lower than those observed for Nordic cities and part of the cities in continental Europe (Fuller et al., 2014; Harrison et al., 2012). Fuller et al. (2014) reported that 9% of ambient $PM_{10}$ in London in 2010 was attributed to RWC. Harrison et al. (2012) reported RWC contributions, which were below 1% of ambient $PM_{2.5}$ concentrations in London and Birmingham. Cordell et al. (2016) evaluated the impacts of biomass burning in the UK, the Netherlands, Belgium and France. Their findings indicated that the contribution of biomass combustion to $PM_{10}$ concentrations during the winter ranged from 2.7 % to 11.6 %. Lanz et al. (2010) reported that wood burning emissions accounted for 17-49 % of organic aerosol in winter across the greater Alpine region during 2002–2009. Yttri et al. (2019) analyzed the carbonaceous particle fraction at nine European locations during winter, spring and autumn. The contribution of RWC was substantial, accounting for 30-50% of the total carbon in particles at most sites.

There are also several publications on RWC in Nordic countries. Im et al. (2019) evaluated that the largest domestic emission sector of $PM_{2.5}$ in Denmark, Finland and Norway was non-industrial combustion. Non-industrial combustion and industry in Sweden were found to contribute to $PM_{2.5}$ emissions with a comparable amount. Im et al. (2019) also estimated that the total premature mortality cases due to air pollution were approximately 4000 in Denmark and Sweden, and approximately 2000 in Finland and Norway. Markers of processes and abundant sources of particles were apportioned based on measurements during a summer campaign at four Norwegian rural background sites in 2009 by Yttri et al. (2011). In late summer, biomass burning contributed only by 3-7 % to the carbonaceous aerosol. According to Hedberg et al. (2006), RWC was responsible for 70 % of the fine particle mass in a small city in Northern Sweden in 2002. In addition, Glasius et al. (2006) reported that $PM_{2.5}$ concentrations in a small Danish rural village were approximately 4 $\mu g/m^3$ higher than at a nearby background monitoring site during the winter period. Their findings regarding the observation of high $PM_{2.5}$ concentrations during the evening and the night were consistent with a local heating source. In a later study, RWC was analyzed in a similar village and season in the same region (Glasius et al., 2008). The local contribution of RWC to $PM_{2.5}$ corresponded to 10% of ambient $PM_{2.5}$.

Moreover, Saarnio et al. (2012) reported that the average contributions of RWC to ambient $PM_{2.5}$ concentrations in the Helsinki Metropolitan Area (HMA) ranged from 18% to 29% at two urban sites and from 31% to 66% at two suburban sites during various periods within the colder half of the year. Local wood combustion sources were reported to be responsible especially for the increased concentrations at suburban sites. Hellén et al. (2017) observed that the local emissions from residential wood combustion caused high benzo(a)pyrene (BaP) and levoglucosan concentrations in the HMA. The BaP concentrations exceeded the European Union target value for the annual average concentrations (1ng $m^{-3}$) in certain suburban detached-house areas.

Some studies have also addressed specifically particulate carbonaceous matter from wood burning (Genberg et al., 2011; Yttri et al., 2011; Szidat et al., 2009; Helin et al., 2018; Aurela et al., 2015).

The overarching aim of this article is to evaluate the influence of RWC within urban regions on air quality in four Nordic cities, in particular, Copenhagen, Helsinki, Oslo and Umeå. The more specific objectives include, first, to present and inter-compare the methodologies for evaluating the emissions and dispersion of fine particulate matter originated from RWC in four Nordic cities. Second, we aim to compare the predicted concentrations with the available air quality measurements. Third, we intend to present and analyze numerical results on the $PM_{2.5}$ concentrations. In particular, we will quantify the influence of RWC in urban regions on the $PM_{2.5}$ concentrations. We will also report and evaluate the current regulations regarding the emissions and concentrations from RWC. This article presents for the first time a systematic assessment of the influences of RWC on air quality in several Nordic cities.

# 2. Methods

This study focuses on three Nordic capital regions: Oslo, Helsinki and Copenhagen, and one smaller city including its neighboring area, Umeå. Our aim was to investigate greater capital or urban areas, instead of solely the areas of the cities. For instance, we address the Helsinki Metropolitan Area which contains four separate cities. However, for simplicity, we chose to refer in the following to the capital regions simply as Oslo, Helsinki and Copenhagen.

Umeå was selected instead of the Swedish capital, due to lack of detailed information on the influence of RWC in Stockholm. This article presents the results for one year for each city. The target years are 2011 for Umeå, 2013 for Helsinki and Oslo, and 2014 for Copenhagen.

We have addressed the contributions of RWC originated from sources within the target urban regions. Clearly, a fraction of the regional background is also originated from RWC that is located outside the considered urban regions.

### 2.1 The considered cities, regions and measurement networks

The locations of the selected cities and the domains are presented in Fig. 1. The considered domain sizes were selected mainly based on the sizes of the cities and their surrounding metropolitan areas; the domain is therefore largest for Copenhagen and smallest for Umeå.

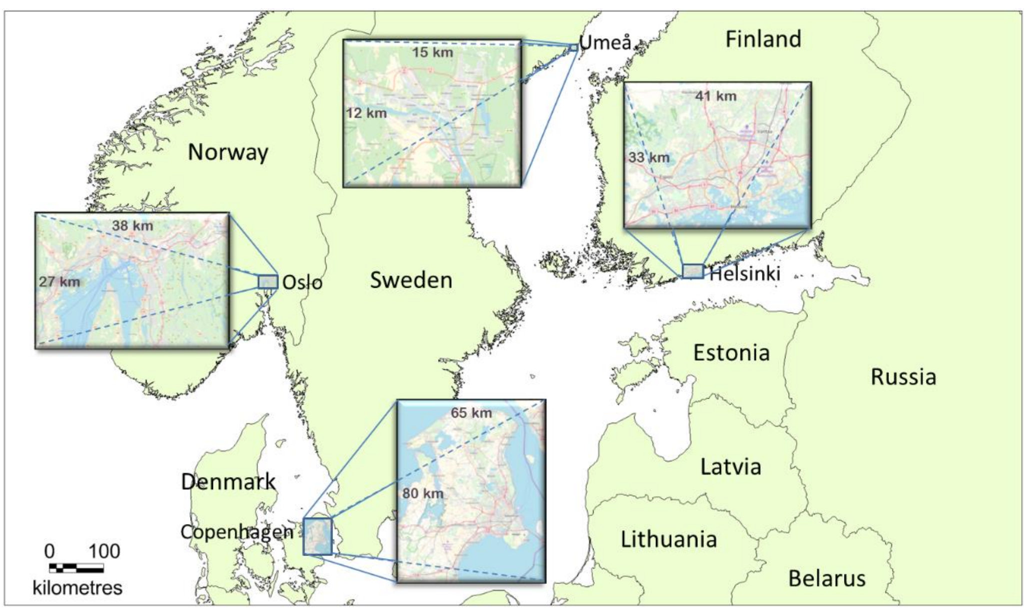
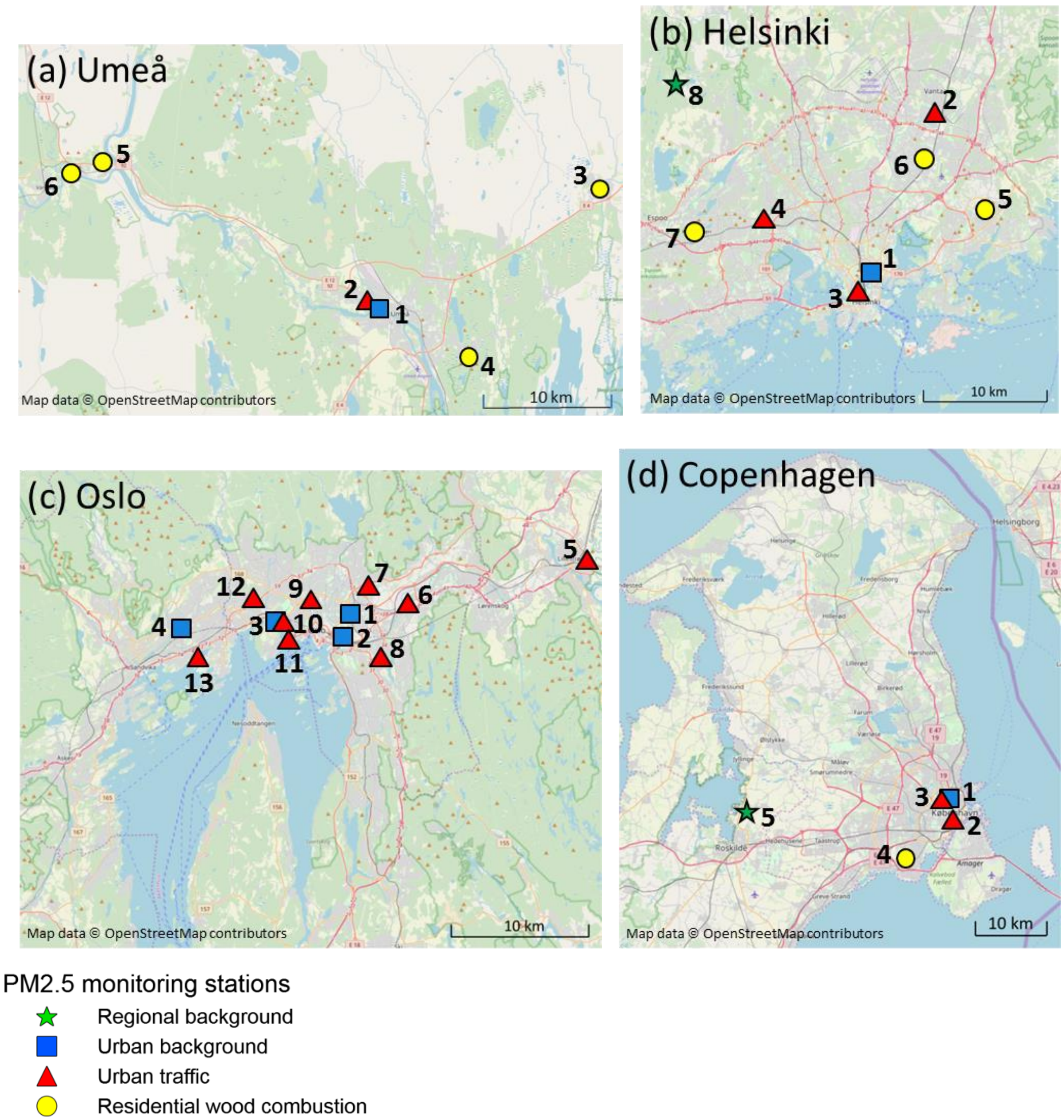


Fig.1. The locations of the selected cities and domains. The physical sizes of the domains have been indicated in the inserted smaller maps.

The geographical locations and the air quality measurement stations addressed in this study are presented in Figs. 2a-d. All the considered cities are located either on the coast or in the immediate vicinity of the coast of the Baltic Sea. Characterizations of the geographical regions and climates of the cities have been presented in Appendix A.




Figs. 2a-d. The geographical locations of the cities and the air quality measurement stations for (a) Umeå, (b) Helsinki, (c) Oslo and (d) Copenhagen. The panels represent the locations of the stations in 2011 for Umeå, 2013 for Helsinki and Oslo, and 2014 for Copenhagen, respectively. The most densely populated central areas of the cities are shown with light mauve color. Notation for the stations: (a) Umeå: 1 Biblioteket; 2 Västra Esplanaden; 3 Sävar; 4 Tavleliden; 5 Vännäsby; 6 Vännäs. (b) Helsinki: 1 Kallio; 2 Tikkurila; 3 Mannerheimintie; 4 Leppävaara; 5 Vartiokylä; 6 Tapanila; 7 Kauniainen; 8 Luukki. (c) Oslo: 1 Sofienbergparken; 2 Grønland; 3 Skøyen; 4 Bekkestua; 5 Vigernes; 6 Alnabru; 7 Rv4, Aker sykehus; 8 Manglerud; 9 Kirkeveien; 10 Bygdøy Alle; 11 Hjortnes; 12 Smestad; 13 Eilif Dues vei. (d)

Copenhagen: 1 HCØ; 2 HCAB; 3 JGTV; 4 Hvidovre; 5 Risø. © OpenStreetMap contributors 2019. Distributed under a Creative Commons BY-SA License.

### 2.1.1 Concentration measurement networks

**Concentration measurements for Umeå**

For Umeå, we took in consideration both long-term measurements and the results of a measurement campaign. The long-term measurements were conducted from 2006 to 2011 at two sites in the city of Umeå (Västra Esplanaden and Biblioteket). The site of Västra Esplanaden is classified as an urban traffic site; it is a roadside station located in a street canyon with relatively dense traffic. The site of Biblioteket is classified as an urban background site; it is located on a rooftop in central Umeå. The long-term measurements were conducted using TEOM 1400A (Thermo Fisher Scientific, Waltham, MA, USA).

A monitoring campaign was also carried out to evaluate the performance of the modelling approach (Omstedt et al. 2014). The measurements were carried out in the villages of Sävar, Vännäs and Vännäsby, situated in the vicinity of Umeå, and at Tavleliden, located in the southernmost outskirts of the city. The stations of Sävar, Vännäs, Vännäsby and Tavleliden are classified as residential sites.

All monitoring campaign measurements of $PM_{2.5}$ were carried out using filter collection. For Sävar and Vännäsby, the filters were changed on a daily basis and for Tavleliden and Vännäs, on weekly intervals. The analysis of the filters was gravimetric (weighting before and after measurements under standardized conditions).

**Concentration measurements for Helsinki**

For this study, we have selected three measurement stations that mainly represent the influence of RWC in residential areas (Vartiokylä, Tapanila and Kauniainen) and three stations that represent either pollution originated from vehicular traffic in the centre of Helsinki (Mannerheimintie) or at smaller regional urban centres within the Helsinki Metropolitan Area (Leppävaara and Tikkurila). In addition, we have selected two stations that represent urban (Kallio2) and regional background (Luukki). All the $PM_{2.5}$ monitors were equivalent reference instruments (i.e., TEOM 1400AB, SHARP 5030, FH 62 I-R and Grimm 180).

**Concentration measurements for Oslo**

All the available monitoring stations in Oslo in 2013 were classified as either urban or suburban traffic, or urban background. There were no stations originally designed to measure the influence of residential combustion; however, several stations were influenced by pollution from RWC.

At all the considered monitoring stations in Oslo, $PM_{2.5}$ is measured by continuous monitors and logged with a time resolution of 1 hour. All monitors are equivalent reference instruments (i.e., TEOM 1400A, TEOM1405DF-FDMS and Grimm-EDM180).

**Concentration measurements for Copenhagen**

The Danish Air Quality Monitoring Network includes five measuring sites in close vicinity of Copenhagen. There are three sites in central Copenhagen: two street sites and one urban background site. We have also used data measured at a suburban site of Hvidovre, located outside of Copenhagen, and at a regional background site in a rural area at Risø. The $PM_{2.5}$ observations were performed using the Low Volume Sampling reference method.

**Inter-comparison of the measurement networks in the target cities**

Generally, the locations of the stations in the target cities have been selected using similar or the same criteria (according to the European Union directives and guidance). For each target city, we have selected regional and urban background stations, and urban traffic and RWC stations. Stations representing all of these categories were available for all the cities.

However, in case of Oslo, the official categorization of the stations did not include any RWC station. We have therefore selected a few urban stations in Oslo, which we considered to be best representative for the pollution attributed to RWC, to stand for RWC in this study.

## 2.2 Emission inventories for the target cities

The assessment of emissions located within the target cities is addressed in this section. The regional and continental scale emissions are discussed in the context of regional dispersion modelling. We present first an overview and summary of the emission modelling both for RWC and for all the other urban sources. More detailed descriptions of the assessment of RWC emissions are presented in the following section.

### 2.2.1 Overview of the emission inventories

An overview of the emission inventories regarding RWC is presented in Table 1. In all the cities, the emissions inventory from RWC was based on (i) surveys regarding the amounts and use of wood stoves, boilers and other relevant appliances, (ii) national or literature-based emission factors, and (iii) the spatial distribution of the emissions. In case of Umeå, Helsinki and Copenhagen, also various national or local register data been used.

Table 1. Assessment of the emissions of $PM_{2.5}$ originated from RWC, and their spatial resolution in the target cities.

| | Umeå | Helsinki | Oslo | Copenhagen |
|---|---|---|---|---|
| **Data and information sources ragarding the use of wood for combustion, and on the combusion appliances** | (i) Survey on the amounts of wood stoves and boilers, and the habits of wood combustion.<br>(ii) Register data gathered by chimney sweepers. | (i) Survey concerning the amount of wood combusted, types and amounts of fireplaces, habits of wood combustion, for detached and semi-detached houses.<br>(ii) Regional basic register for dwellings | (i) Survey regarding the amount and temporal variability of wood combusted by Statistics Norway. | (i) Survey on the unit consumption and age for different types of residences.<br>(ii) Register data on the location of the appliances from the chimney sweepers.<br>(iii) Danish energy statistics and building and dwelling register.<br>(iv) The spatial distribution is evaluated by the SPREAD model (Plejdrup et al., 2016) |
| **Assessment of emission factors** | Combination of results from national measurement programmes and available literature (Omstedt et al., 2014) | Combination of results from national measurement programmes and available literature. (Kaski et al., 2016, Savolahti et al., 2016) | National measurements reported by Haakonsen and Kvingedal (2001). | Combination of results from the EMEP/EEA Guidebook (EMEP/EEA, 2016) and national measurements. |
| **Spatial resolution of the predicted emissions of $PM_{2.5}$** | Appliances were treated as point sources. | 100 x 100 $m^2$ | 1 x 1 $km^2$ | 1 x 1 $km^2$ |
| **Basis for spatial allocation of emissions, i.e., gridding** | Geocoded addresses of combustion appliances, based on a survey and chimney sweeper register | Average wood use for houses with different primary heating methods. Location of the houses from local building and dwelling register. | The amount of wood consumed in the districts in Oslo, based on a survey carried out by Statistics Norway. | Average wood consumption in different types of houses and location of the appliances based on chimney sweepers register. Location of the houses from Danish building and dwelling register. |
| **Basis for temporal allocation of emissions** | Measured local contributions of the concentrations of $PM_{2.5}$ as a proxy variable | Information gathered in questionnaires (Kaski et al., 2016) | Based on a survey carried out by Statistics Norway | Temporal profile evaluated by Friedrich and Reis (2004) |

Information on the combusted wood is subsequently combined with the corresponding emission factors. The assessment of emission factors has been based on either on national measurements (Oslo), or a combination of national measurements and results from the available literature (Umeå, Helsinki and Copenhagen). All measurements that were used for the assessment of emission factors were based on methodologies using cooled flue gases and dilution chambers.

Clearly, the RWC emissions are dependent on the temporal variation of the meteorological conditions, especially on the ambient temperature. In case of Oslo, the variation of emissions on the ambient temperature has also been taken into account, based on measured weekly average ambient temperatures.

In the inventory for Umeå, the individual RWC sources were treated separately. For the other cities, the computed RWC emissions have been gridded on various spatial resolutions, from 100 x 100 $m^2$ (Helsinki) to 1 x 1 $km^2$ (Oslo and Copenhagen).

An overview of the emission inventories for the other relevant source categories is presented in Table 2. The vehicular traffic exhaust emissions have been included for all the cities. The suspension emissions originated from vehicular traffic have been included for Umeå, Helsinki and Oslo. The emissions from shipping have been included for Umeå, Oslo and Copenhagen. In case of Helsinki, Kukkonen et al. (2018) presented a detailed analysis regarding the contribution of shipping on the $PM_{2.5}$ concentrations, based on computations for a three-year period. They found that the contribution of shipping, including harbour activities, to the ambient air $PM_{2.5}$ concentrations varied from 10 to 20 % near major harbours to a negligible contribution in most other parts of the metropolitan area.

Table 2. Assessment of the traffic flows and emissions from vehicular traffic and other source categories, except for RWC, in the target cities.

| | | **Umeå** | **Helsinki** | **Oslo** | **Copenhagen** |
|---|---|---|---|---|---|
| **Vehicular traffic flows and emissions** | **Vehicular traffic flows** | Traffic flow model EMME/2 and measured data | Traffic flow model EMME/2 and measured data | Traffic flow model RTM23+ | National GIS-based road network and traffic database. The spatial distribution is done by the SPREAD model |
| | **Vehicular exhaust emissions** | Emission factors by Hausberger et al. (2009) | The LIPASTO emission model | NILUs traffic emission model | Danish area: the SPREAD emission model |
| | **Vehicular suspension emissions** | Resuspension model by Omstedt (2005) | The FORE traffic suspension emission model (Kauhaniemi et al., 2011) | The NORTRIP traffic suspension emission model (Denby et al., 2013) | Not included |
| **Shipping emissions** | | Modelled using SHIPAIR (Segersson, 2014) | Not included in the modelling | Based on López-Aparicio et al. (2017b) and US EPA (2009) | An updated version of AIS based inventory for Denmark (Olesen et al., 2009). |
| **Other sources** | | National compilation of emissions originated from off-road machinery and major point sources in Sweden. | Not included in the modelling | Industrial emissions and emissions from off-road mobile combustion | Fugitive emissions from fuels, emissions from industrial processes, agriculture and waste modelled by SPREAD |

However, the emission inventories for other source categories except for RWC were not the main focus of this article. Their more detailed descriptions have therefore been presented in Appendix B.

### 2.2.2 Detailed descriptions of the assessment of emissions from RWC

For the estimation of the emissions of wood combustion, one needs to know numerous factors, including (i) the spatial distributions of the various categories of buildings using wood combustion, (ii) the amounts and distribution of firewood used, (iii) the shares of primary and secondary heating sources, (iv) the amounts of wood used and the numbers of boilers, stoves, fireplaces, sauna stoves and other heating devices, and (v) the emission factors for the different types of heating devices (Kukkonen et al., 2018).

The information on the use of wood and the heating device technologies is mostly based on surveys. Moreover, in case the survey year and the study year are not the same, the information on the changes of technologies and fuels in time is also needed. There are also other factors that may have a substantial influence on the assessment of RWC emissions, which are commonly estimated in a simplified manner, or even neglected in evaluating the emissions of RWC (e.g., Savolahti et al., 2016). These include (i) the compositions of wood fuels, e.g., their humidity, the tree species and the pre-processing and storage of wood, and (ii) the variations of the habits and procedures of combustion (Kukkonen et al., 2018). For these reasons, the uncertainties in the RWC emission estimates of $PM_{2.5}$ are commonly relatively higher than those for most other major emission source sectors (e.g., Karvosenoja et al., 2018).

**The assessment of emissions from RWC for Umeå**

A survey regarding the habits of wood consumption and combustion was carried out in four areas in 2013, which included a recently constructed suburb and three small towns. The survey included also an air quality monitoring campaign. Based on the register data gathered by the chimney sweepers, we selected a representative sample of 178 houses with a stove or a boiler. A total of 176 houses were willing to participate to the survey; these households were subsequently visited. The residents were interviewed using a form with questions mainly regarding the type of stove or boiler, the principal type of heating, biofuel consumption, biofuel type, combustion habits and the actions to reduce energy consumption.

A bottom-up inventory was made on the amounts of wood stoves and boilers, based (i) on the above-mentioned survey on the habits of wood consumption and combustion, and (ii) register data that had been gathered by the local chimney sweepers. In combining these two information sources, we have extended the information of the above-mentioned survey to the whole building stock, i.e., we have assumed that the habits of wood consumption and combustion are the same also in the households that were not included in the survey.

The inventory was compiled in the Västerbotten county in 2009. This dataset included information on the types of equipment, such as boilers (wood or oil), stoves, pellets boiler and open fireplaces, and their geocoded addresses. A total of more than 54 thousand appliances were identified within the county. About 23 % of them were wood boilers, 10 % pellet boilers, 64 % stoves and 3 % oil boilers.

We estimated the amounts of combusted wood and the emission factors based on dilution chamber experiments by Omstedt et al. (2014). Separate emission factors were used for (i) wood, (ii) pellet and (iii) oil fueled boilers, (iv) fireplaces and stoves, and (v) summer houses and cottages.

The temporal variations of the emissions originated from wood combustion were evaluated using the measured local contributions of the concentrations of $PM_{2.5}$ as a proxy variable. The local contributions of the $PM_{2.5}$ concentrations were estimated by subtracting the modelled regional background concentration from the local measurements. All measurement stations used for these estimations were located in areas with a substantial amount of RWC.

**The assessment of emissions from RWC for Helsinki**

Emissions from RWC were based on an emission inventory for the years 2013-2014, including the spatial and temporal variation of emissions. We estimated by using a questionnaire the amount of wood combusted in 12 different fireplace types, and the procedures and habits for the combustion. Its results were applied for all detached and semi-detached houses in the area.

The spatial distribution of the emissions was based on average wood use per combustion appliance type for each main heating method of a house, based on the questionnaires (Kaski et al., 2016). The emissions were allocated to the location of the houses available in local building and dwelling register, and the emissions were allocated to the 100 x 100 $m^2$ grid.

The temporal variation (monthly, weekly, hourly) of emissions was estimated based on the information gathered in questionnaires (Kaski et al., 2016). The temporal variation was estimated separately for three different source categories: heating boilers, sauna stoves, and other fireplaces. However, the information was not sufficient to model quantitatively the influence of meteorological variables on the emissions.

The emission factors for different types of fireplaces were adopted based on the results of national measurement programmes and the literature (Kaski et al., 2016; Savolahti et al., 2016). The spatial distribution of RWC emissions was based on the regional basic register for dwellings, provided by the Helsinki Region Environmental Services Authority; this register contains information on primary heating methods.

**The assessment of emissions from RWC for Oslo**

The RWC emissions were estimated based on a bottom-up approach by using the data of a dedicated survey. The survey was carried out by Statistics Norway; its aim was to assess the use of wood combustion and heating habits in Oslo. The results of the survey include information on the amount of wood consumed in the districts in Oslo, and information on how the wood combustion varies temporally, in terms of weeks, days and hours of the day. Information on the amount of wood combusted was collected based on the survey in terms of the type of technology, i.e., open fireplace, wood stove produced before 1998 and wood stove produced after 1998.

The emission factors were extracted from Haakonsen and Kvingedal (2001), which were based on a review of the results from different tests for various fireplaces in Norway. Separate emission factors were used for conventional wood stoves, certified wood stoves and open fireplaces.

The seasonal variations of emissions were taken into account, by modelling their variation using their dependency on the ambient temperature, based on observed weekly average ambient temperatures. The weekly mean temperatures measured at the station of Blindern in 2013 were used in the parametrization.

**The assessment of emissions from RWC for Copenhagen**

A survey was conducted regarding the unit consumption of wood and age of different types of residences by the Danish Technological Institute in 2015. A distinction was made between villas, apartments and allotments, either connected or unconnected to district heating. The survey also included information on the age of the appliance, distributed into four age categories. For RWC in the Copenhagen area, detailed data was also used on the location of the appliances based on the chimney sweeper register data for Copenhagen in 2015.

The assessment of the emissions for the Danish area were based on the SPREAD model. The SPREAD model is an integrated database system for high-resolution (1 km x 1 km) spatial distribution of emissions (Plejdrup et al., 2016 and 2018). The SPREAD model includes emission distributions for each sector in the Danish emission inventory system. In this study, the emission factors included in this national inventory were used (Nielsen et al., 2017). These were based on emission factors of the EMEP/EEA Guidebook (EMEP/EEA, 2016) and national measurements.

The emission inventory for RWC was also based on wood consumption information by the Danish energy statistics. The spatial distribution of RWC emissions was based on the Danish building and dwelling register, which includes information on building use, and on primary and secondary heating installations.

### 2.2.3 Inter-comparison of the emission inventories in the target cities

For all target cities, we have included the most important emission source categories. The emissions from vehicular traffic exhausts and RWC have been included for all the cities, and the suspension emissions originated from vehicular traffic for all cities except for Copenhagen. In case of Copenhagen, traffic suspension emissions have only a minor importance, mainly due to the fact that studded tyres are not used, in contrast with the other target cities. The emissions from shipping have been included for all cities except for Helsinki, as the contribution of shipping has previously been found to have a relatively minor influence on the concentrations of $PM_{2.5}$ (Kukkonen et al., 2018). In summary, we can evaluate that these omissions in the emission inventories will result only in minor uncertainties to the final results of this study.

Based on previous studies, the uncertainties related to the estimation of RWC emissions were expected to be relatively larger, compared with those for the other included source categories. However, detailed high-resolution emission inventories of RWC were available for all target cities. The emission inventories for RWC were based on similar, although not identical, methodologies in the target cities. The inventories were in all cities based on surveys regarding the amounts and use of relevant appliances, national or literature-based emission factors, and the evaluations of the spatial distribution of emissions.

## 2.3 Atmospheric dispersion modelling for the target cities

We present first an overview and summary of the dispersion modelling, and second, a more detailed description of dispersion modelling in the target cities.

### 2.3.1 Overview of dispersion modelling

An overview of the dispersion modelling has been presented in Table 3. The assessment of the regional background concentrations was based on chemical transport modelling in all the cities, except for Umeå, for which the assessment of the regional background was based on a combination of measured data and the results of regional background modelling. For the urban scale assessments, multiple-source Gaussian modelling systems were used for all the cities. As the focus on this study was on RWC, the dispersion in street canyons was modelled only for one street canyon measurement station in Umeå. The spatial resolutions of the modelling of the dispersion originated from RWC ranged from a couple or a few tens of metres (Oslo, Umeå) to 100 m (Helsinki) and 1 km (Copenhagen).

Chemical reactions were included in the regional scale computations, for all the cities. However, chemical reactions and aerosol transformation processes were not included in the urban scale computations. However, it has previously been shown that gas-to-particle transformation reactions do not have a major influence on the annual average $PM_{2.5}$ concentrations in Nordic cities on urban distance scales (Kukkonen et al., 2016; Karl et al., 2016). The impacts of aerosol processes (such as nucleation, condensation and evaporation, and coagulation) on the annually averaged $PM_{2.5}$ concentrations have been found to be minor, although these can be significant in specific dispersion conditions and for the finer aerosol modes (Karl et al., 2016; Pohjola et al., 2007).

Table 3: Atmospheric dispersion modelling and its spatial resolution in the target cities.

| | | **Umeå** | **Helsinki** | **Oslo** | **Copenhagen** |
|---|---|---|---|---|---|
| **Assessment of regional background concentrations** | | Measured values at a regional background station | Predictions of the regional and global scale chemical transport model SILAM | Predictions of model ensemble, using seven regional-scale chemical transport models | Predictions of the hemispheric chemical transport model DEHM |
| **Urban scale dispersion modelling** | **Residential wood combustion** | Multiple-source Gaussian model DISPERSION | Multiple-source Gaussian model UDM-FMI | Multiple-source Eulerian model EPISODE | The Gaussian plume-in grid model - Urban Background Model (UBM) |
| | **Vehicular traffic for the whole city** | Multiple-source Gaussian model DISPERSION | Roadside dispersion model CAR-FMI | Multiple-source Eulerian model EPISODE, including sub-grid Gaussian line source modelling | The Gaussian plume-in grid model - Urban Background Model (UBM) |
| | **Vehicular traffic in street canyons** | Street canyon dispersion model OSPM was used. | Street canyon modelling (OSPM) is included in the modelling system, but was not used in this study. | Street canyon modelling was not included in the modelling system. | Street canyon modelling (OSPM) was included in the modelling system, but was not used in this study. |
| **Spatial resolution** | | Near the sources 50 x 50 m$^2$, at substantial distances from the sources 3 km$^2$ | Vehicular traffic: from 20 m in the vicinity of traffic sources to 500 m on the outskirts of the area. RWC: 100 x 100 m$^2$ | For the entire modelling domain 20 x 20 m$^2$ | For the entire modelling domain 1 x 1 km$^2$ |

### 2.3.2 Detailed descriptions of dispersion modelling

For each domain, we address first the assessment of the regional background concentrations, and second, the dispersion of urban contributions to concentrations.

#### Atmospheric dispersion modelling for Umeå

The regional background contribution was estimated based on the measured data from two regional background stations (Bredkälen and Vindeln) and on the modelled spatial concentration distributions. The stations of Bredkälen and Vindeln are situated approximately 350 km to the west, and 50 km north-west of Umeå, respectively. For the year 2013, to account for the influence of concentration gradients between Umeå and the station of Bredkälen, we have added a contribution of 1.28 µgm$^{-3}$ to the measured concentrations at Bredkälen, based on the computations by Omstedt et al. (2014). Similar yearly adjustments were made also for years 2006-2011, based on results from the atmospheric chemistry transport model MATCH and corrections using earlier measurements at the more nearby station Vindeln (Segersson et. al. 2017).

The larger spatial scale meteorological values were extracted from the predictions of the Swedish version of the numerical weather prediction model HIRLAM, with a horizontal resolution of 22 km. The finer, mesoscale meteorological data for dispersion modelling was provided by the operational mesoscale analysis system Mesan (Häggmark et al., 2000), which is based on an optimal interpolation technique. All available measurements from synoptic and automatic stations, radars and satellites were analyzed with hourly time resolution on an 11x11 $km^2$ grid across northern Europe. The following meteorological parameters were used: wind speed and direction at a height of 10 m, ambient temperature and humidity at a height of 2 m, cloud cover, global radiation and precipitation. Boundary layer parameters such as friction velocity, sensible heat flux and boundary layer height were calculated using methods from van Ulden and Holtslag (1985), Holtslag et al. (1995) and Zilitinkevich and Mironov (1996).

The dispersion of pollutants from RWC and vehicular traffic were modelled using the Gaussian multiple-source dispersion model DISPERSION (Omstedt, 1988). The DISPERSION model contains a Gaussian finite length line source dispersion model. For point sources, the DISPERSION model includes a revised version of the Gaussian OML (Operational Meteorological Air Quality model*)* point-source model (Omstedt et al. 2011). For a more detailed description of the model and its evaluation against experimental data, the reader is referred to Omstedt et al. (2011) and Gidhagen et al. (2013).

The dispersion parameters of the DISPERSION model are continuous functions of boundary-layer parameters, such as friction velocity, sensible heat flux and boundary layer height. The model also includes a detailed description of plume rise and building downwash effects. The OML model has previously been used to investigate the influence of wood combustion on particulate matter concentrations in residential areas in Denmark (Glasius et al., 2008) and in the northern part of Sweden (Omstedt et al., 2011). In case of sources described using spatially gridded emissions, a Gaussian model included in the Airviro air quality management system was applied (SMHI, 2017). Segersson et al. (2017) have presented a more detailed description of dispersion modelling methodology for other sources than RWC.

The chimney height for RWC was set to 5 m and the effective plume rise was then evaluated by the model depending on meteorological conditions. The concentrations were computed on a receptor grid that was different for the contributions from RWC and vehicular traffic.

The OSPM model (Operational Street Pollution Model; Berkowicz, 2000) can be used to estimate the dispersion and transformation of vehicular and urban background pollution in a street canyon. In this study, the model was used to estimate the concentrations at the considered street canyon measurement station. The OSPM model was run twice, both with and without the influence of the surrounding buildings. The difference between these two model computations is a measure for the concentration increment caused by the buildings. This concentration difference was subsequently added to the values obtained by the urban background computations.

### Atmospheric dispersion modelling for Helsinki

The regional background concentrations were computed using the SILAM model (Sofiev et al., 2006; 2015) for the European domain. A detailed description of these computations has been presented by Kukkonen et al. (2018). For this study, we selected four grid points of the SILAM computations that were closest to the Helsinki Metropolitan Area (HMA), but outside the urban domain. We then computed an hourly average of the concentration values at these four locations, and used that value as the regional background for all the chemical components of particulate matter, except for mineral dust. In case of mineral dust, we used the lowest hourly value within the four selected points. The latter procedure was adopted to avoid the potential double counting of occasional releases of dust originating from the considered urban area.

The meteorological input variables for the urban scale modelling were based on synoptic weather observations from the stations of Helsinki-Vantaa airport (18 km north of city center) and Harmaja (marine station south of Helsinki), radiation measurements of Helsinki-Vantaa, and sounding observations from Jokioinen (90 km northwest of Helsinki) for the year 2013. Measured meteorological data was analyzed using the meteorological pre-processing model of the Finnish Meteorological Institute (MPP-FMI) adapted for urban environment (Karppinen et al 2000a). The MPP-FMI model is based on the energy budget method of van Ulden and Holtslag (1985), and its output consists of hourly time series of meteorological data needed for the dispersion modelling, including temperature, wind speed, wind direction, Monin-Obukhov length, friction velocity, and boundary layer height. The same meteorological parameters were used for the whole HMA.

For urban dispersion modelling, we used a roadside dispersion model and a multiple source Gaussian model. We did not model dispersion in street canyons.

The urban scale dispersion of vehicular emissions was evaluated with the CAR-FMI model (Contaminants in the Air from a Road – Finnish Meteorological Institute; e.g., Kukkonen et al. 2001). The model is a Gaussian finite line source model, which computes an hourly time-series of the pollutant dispersion. The dispersion parameters are modelled as a function of Monin-Obukhov length, friction velocity, and boundary layer height. The modelling system containing the CAR-FMI model has been evaluated against the measured data of urban measurement networks for gaseous pollutants and particulate matter in the HMA, London and Birmingham, U.K. (e.g. Karppinen et al., 2000c; Kousa et al., 2001; Kauhaniemi et al., 2008; Aarnio et al., 2016; Sokhi et al., 2008; Singh et al., 2014; Srimath et al., 2017), and for gaseous pollutants also against the results of a field measurement campaign and other roadside dispersion models (Kukkonen et al., 2001; Ottl et al., 2001; Levitin et al., 2005).

Overall, the model performance for predicting the $PM_{2.5}$ concentrations has been fairly good or good. For instance, for the predicted and measured hourly concentrations at 18 sites in London, the medians of correlation, index of agreement and factor of two of all stations were 0.80, 0.86, and 74 %, respectively (Singh et al., 2014).

The dispersion of RWC emissions was evaluated with Urban Dispersion Model of Finnish Meteorological Institute UDM-FMI (Karppinen et al., 2000b). The model is a multiple-source Gaussian dispersion model for various stationary source categories (point, area and volume sources). The modelling system has been evaluated against measurement data of urban measurement networks (e.g., Karppinen et al., 2000b; Kousa et al., 2001).

In this study, the RWC emissions were treated as area sources of the size of 100 m x 100 m. The height of the sources was assumed to be equal to 7.5 m, including the initial plume rise. This altitude was assumed to be the combined average height of detached and semi-detached houses and chimneys in the area.

**Atmospheric dispersion modelling for Oslo**

The regional background concentrations were extracted from the ensemble reanalysis that comprised of seven regional-scale chemical transport models (Marécal et al., 2015): CHIMERE, EMEP, EURAD-IM, LOTOS-EUROS, MATCH, MOCAGE and SILAM. Within this ensemble, the models had a common framework in terms of meteorology, chemical boundary conditions and emissions. However, the models have differences in terms of their aerosol representations, chemistry schemes, physical parametrizations, and different implementations for use of the input data.

The meteorological variables used as modelling input were hourly measurements extracted from the data of the meteorological stations in the simulated domain (the stations of Valle Hovin, Blindern, Alna, Tryvannshøgda and Kjeller). All these stations are located within the Oslo municipality, except for the station of Kjeller, which is located at a distance of approximately 25 km northeast. The variables related to wind and atmospheric stability were used as input in a preprocessing diagnostic wind field model. The hourly wind field data produced by the wind field model were input to the urban scale dispersion modelling.

The atmospheric dispersion modelling was done with the EPISODE model. This model is a combined three-dimensional Eulerian and Lagrangian air pollution dispersion model, which has been developed for urban and local scale applications (Slørdal et al., 2003; 2008). The Eulerian part of the model consists of a numerical solution of the atmospheric mass conservation equation of the pollutant species in a three-dimensional grid. The Lagrangian part consists of separate subgrid-models for line and point sources. Topography has been included as input data in the regional scale modelling for Oslo domain. The topography within the domain is defined on the Eulerian grid in terms of the elevation above sea level.

The line source model is an integrated Gaussian type model, whereas the point source model is a Gaussian puff trajectory model. The EPISODE model has been used for a large number of applications, including the assessment of air quality and air pollution control measures in urban areas (e.g., Sundvor and López-Aparicio, 2014), and in a forecasting system for seven city regions in Norway.

**Atmospheric dispersion modelling for Copenhagen**

The Danish multiscale integrated model system THOR (Brandt et al., 2001; 2003) has for this study been setup for a domain over Greater Copenhagen. The system combines the Danish Eulerian Hemispheric model (DEHM) and the Urban Background Model (UBM).

The DEHM model (Christensen 1997) is a chemistry-transport model describing the concentration fields of 73 photo-chemical compounds (including NOx, SOx, VOC, NHx, CO, etc.) and nine classes of particulate matter (e.g., $PM_{2.5}$, $PM_{10}$, TSP, seasalt, and fresh and aged black carbon). The regional model covers the Northern Hemisphere, with higher resolution over Europe (50 km x 50 km), Northern Europe (16.7 km x 16.7 km) and Denmark (5.6 km x 5.6 km). The DEHM model has been extensively evaluated (Brandt et al. 2012; Zare et al. 2014; Solazzo, et al. 2012a, b).

The regional background concentrations were extracted on a 5.6 x 5.6 $km^2$ grid. The meteorological fields were provided by the Weather Research and Forecasting (WRF) Model (Skamarock et al., 2008), using the same domains as the DEHM model. The anthropogenic emissions for the regional modelling were based on a combination of a number of emission inventories including in particular the EMEP emissions for Europe (http://www.ceip.at/webdab_emepdatabase/emissions_emepmodels/). Within the Danish area, the emissions were based on the SPREAD emissions model. Temporal profiles of emissions, depending on the emission type were included.

The Urban Background Model (UBM) is a Gaussian plume model, including a simplified description of photochemical reactions of $NO_x$ and ozone. The model was set up for the selected urban domain on a resolution of 1 x 1 $km^2$ and hourly background concentrations were provided by the DEHM model. The UBM model has been used for assessments of air pollution in Denmark, e.g., as part of the Danish AirGis system (Hvidtfeldt et al., 2018; Khan et al., 2019).

### 2.3.3. Inter-comparison of the dispersion modelling in the target cities

The regional background concentrations were computed using chemical transport models for all the cities, except for Umeå, for which these were assessed based on both measured data and the results of chemical transport models. All of the applied chemical transport models for Copenhagen, Helsinki and Umeå (DEHM, SILAM and MATCH) have previously been extensively evaluated against experimental data. The regional background assessment for Oslo was based on an ensemble of seven European models. The uncertainties of the estimates on regional background are therefore not expected to have a major influence on the results and conclusions of this study.

Multiple-source Gaussian modelling systems were used for the urban scale assessments in all target cities. All of these modelling systems (DISPERSION, UDM-FMI and CAR-FMI, EPISODE and UBM) have been previously widely used and analyzed against measured data. However, the spatial resolutions of the modelling of the dispersion varied between the cities, from a couple of tens of metres (in Helsinki and Oslo) to 1 kilometre (Copenhagen). These differences of resolution have to be taken into account in the interpretation of the results.

## 2.4 Statistical model performance parameters

For simplicity, we have mainly considered two selected statistical model performance parameters: the index of agreement (IA) and the fractional bias (FB). The IA is a measure of the agreement of the measured and predicted timeseries of concentrations, and the FB is a measure of the agreement of the longer term (e.g., annual) average concentrations.

The index of agreement is defined as (Willmott, 1981)

$$IA = 1 - \frac{\sum_{i=1}^{n}(P_i - O_i)^2}{\sum_{i=1}^{n}(|P_i - \bar{O}| + |O_i - \bar{O}|)^2} \qquad (1)$$

where n is the number of data points, and P and O refer to predicted and observed pollutant concentrations, respectively. Overbar refers to an average value. Factor-of-two is defined as the fraction of data for which $0.5 \leq P/O \leq 2$.

Fractional bias is given by

$$FB = \frac{2(\bar{P} - \bar{O})}{\bar{P} + \bar{O}} \qquad (2)$$

where P and O are the mean values of the predicted and observed values.

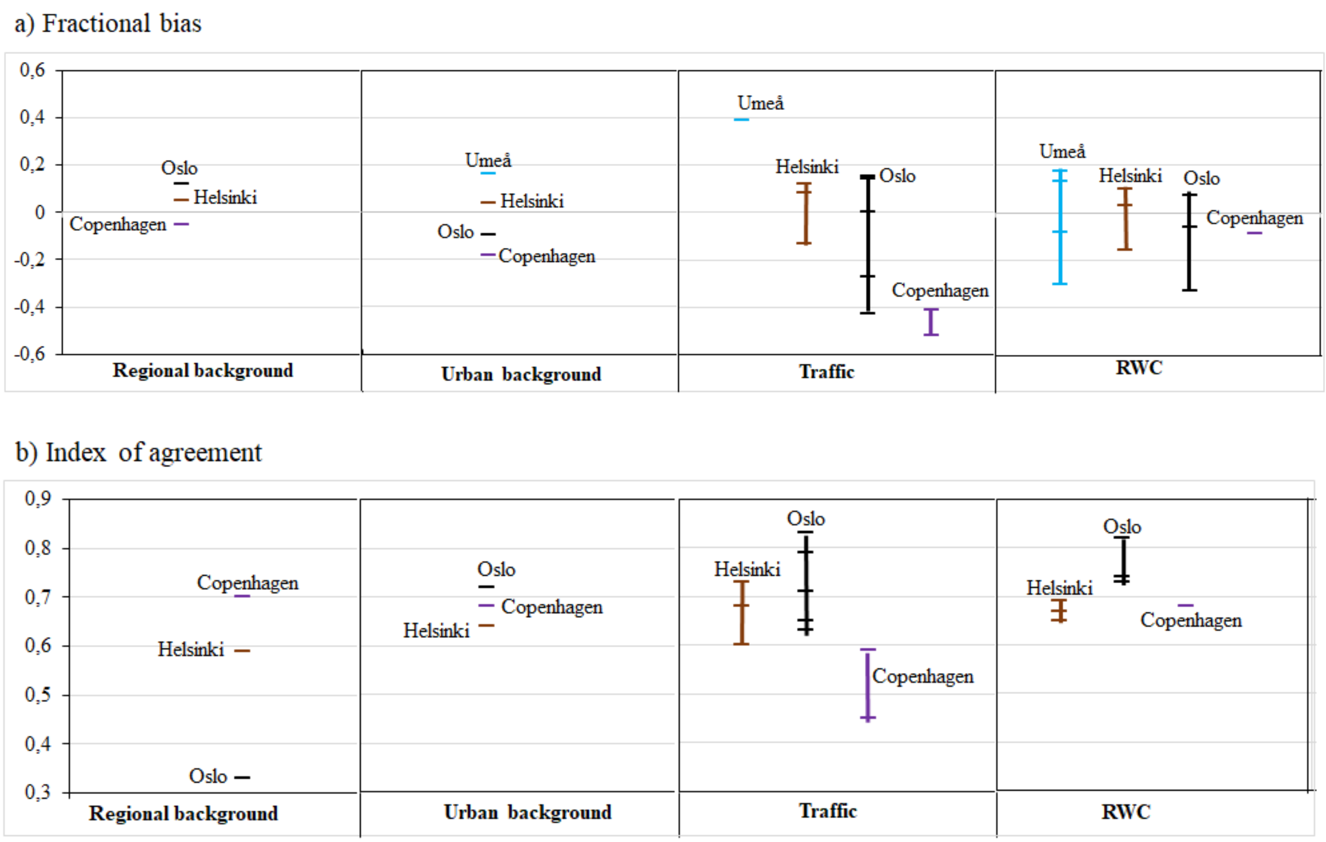
# 3. Results

First, the numerical predictions will be evaluated against measured urban scale data regarding the $PM_{2.5}$ concentrations in the four target cities. Second, the predicted emissions originated from RWC will be presented and analyzed. Third, the ambient air concentrations of $PM_{2.5}$, and the contributions from RWC to these concentrations will be presented and discussed. We have also presented an overview of the regulatory frameworks regarding RWC in four Nordic countries in Appendix E.

### 3.1 Evaluation of the predicted concentrations against measured data

The results of the model evaluation are summarised and reviewed in this section. The detailed model evaluation results have been presented in Appendix C.

The ranges of values of two statistical parameters, Index of Agreement (IA) and Fractional Bias (FB), for the daily average concentration values of $PM_{2.5}$ values are presented in Figs. 3a-b. The IA is a measure of the agreement of the measured and predicted time series of concentrations, whereas FB is a measure of the agreement of the average (annual or during several months) values of the concentrations. In case of regional and urban background stations, we have selected one station for each city, whereas for traffic and RWC stations, the range of values is shown by a vertical line, and the value for each station is shown by short horizontal lines.




Figs. 3a-b. Values of two statistical model performance measures for the target cities, for various categories of stations. Upper panel presents the fractional biases, the lower panel the index of agreement. In case of Oslo, we have selected three stations to be representative for RWC (Akerbergveien, Bygdoy Alle and Kirkeveien), although these were officially classified as traffic monitoring stations.

In case of Umeå, the distributions of the temporal variations of the emissions originated from wood combustion were evaluated using the measured concentrations of $PM_{2.5}$. It was therefore not reasonable to perform an evaluation of the temporal variation of the predicted values for Umeå; this would have implied evaluating modelling that is partly based on experimental values to the same experimental values. The IA values have therefore not been presented for that city.

In case of Oslo, there were no measurement stations that would have been officially nominated by the local authorities as measuring the influence of RWC. We have therefore selected three stations that we considered to be most influenced by RWC.

The results in Figs. 3a-b facilitate an assessment of model performance in terms of the cities and the categories of the stations. The FB values are reasonably good, considered here as the range from -0.20 to + 0.20, for all the regional and urban background values, and for most of the traffic and RWC stations. However, for some of the traffic and RWC sites, the FB values are substantial, especially for two traffic stations in Copenhagen (substantial underprediction of the model), one traffic station in Umeå (over-prediction), and two traffic and one RWC station in Oslo (underprediction). In case of the stations in

Copenhagen, the under-prediction is to be expected, as we have applied an urban background model on a spatial resolution of 1 x 1 km$^2$.

The IA values are also fairly good, considered here as IA > 0.55, in most cases. The agreement of the time series of daily measured and modelled values is relatively worse for the regional background values in Oslo, and for one traffic station in Copenhagen. In particular, the IA values for the traffic stations are lower for Copenhagen, compared with the corresponding values in Helsinki and Oslo. This is due to the coarser spatial model resolution (1 x 1 km$^2$) in Copenhagen, compared with those in the other three target cities, which tends to result in an underprediction of the local influence of vehicular traffic. A better model performance was obtained in a previous study for the street stations in Copenhagen, when the street pollution model OSPM was used (Khan et al., 2019). For the finer resolution computations for Helsinki and Oslo, there is no substantial systematic difference between the model performance at traffic stations compared with the corresponding RWC stations.

The measured and predicted annual average concentrations have been summarised in Fig. 4. Both the measured and predicted concentration values are the highest for Copenhagen, caused mainly by the relatively higher regional background contributions, compared with the other three cities. The concentrations are second highest for Oslo, mainly due to substantial urban contributions. In case of the computations for Denmark, the predicted regional background has been evaluated at the station of RISO; however, this station is not optimally representative for the regional scale background of Copenhagen.

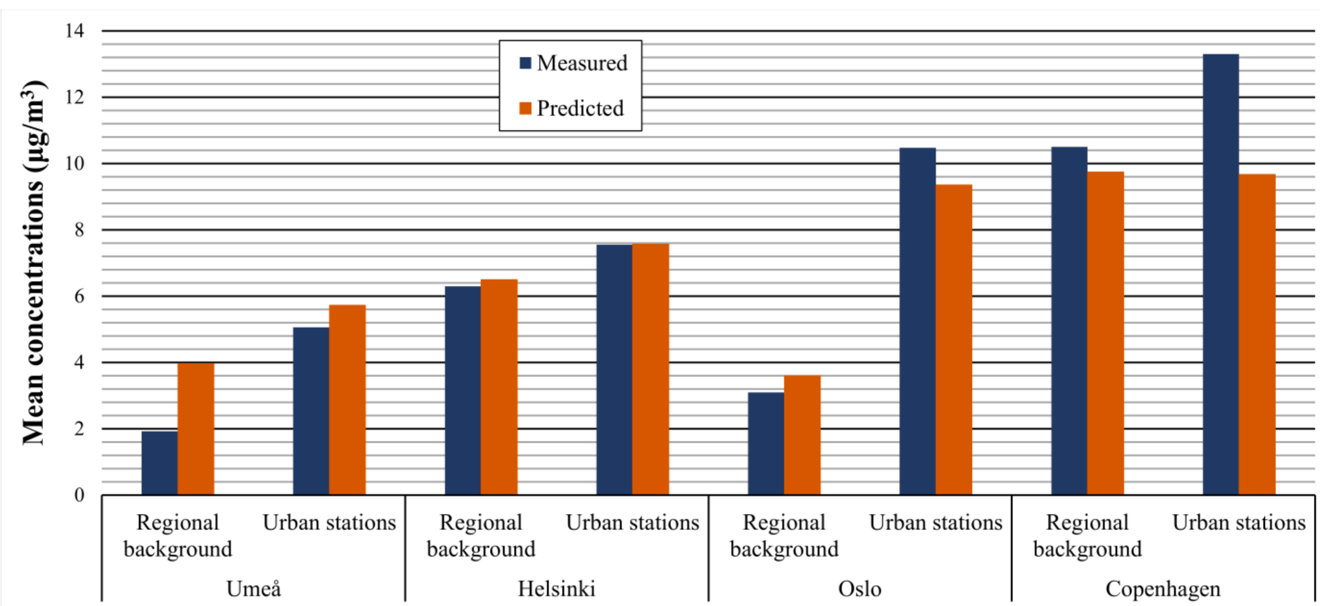
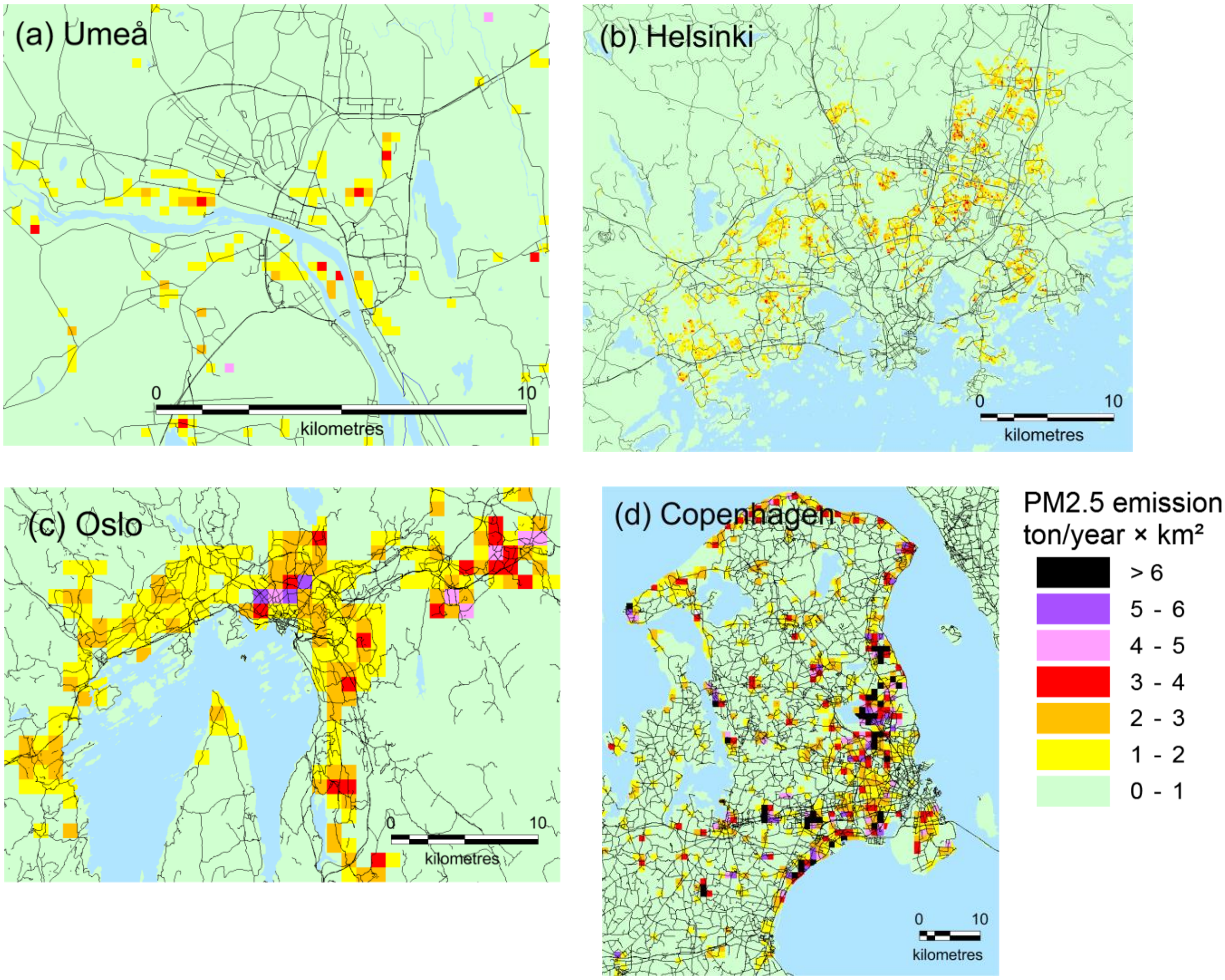



Fig. 4. The measured and predicted annual average concentrations of $PM_{2.5}$ in the target cities. Both the predicted and measured values at the urban stations in the target cities are averages over all the considered urban measurement stations in each city.

### 3.2 Emissions of $PM_{2.5}$ originated from RWC

The results of the emission inventories regarding RWC for $PM_{2.5}$ have been presented in Figs. 5a-d.




Figs. 5a-d. The predicted emissions of $PM_{2.5}$ originated from RWC in Umeå (a), Helsinki (b), Oslo (c) and Copenhagen (d). The spatial resolution is 250 x 250 $m^2$ for Umeå, 100 x 100 $m^2$ for Helsinki, and 1 x 1 $km^2$ for Oslo and Copenhagen. The unit is ton/(year $km^2$) for all the domains. The sea and inland water areas have been presented using light blue color. The results represent the year 2011 for Umeå, 2013 for Helsinki and Oslo, and 2014 for Copenhagen. The physical scales of the domains have also been indicated. © OpenStreetMap contributors 2019. Distributed under a Creative Commons BY-SA License.

The results show that the emission values originated from RWC were the highest for the domains of Copenhagen and Oslo; these range from negligible to more than 5.0 or 6.0 ton/(year $km^2$) in some limited areas in Oslo and Copenhagen, respectively. The emission values within the domains of Helsinki and Umeå reach up to a few ton/(year $km^2$).

In the case of Helsinki and Copenhagen, the highest emission values of RWC were mainly located outside the city centers. In particular, in the Helsinki region, the highest emissions were detected in detached and semi-detached house areas; these were situated to the west, east and north of the centre of Helsinki. The detailed locations of these areas were reported by Hellén et al. (2017). For Copenhagen, the highest emission strengths were also slightly outside the most densely built city centre; highest concentrations were observed in the suburban areas of Copenhagen.

In the Helsinki area, the buildings are mainly kept warm using an extensive district heating system, and by electricity heating and by using geothermal heat pumps. However, these systems have only a minor impact on the local air quality. The district heating is mainly produced in energy plants burning fossil fuels; most of these plants have very high stacks. On the other hand, wood combustion is mainly used as a secondary heating system in detached or semi-detached houses. In addition, it is common to use fireplaces and sauna stoves in suburban detached houses. Wood combustion appliances were used in approximately 90 % of the detached houses in the Helsinki area in 2013. Helsinki was the only target city, in which sauna stoves were an important source of $PM_{2.5}$ emissions. There is a high correlation of the spatial density of the detached or semi-detached houses, and that of the emissions from RWC in the Helsinki region.

Domestic heating in the Copenhagen area is dominated by district heating, which was used in 80 % of the residences on a national level at the time. Wood combustion was most commonly used as a secondary heating method in wood stoves in residential detached or semi-detached houses as in Helsinki. Such detached houses are mainly located in suburban regions, outside the city centre. Wood is mainly residentially combusted in the Copenhagen area in wood stoves, instead of boilers.

In addition to suburban regions, there is a significant number of wood stoves used in apartments in the city centre of Copenhagen. The stoves in these apartments have on average a lower rate of wood consumption compared to the ones in detached and semi-detached houses. Wood stoves can also be located in the cottages in allotments. The emission gridding methodology used in this study has taken into account both the differences of the rate of consumption for the different building types, and those for the RWC used as primary and secondary heating.

For Oslo and Umeå, the highest emission values from RWC were located within the city centres. Concerning Oslo, the highest $PM_{2.5}$ emissions were attributed to residential areas, which contain aged blocks of flats and multifamily dwellings, both located in the Oslo city centre and its surroundings. A major fraction of these buildings was constructed in the beginning of the 20$^{th}$ century, and wood stoves are commonly used for heating. There were also relatively high emission densities in the densely inhabited eastern parts of Oslo and in the neighbouring municipalities to the east of Oslo.

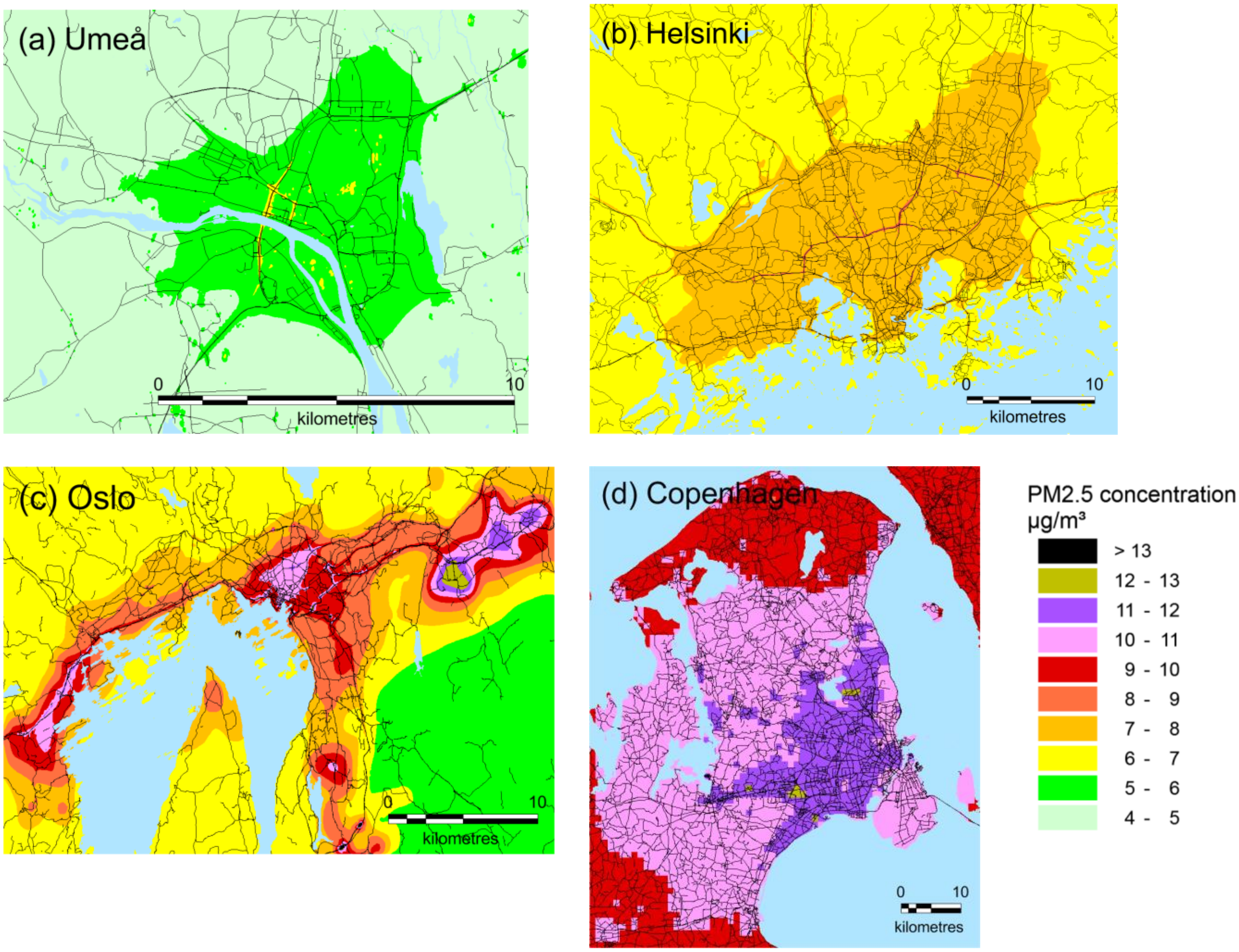
In Umeå, the largest emissions were originated from relatively old buildings, which were not connected to the district heating system. In such buildings, wood boilers are commonly used as a primary heating source. Umeå was the only target city, in which boilers were widely used inside the urban area. Although there was a smaller number of this kind of buildings within the centre of the city, a majority of them were detached or semi-detached houses located outside of the city of Umeå. In residential areas connected to the district heating system, stoves are commonly used as a secondary heating source. The wood consumption for such stoves is considerably lower than that for boilers. However, the number of stoves is substantially higher; the stoves were therefore the main source of RWC emissions within the most recently constructed residential areas.

### 3.3 $PM_{2.5}$ concentrations and source contributions from RWC

The urban scale concentration distributions are determined by the corresponding spatial and temporal distributions of the urban emissions and the meteorological conditions. The contributions from RWC are strongly influenced by the district heating systems, the spatial distributions of residential areas, and the types of usage of the combustion devices. The concentration distributions from RWC can be correlated with the corresponding distributions of residential areas. The residential areas are often situated mainly in suburban regions; however, there can be also a substantial number of residents in the city centres or in regional urban centres.

Clearly, the dilution of pollution is dependent on the meteorological conditions during any selected year. In addition, the amount of wood combustion is influenced by the evolution of the ambient temperatures, especially during the colder winter periods. The strengths of other urban pollution sources and of the regional background are essential factors in terms of the source contributions to RWC.

The predicted $PM_{2.5}$ concentrations in ambient air have been presented in Figs. 6a-d. These include the contributions originated from all the main source categories in the four Nordic cities (Umeå, Helsinki, Oslo and Copenhagen), and the regional background concentrations. The results have been computed on fine urban scale resolutions for Umeå, Helsinki and Oslo; of the order of from 20 to 50 m in the vicinity of the local sources, such as vehicular traffic, industrial sources and energy production. In case of RWC, the spatial resolutions of the dispersion modelling were approximately the same as for the emission inventories in each city, corresponding to that source category. For the Copenhagen domain, the spatial resolution of the dispersion modelling was 1 x 1 $km^2$.



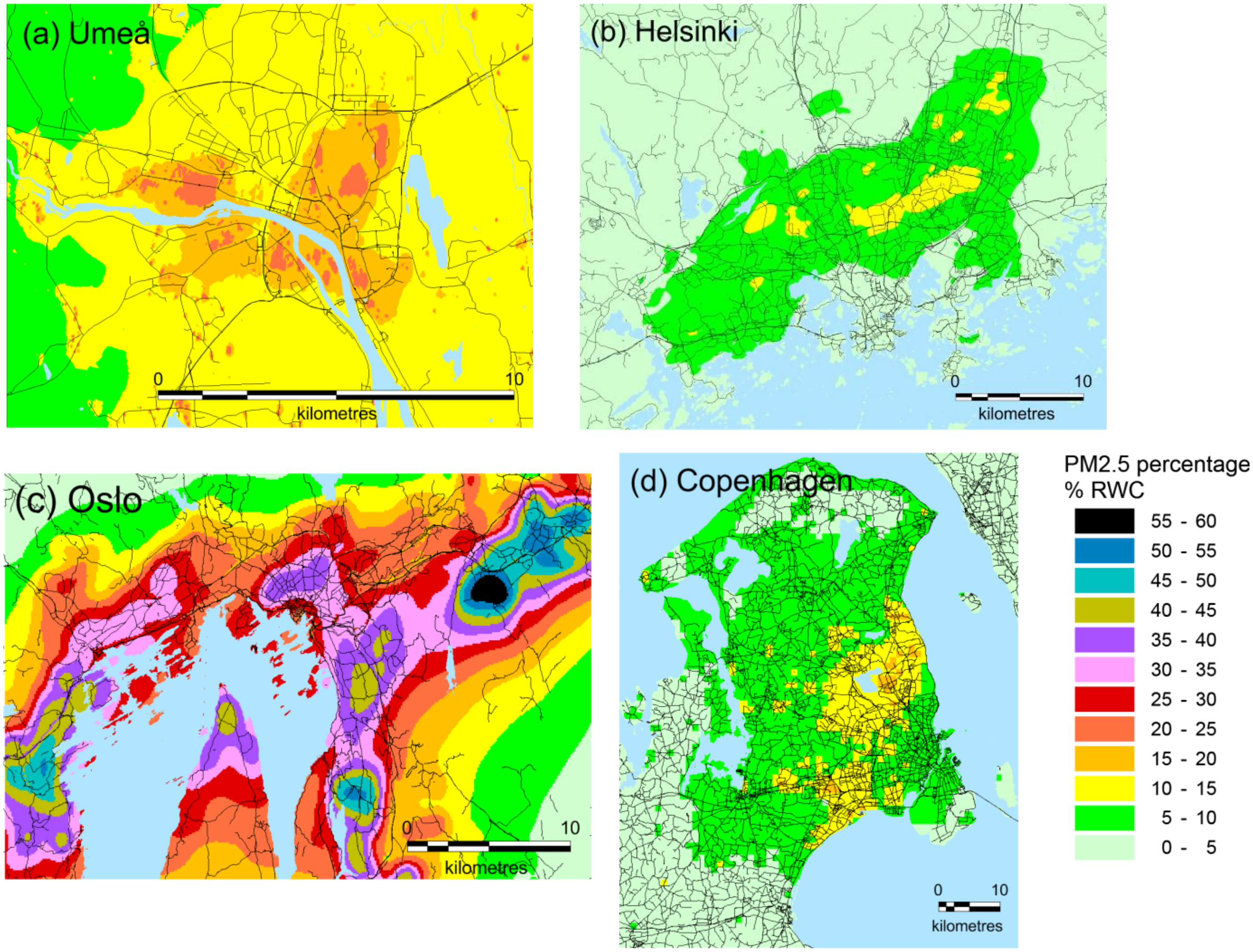

Figs. 6a-d. The predicted concentrations of $PM_{2.5}$ in Umeå (a), Helsinki (b), Oslo (c) and Copenhagen (d). The results represent the year 2011 for Umeå, 2013 for Helsinki and Oslo, and 2014 for Copenhagen. The main road and street networks have been presented as black lines. © OpenStreetMap contributors 2019. Distributed under a Creative Commons BY-SA License.

The concentration distributions are influenced also by the atmospheric dispersion conditions in the cities. In particular, Oslo is surrounded by higher ground and numerous hills, which tend to reduce the dilution of pollution, whereas the other target cities are located in a fairly flat terrain. Copenhagen and Oslo are located in a maritime climate, whereas Umeå and Helsinki are to a larger extent influenced also by continental climate conditions (Brandt et al., 2012; Segersson et al., 2017; Kukkonen et al., 2016, 2018; Yttri et al., 2019).

The annual average concentrations ranged spatially from 4 to 7 µg/m$^3$, from 6 to 10 µg/m$^3$, from 4 to more than 13 µg/m$^3$ and from 9 to more than 13 µg/m$^3$ in Umeå, Helsinki, Oslo and Copenhagen, respectively. The regional scale $PM_{2.5}$ concentrations in Denmark were higher than those in the other Nordic countries, due to the higher long-range transported contribution. Both regional background and local contributions were the lowest for Umeå. The reasons were that Umeå is clearly the smallest of the target cities as well as it is situated at larger distance from the main pollution source areas in Central, Central Eastern and Eastern Europe.

For Umeå, Oslo and Copenhagen, the highest concentrations occurred mainly in the city centres. For Helsinki, the detailed numerical data showed that the highest concentrations occurred (i) in the residential areas that are mainly situated north of the city centre and (ii) in the vicinity of the densely trafficked roads, and near the junctions of such roads. The influence of major traffic networks for all the cities are evident in the figures. Particularly, the overall distribution of concentrations in Oslo is very similar to that of the residential areas, and the concentrations tend to be relatively higher in the areas characterized by residential houses.

The fractions of RWC contributions to the $PM_{2.5}$ concentrations of $PM_{2.5}$ within the selected domains have been presented in Figs. 7a-d. The predicted fractions originated from RWC of the concentrations ranged spatially from 0 to 15 %, from 0 to approximately 20 %, from 8 to around 22 % and from 0 to 60 % in Helsinki, Copenhagen, Umeå and Oslo, respectively. The contributions of RWC in Oslo were clearly the highest within the target cities.




Figs. 7a-d. The spatial distributions of the source contributions of RWC to the concentrations of $PM_{2.5}$ as percentages in Umeå (a), Helsinki (b), Oslo (c) and Copenhagen (d). The results represent the year 2011 for Umeå, 2013 for Helsinki and Oslo, and 2014 for Copenhagen. © OpenStreetMap contributors 2019. Distributed under a Creative Commons BY-SA License.

These ranges of these annual average fractions have also been summarized in Fig. 8., presented both as percentages and as absolute concentration values. The RWC contributions were clearly highest for Oslo, both in terms of absolute concentrations and their proportions of the total $PM_{2.5}$ concentrations.

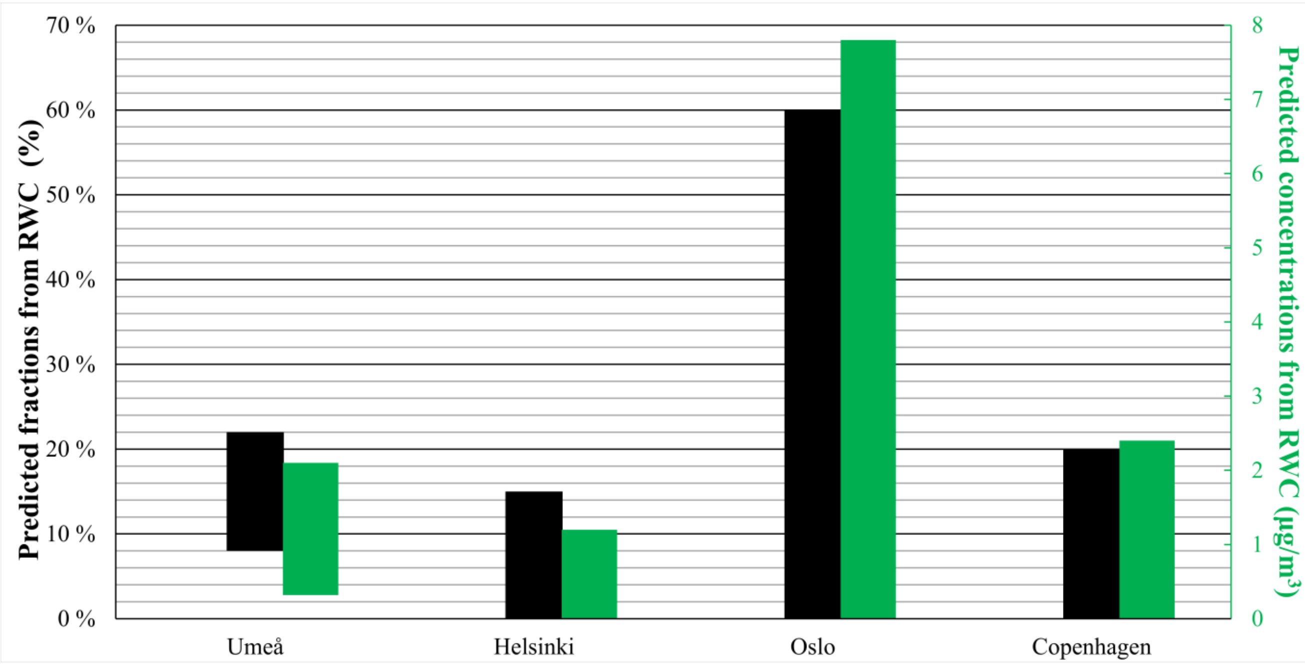



Fig. 8. The ranges of the source contributions of RWC to the concentrations of $PM_{2.5}$ as percentages (left-hand side axis) and as absolute concentrations (right-hand side axis) within the considered domains in Umeå, Helsinki, Oslo and Copenhagen. The results represent the year 2011 for Umeå, 2013 for Helsinki and Oslo, and 2014 for Copenhagen.

The fractions have been evaluated on an annual average level. In general, wood combustion is mainly used during the colder half of the year, and especially during the winter months; the fractions evaluated solely for the colder periods are therefore substantially higher.

In Umeå, the highest fractions occurred both in the city centre and in the vicinity of the nearby villages with a relatively larger density of residences. In Helsinki, the highest fractions occurred in the residential areas that are mainly situated west, north and north-east of the city centre. The use of wood combustion in the centre of Helsinki is negligible. In Copenhagen, the highest fractions were located in the residential areas situated to the north and west of the city centre, similarly to the case of Helsinki.

In Oslo, the contributions of RWC ranged from negligible at the outskirts of the domain to up to 60 % in some of the easternmost parts of the domain. Wood combustion contributed more than 40 % in central Oslo. The areas, in which the annual mean $PM_{2.5}$ concentrations were the highest (Fig. 6c) coincided with the areas, in which the source contribution from RWC was the highest (Fig 7c).

The high concentrations attributed to RWC in the eastern part of the domain in Oslo were caused by several factors. We have assessed the largest factor to be the intensive residential wood combustion activity in this region. Topography has also influence, as this region is at lower ground than the surrounding hills. However, the topography of this region is not substantially different from most other regions within this domain. It can also be seen based on Fig. 5c that the road and street network is fairly dense in this region; the vehicular emission is also a contributing factor.

The high percentages for Oslo were caused mainly by two reasons. First, wood combustion is used extensively within a fairly limited area (compared to the other target cities), for both the heating of smaller detached or semi-detached houses and for larger blocks of flats. The shares of wood combustion in Oslo were recently studied based on citizen involvement (López-Aparicio et al., 2017c). It was found that 46% of the wood used for residential heating was applied in residences in blocks of flats. Such flats use commonly wood stoves or open fireplaces as heating installations. The rest of the wood combustion took place in detached and semidetached houses, duplexes and townhouses.

The fractions of the $PM_{2.5}$ concentrations originated from RWC in Oslo were similar to those that have been previously evaluated by Tarrasón et al. (2018) for other Norwegian cities. Tarrasón et al. (2018) also evaluated such fractions using emission inventories, combined with dispersion modelling. According to that study, the annually averaged fractions from RWC in the areas of 13 Norwegian cities ranged from 20 to 75 %.

In principle, one or more of the considered years could be meteorologically exceptional or very rare, in terms of the ambient air temperatures. This could have a substantial effect on the use of wood combusted. We have therefore analyzed the seasonal variation of temperatures in the selected cities for four years. The main results of this analysis are presented in Appendix D. It can be concluded that none of the considered years was exceptional or rare in any of these cities, in terms of the seasonal variation of the ambient temperatures.

# 4. Conclusions

It has been evaluated that small-scale residential wood combustion (RWC) is a substantial source of atmospheric particulate matter globally, especially in Europe, Asia and Africa (e.g., Vicente and Alves, 2018; Butt et al., 2016). RWC has been found to be a significant source of pollution in all European regions, especially in Northwestern, and Central and Eastern parts of Europe (e.g., Karagulian et al., 2015).

In the continental Nordic countries (i.e., Denmark, Norway, Sweden and Finland), previous literature has addressed the emissions and concentrations attributed to RWC in various cities and regions, for various pollutants. However, there has been no harmonized analysis up to date of the situation in several Nordic cities, within the whole of the continental Nordic region. In this study, we have evaluated the emissions and ambient air concentrations of fine particulate matter in four Nordic cities, with a special emphasis on the contributions originated from small-scale residential wood combustion (RWC).

The reliable and accurate assessment of the emissions from RWC still remains a challenge. For the estimation of the emissions of wood combustion, one needs to survey and quantify numerous characteristics and factors of a multitude of individual residential sources. In principle, one needs to quantify the spatial distributions of the various categories of buildings using wood combustion, the amounts and distribution of firewood, the shares of primary and secondary heating sources, the amounts of wood used and the numbers of boilers, stoves, fireplaces, sauna stoves and other heating devices, and the emission factors for the different types of heating devices (e.g., Savolahti et al., 2016; Kukkonen et al., 2018; Karvosenoja et al., 2018; Grythe et al., 2019).

Due to the above-mentioned reasons, wood combustion emissions are commonly known less accurately than those from most other source categories, such as, e.g., vehicular traffic, larger scale energy production or industry (e.g., Karvosenoja, 2008; Karvosenoja et al., 2018). In this study, the largest uncertainties to the computed fractions of $PM_{2.5}$ concentrations attributed to RCW are probably caused by the inaccuracies of the emission inventories of RWC.

The numerical predictions were evaluated against measured urban scale data regarding the $PM_{2.5}$ concentrations in the four target cities. The fractional bias values were reasonably good for all the regional and urban background values, and for most of the traffic and RWC stations. The agreement of the daily modelled and measured time series was also fairly good in most cases.

The spatially averaged maximum emission values were the highest for Copenhagen and Oslo. The highest emissions from RWC were mostly located outside the city centres for Helsinki and Copenhagen. In the Helsinki region, the highest RWC emissions occurred in detached and semi-detached house areas, which were located outside the centre of Helsinki. For Copenhagen, the highest emission strengths were also found outside the most densely built city centre. In the Helsinki area, there is an extensive district heating system, and wood combustion is mainly used as a secondary heating system in detached or semi-detached houses.

In contrast to the above results for Copenhagen and Helsinki, the highest emission values from RWC in Umeå and Oslo were located within the city centres. In particular, in Oslo, the highest $PM_{2.5}$ emissions correspond to residential areas, which include aged blocks of flats and multifamily dwellings. There are also relatively high emission densities in the densely inhabited eastern parts of Oslo and in the neighbouring municipalities situated east of Oslo. In Umeå, the largest emissions were originated from relatively older buildings, which have not been connected to the district heating system.

Both the measured and predicted $PM_{2.5}$ concentration values were the highest for Copenhagen, caused mainly by the relatively higher regional background contributions, compared with the other three cities. The concentrations were second highest for Oslo, mainly due to substantial urban contributions. For Umeå, Oslo and Copenhagen, the highest concentrations occurred mainly in the city centres. In contrast, for Helsinki, the highest concentrations occurred in the suburban residential areas and in the vicinity of the densely trafficked roads. Major traffic networks had a substantial influence on the air quality for all these four cities.

The annual average fractions of RWC contributions to the concentrations of $PM_{2.5}$ ranged spatially from 0 to 15 %, from 0 to 20 %, from 8 to approximately 22 % and from 0 to 60 % in Helsinki, Copenhagen, Umeå and Oslo, respectively. The contributions of RWC in Oslo were clearly the highest in the target cities. In Oslo, the RWC contributions were up to 60 % in some of the easternmost parts of the domain, and larger than 40 % in central Oslo. In Copenhagen, the highest fractions were also located slightly outside the city centre, similarly to the case of Helsinki. The high percentages of the contributions of RWC in Oslo were mainly attributed to the fact that wood combustion was used extensively within a fairly limited area, and it was used both for the heating of smaller detached or semi-detached houses, and for larger blocks of flats.

The most significant research needs for an improved assessment of the concentrations from and exposure to RWC include the following. (i) The influence of actual meteorological parameters (especially the temporal variation of the ambient temperatures) should be explicitly allowed for in the modelling of the RWC emissions. (ii) The shorter-term temporal variations (daily, weekly and seasonal) of the RWC emissions should be evaluated more accurately. This could be done using measurements of chemical compounds that are specific for the emissions originated from RWC. (iii) More accurate experimental evaluations of the emission factors for the different types of heating devices, used in different conditions, would be needed. (iv) Improved diagnostic evaluation of the emission and atmospheric dispersion modelling against measured data, such as the evaluation in terms of the meteorological conditions and shorter-term temporal evaluation, would be needed, for developing more accurate emission and dispersion methodologies.

Whereas attempts have been made to regulate RWC in the Nordic countries, there are grounds for increased policy and technical measures, to avoid and alleviate harmful impacts of RWC, including especially those on human health. Regulation should consider the whole chain of events from emissions through population exposures to impacts. Both generic and more specific, targeted measures would be useful, in connection with general policies on air pollution, the environment, energy and climate, and those on urban planning, housing and buildings. The range of measures could include regulatory ones, information campaigns and economic steering, and their combinations.

# 5. Code and data availability

The SILAM code is publicly available. The emission data, and the measured and predicted concentration data used in this study is available, by contacting the responsible authors in each country, i.e., J. Kukkonen, C. Geels, S. López-Aparicio and D. Segersson.

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## Appendix A. Geographical regions and climates of the cities

Umeå is the largest city and the capital of the county of Västerbotten in northern Sweden. The domain addressed in this study includes the city and its surrounding areas. The terrain in the county rises from the gulf through a forested upland zone, and culminates in mountains near the Norwegian frontier. The city of Umeå is a medium sized Swedish municipality with 0.12 million inhabitants (all the population counts in this section are for 2018). The Ume River flows through the middle of the city and enters a bay in the Baltic Sea, at a distance of approximately five kilometers downstream of the city border. The yearly average temperature is 2.7 °C; average monthly temperatures vary from -7 °C in February to +16 °C in July (all the average temperatures presented in this section are based on the standard period of 1961-1990).

The Helsinki Metropolitan Area includes four cities: Helsinki, Espoo, Vantaa and Kauniainen. The total population in the area was approximately 1.1 million, whereas the population of Helsinki was about 0.64 million in 2018. The Helsinki Metropolitan Area is situated on a fairly flat coastal area. The annual mean temperature in Helsinki is 5.9 °C and the average monthly temperatures vary from -5 °C in February to +18 °C in July.

The city of Oslo and the Greater Oslo Region are situated at the northernmost end of a fjord and surrounded by hills that have heights of approximately 500 m above the sea level. The total population in Oslo was approximately 0.63 million in 2018, whereas the metropolitan area had a population of 1.7 million. Oslo has a humid climate; average monthly temperatures vary from -5 °C in January to +17 °C in July.

Copenhagen is situated on the flat eastern coast of the island Zealand; it is separated from Sweden by the narrow Øresund strait. The total population in the urban area of Copenhagen is approximately 1.6 million, whereas the city of Copenhagen has a population of 0.78 million. For the greater area of Copenhagen, the average monthly temperatures range from 0 °C in February to +16 °C in July.

## Appendix B. The assessment of emissions from other source categories in the target cities, in addition to RWC

### B1. Umeå

Exhaust emissions originated from vehicular traffic were estimated using measured traffic flow information, the traffic flow model EMME/2, and the emission factors of Handbook Emission Factors for Road Transport by Hausberger et al. (2009). Measured traffic flow information included separately light and heavy duty vehicles; this information was complemented with predictions provided by a traffic flow model. The information on the vehicle fleet composition was derived based on the national vehicle registry; however, this information was refined to allow for the local information regarding the share of heavy vehicles.

A resuspension model by Omstedt et al. (2005) was applied to evaluate the non-exhaust emissions. The model can be used to analyze also the wear due to studded tyres and street sanding.

Emissions from the category of other sources were extracted from the yearly national compilation of spatially distributed emissions in Sweden. The emissions originated from shipping were evaluated using the SHIPAIR model (Segersson, 2014). The largest contributions from the other sources were originated from off-road machinery and major point sources (Segersson et al., 2017).

### B2. Helsinki

An emission inventory for vehicular traffic for 2013 was used. This inventory included both traffic exhaust and traffic suspension emissions for the network of roads and streets in the Helsinki Metropolitan Area (HMA). The spatial distribution of vehicular emissions was based on detailed information on the line source network in the HMA provided by the Helsinki Region Transport. The number of line sources in the revised inventory was 26 536. The traffic volumes and average travel speeds at each traffic link were computed using the EMME/2 transportation planning system for three time periods of the day (HSL, 2011). The hourly traffic volumes were computed using a set of regression-based factors.

The total $PM_{2.5}$ exhaust emission values in the HMA for 2013 were estimated using data of the national calculation system for traffic exhaust emissions and energy consumption in Finland, called LIPASTO (Mäkelä and Auvinen, 2009), containing city-level data on emissions and mileages for various classes of vehicle types, and for streets and roads.

We have evaluated the hourly vehicular suspension emissions of $PM_{2.5}$ using emission factors computed using the FORE model (Kauhaniemi et al., 2011; 2014), which is based on the resuspension model by Omstedt et al. (2005). The same traffic mileage data was applied as for the estimation of exhaust emissions.

### B3. Oslo

We have considered the most important local emission source categories, such as RWC, on-road and non-road traffic, industry and shipping (López-Aparicio et al., 2017a). The emissions have been evaluated for the year 2013.

On-road traffic emissions were estimated at the road links, taking into account road type, width, length, the average daily traffic, the road vehicle distribution as vehicle class and vehicle technology class. The baseline emission factors were selected based on the Handbook Emission Factors for Road Transport (HBEFA, 2010), and they are adjusted based on the ageing of the vehicle, as a function of the mileage, and factors that relate to speed dependency.

Vehicular non-exhaust emissions of $PM_{2.5}$, due to suspension of road dust, were calculated based on a simplified version of the NORTRIP model (Denby et al., 2013). The NORTRIP model can be used to compute road surface moisture and dust production, dust loadings and suspended particulate emissions to the air. In its original form, the NORTRIP model is used to calculate the surface moisture separately for each and every road in the considered domain. However, when the model was applied in a simplified way, it was used to compute the surface moisture for two road categories: (i) a characteristic heavily trafficked road with salting and (ii) a less densely trafficked road without salting. The moisture at every road is then evaluated as a weighted average, depending on the road type.

Emissions from shipping were estimated based on the detailed shipping activity data from the port of Oslo following a bottom-up approach (López-Aparicio et al., 2017b). The emissions were computed following the method suggested by US EPA (2009). These are based on detailed information regarding the individual vessels visiting the port, the emission factors for each vessel category and operational modes of ships. The modelled industrial emissions consisted of emissions from point sources and diffuse emissions. The emissions from off-road mobile combustion included construction machinery, tractors, households and gardening.

**B4. Copenhagen**

The assessment of other emissions for the Danish area were based on the SPREAD model (Plejdrup et al., 2016; Plejdrup and Gyldenkærne, 2011). The main emission sectors included were stationary combustion, mobile combustion, fugitive emissions from fuels, industrial processes and product use, agriculture and waste. The SPREAD model evaluates yearly average emissions.

In this study, the road transport emissions were used as included in the national emission inventory within the SPREAD model. The emission factors were based on the COPERT V model, which was adapted to national conditions (Nielsen et al., 2017). The spatial distribution of the national road transport emissions was based on the Danish national GIS-based road network and traffic database, which includes mileage data in terms of road type and vehicle composition.

**Appendix C. A more detailed description of the comparison of model predictions and measured data.**

**C1. Evaluation of the predicted concentrations for Umeå**

For the model evaluation, we used the concentration data from a regional background station (Bredkälen), an urban background station (Biblioteket), one station in vehicular traffic environment (Västra Esplanaden), and four stations specially located to measure the contributions from RWC (Sävar, Vännasby, Vännas and Tavleliden). The results of these comparisons are presented in Table C1. The model computations have been performed for the years 2006-2013 (Omstedt et al., 2014; Segersson et al., 2017).

Table C1. Selected statistical parameters on the agreement of predictions and measurements for the daily concentrations of $PM_{2.5}$ in the Umeå area. The measured values at the station of Bredkälen have been adjusted slightly, based on regional-scale dispersion model computations. For Bredkälen, the results for two different periods have been separately presented.

| Name of the station | Classification | Observed mean (μg/m$^3$) | Predicted or adjusted (in case of Bredkälen) mean (μg/m$^3$) | Fractional bias | Number of data points | Measurement period |
|---|---|---|---|---|---|---|
| Bredkälen | Regional background | 1.93 | 3.97 | - | 1389 | 2009 -2012 |
| Bredkälen | Regional background | 1.90 | 3.18 | - | 196 | Nov 2012 – May 2013 |
| Biblioteket | Urban background | 4.90 | 5.70 | 0.16 | 49553 | 2006-2012 |
| Västra Esplanaden | Traffic | 7.80 | 11.60 | 0.39 | 48211 | 2006-2011 |
| Sävar | RWC | 3.60 | 4.30 | 0.18 | 399 | Nov 2012 - Dec 2013 |
| Vännasby | RWC | 4.20 | 4.80 | 0.13 | 286 | Nov 2012 - Dec 2013 |
| Vännas | RWC | 6.10 | 4.50 | -0.30 | 25 | Nov 2012 - May 2013 |
| Tavleliden | RWC | 3.80 | 3.50 | -0.08 | 20 | Jan 2013 - May 2013 |

The values include the annual average concentrations for the years 2006-2011 at the stations in the city of Umeå (Västra Esplanaden and Biblioteket). For the other stations (Sävar, Vännasby, Vännas and Tavleliden), the measured values are based on daily or weekly samples during the above-mentioned periods. The measurement sites represent a densely trafficked street canyon (Västra Esplanaden), urban background (Biblioteket), and residential environments (Sävar, Vännasby, Vännas and Tavleliden).

The agreement of the measured and modelled long-term average values can be considered to be fairly good for all the sites, except for the street canyon site (Västra Esplanaden).

The method used for the evaluation of the temporal variation of concentrations originated from RWC uses the measured concentration values of $PM_{2.5}$. We therefore cannot independently evaluate the performance of the model, with regard to the temporal correlations of the measured and predicted time series of concentrations.

### C2. Evaluation of the predicted concentrations for Helsinki

For the model evaluation, we used the concentration data from the following stations: regional background station of Luukki, the urban background station of Kallio, three stations in vehicular traffic environments (Mannerheimintie, Leppävaara, Tikkurila), and three stations specially located to measure the contributions from RWC (Vartiokylä, Tapanila and Kauniainen). The results of these comparisons are presented in Table C2.

Table C2. Selected statistical parameters on the agreement of predictions and measurements for the daily concentrations of $PM_{2.5}$ in the Helsinki area in 2013. Notation: RWC = Residential Wood Combustion.

| Name of the station | Classification | Observed annual mean (µg/m$^3$) | Predicted annual mean (µg/m$^3$) | Index of agreement | Factor-of-two (%) | Fractional bias | Number of data points |
|---|---|---|---|---|---|---|---|
| Luukki | Regional background | 6.3 | 6.7 | 0.59 | 58 | 0.05 | 364 |
| Kallio | Urban background | 7.0 | 7.2 | 0.64 | 65 | 0.04 | 364 |
| Mannerheimin-tie | Traffic | 8.6 | 7.6 | 0.60 | 64 | -0.13 | 363 |
| Leppävaara | Traffic | 7.1 | 8.1 | 0.68 | 74 | 0.12 | 363 |
| Tikkurila | Traffic | 7.2 | 7.8 | 0.73 | 75 | 0.08 | 363 |
| Vartiokylä | RWC | 6.8 | 7.5 | 0.69 | 68 | 0.10 | 351 |
| Tapanila | RWC | 9.1 | 7.8 | 0.67 | 65 | -0.16 | 360 |
| Kauniainen | RWC | 7.1 | 7.3 | 0.65 | 65 | 0.03 | 360 |

Overall, the modelled $PM_{2.5}$ concentrations agreed fairly well or well with the measured data. The values of the index of agreement (IA) and the factor-of-two (F2) were slightly lower at the regional background station of Luukki, compared with the corresponding values for the urban stations.

The range of model performance was similar at the three traffic stations compared to the corresponding performance at the RWC–influenced stations. For instance, the IA values ranged from 0.60 to 0.73, and from 0.65 to 0.69 at the traffic and RWC –influenced stations, respectively. Concerning the traffic station of Mannerheimintie in the centre of the city, there is an under-prediction (FB = - 0.13), which can be attributed to the reduced dilution caused by buildings and the frequent congestion of traffic. There is also under-prediction at the station of Tapanila (FB = – 0.16), located in a residential area.

## C3. Evaluation of the predicted concentrations for Oslo

The modelled regional background $PM_{2.5}$ concentrations were compared to the regional background $PM_{2.5}$ measurements at the station of Hurdal in southern Norway. The mean fractional bias varied from -0.54 to 0.65 during the whole year. We noticed that the modelled ensemble results in $PM_{2.5}$ weekly means were lower in summer when compared to the measurements, whereas during the rest of the seasons they are remarkably higher, especially from October to December. These differences might be explained by (i) the inaccuracies related to partially missing secondary organic aerosol formation in the model ensemble, (ii) the inaccuracies in modelling particulate matter originated from biogenic sources (Aas et al., 2014), and (iii) uncertainties in primary aerosol emissions (Marécal et al., 2015). The predicted regional background concentrations were based on an ensemble of seven chemical transport models, four of which did not include secondary organic aerosol formation processes. The contribution of the background concentrations to the $PM_{2.5}$ results within the city of Oslo is on average 56%.

For the urban scale model evaluation we utilized the concentration data from all the available permanent measurement stations within the selected domain in 2013. All the stations in the area were designed as traffic stations, except for one urban background station. Even though there are no stations which could be used to measure the contributions of RWC, Akerbergveien, Bygdoy Alle and Kirkeveien traffic monitoring stations could be considered as the most influenced by RWC emissions. These stations are located in urban roads surrounded by residential areas (i.e., blocks of flats) characterized by intense wood burning activity. The selected statistical parameters of this evaluation are presented in Table C3.

Table C3. Selected statistical parameters on the agreement of predictions and measurements for the daily concentrations of $PM_{2.5}$ in the Oslo area in 2013.

| Name of air quality station | Classification | Observed annual mean ($\mu g.m^{-3}$) | Modelled annual mean ($\mu g.m^{-3}$) | Index of agree-ment | Factor-of-two (%) | Fractional bias | Number of data points |
|---|---|---|---|---|---|---|---|
| Hurdal | Regional | 3.1 | 3.6 | 0.33 | 59 | 0.12 | 51 |
| Sofienbergparken | Urban background | 11 | 10 | 0.72 | 67 | -0.096 | 365 |
| Akebergveien | Traffic | 9.2 | 9.9 | 0.82 | 83 | 0.072 | 335 |
| Alnabru | Traffic | 16 | 10 | 0.63 | 66 | -0.43 | 356 |
| Bygdoy Alle | Traffic | 12 | 9 | 0.74 | 71 | -0.33 | 365 |
| Hjortnes | Traffic | 9.4 | 9.4 | 0.79 | 89 | 0.0028 | 364 |
| Kirkeveien | Traffic | 8.6 | 8.0 | 0.73 | 83 | -0.062 | 345 |
| Manglerud | Traffic | 9.0 | 10 | 0.71 | 88 | 0.15 | 361 |
| RV4 Aker Sykehus* | Traffic | 9.1 | 10 | 0.65 | 78 | 0.14 | 201 |
| Smestad* | Traffic | 10 | 7.9 | 0.83 | 92 | -0.27 | 193 |

* RV4 Aker Sykehus and Smestad were not in operation from May to mid-October.

The model simulation results have been benchmarked with the DELTA tool (http://aqm.jrc.ec.europa.eu/index.aspx), which has been developed by the Joint Research Centre in the framework of the FAIRMODE action (Forum for air quality modelling in Europe; http://fairmode.jrc.ec.europa.eu/). The analysis that the results of all considered air quality stations in Oslo fulfill the Model Quality Objectives, as defined by this assessment tool.

Results showed that the comparison of the model performance and the agreement between measurements and predictions amongst traffic stations was poorer for the station of Alnabru. This station is located between two roads in a valley, along where winds from the Oslo fjord frequently transport substantial pollution from central Oslo.

As expected, the highest $PM_{2.5}$ concentrations were observed in winter and daily values at the urban background station (Sofienbergparken) can be higher than 40 $\mu gm^{-3}$. According to a survey on wood combustion by Statistics Norway, emissions from RWC are diurnally the highest in the evening.

### C4. Evaluation of the predicted concentrations for Copenhagen

For the model evaluation, we exploited the concentration data from the regional background station of Risø, the urban background station of HCØ (H.C. Ørsted Institute), two stations in vehicular traffic environments (JGTV, Jagtvej and HCAB, H.C. Andersens Boulevard), and one station in a suburban area (Hvidovre). The latter site was selected to represent the influence of residential small-scale combustion. The results of these comparisons are presented in Table C4.

Table C4. Selected statistical parameters on the agreement of predictions and measurements for the daily concentrations of $PM_{2.5}$ in the Copenhagen area. The results correspond to the period 2013-2017, except for the stations of JGTV and Hvidovre, for which the measurements were started later on, in November, 2013 and in June, 2015, respectively. Notation: RWC = Residential Wood Combustion.

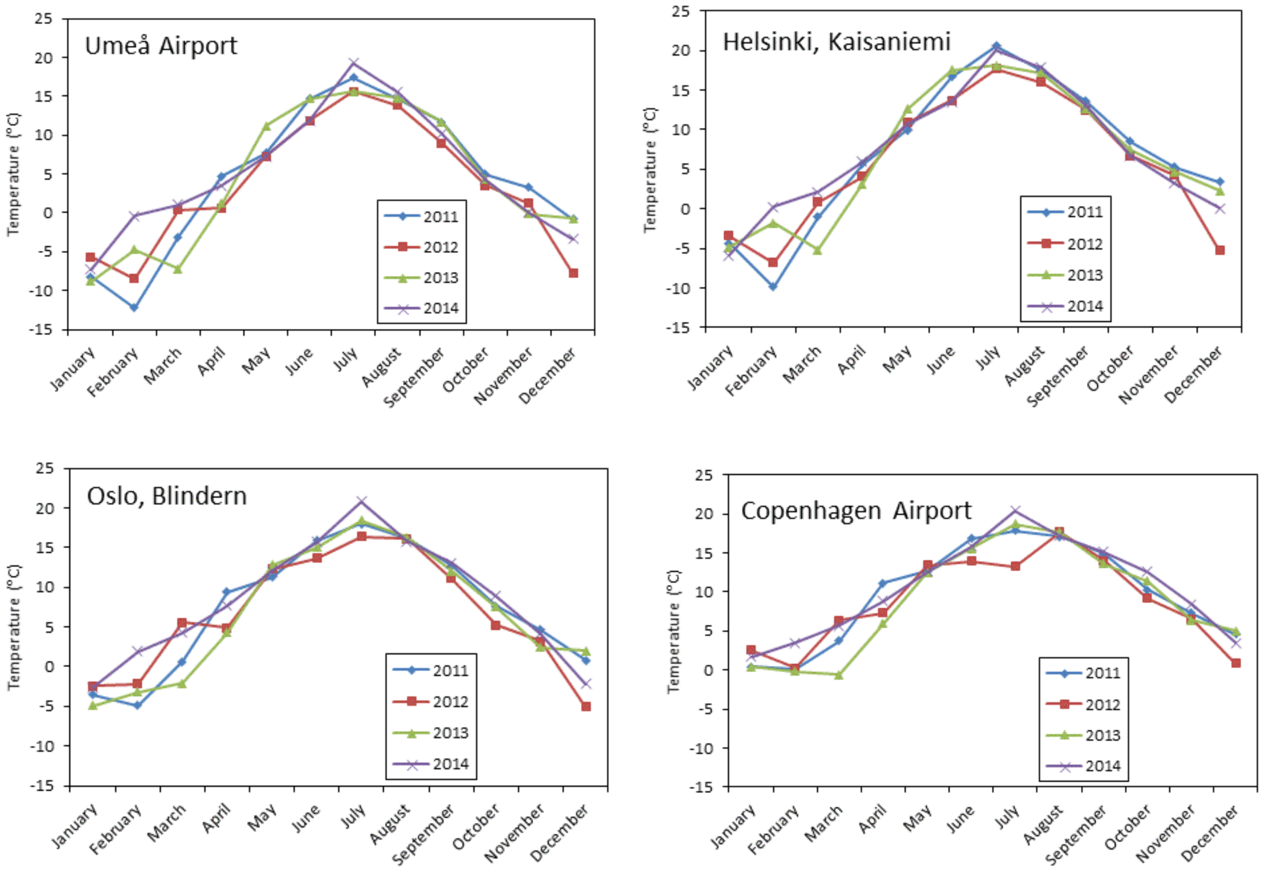
| Name of the station | Classification | Observed annual mean ($\mu g/m^3$) | Predicted annual mean ($\mu g/m^3$) | Index of agreement | Factor-of-two (%) | Fractional bias | Number of data points |
|---|---|---|---|---|---|---|---|
| Risø | Regional background | 10.5 | 10.0 | 0.70 | 95 | -0.05 | 1753 |
| HCØ | Urban background | 11.6 | 9.7 | 0.68 | 93 | -0.18 | 1649 |
| JGTV | Traffic | 14.7 | 9.7 | 0.59 | 82 | -0.41 | 1415 |
| HCAB | Traffic | 16.6 | 9.8 | 0.45 | 70 | -0.52 | 1638 |
| Hvidovre | RWC | 10.3 | 9.5 | 0.68 | 94 | -0.09 | 870 |

Regarding regional background concentrations, the model predicted well both the long-term averages (FB = - 0.05) and their daily variability (IA = 0.70). For the urban background site, the model predictions were also good or fairly good (FB = - 0.18, IA = 0.68).

The modelling was done using a spatial resolution of 1 x 1 $km^2$. The concentrations at the two vehicular traffic sites were substantially underestimated (FB = - 0.41 and – 0.52). The agreement of the daily temporal variations at the traffic sites were also lower than the corresponding one for the other selected stations. With respect to the site affected by RWC, the model performance was fairly good.

**Appendix D. Evaluation of the variations of the ambient temperatures during the period 2011 – 2014 in the four selected cities.**

The seasonal variations of temperatures at four measurement stations in the four cities have been presented in Figs. D1 a-d. The data has been extracted from the open data portals of the Swedish, Finnish, Norwegian and Danish meteorological institutes. The selected stations can be considered to be representative for the meteorological conditions in these cities. For Copenhagen, the earliest part of the data, i.e., from 1 Jan, 2011 to 30 May, 2012 has been interpolated, based on data from several meteorological stations in Denmark.




Figs. D1 a-d. The monthly averaged values of the measured ambient temperatures in the four selected cities, for the years 2011 – 2014.

None of the considered years can be considered to be exceptional or rare at any of these locations, in terms of the ambient temperatures.

## Appendix E. An overview of the regulatory frameworks for RWC in four Nordic countries

We address both (i) the EU policies on regulation of emissions from RWC, and (ii) their implementation into national laws and regulations at member-state level in the four considered Nordic countries. Sweden, Denmark and Finland are EU member states. Norway is a country associated with EU; however, it also complies with the same directives.

The main characteristics of the air quality policies are similar across the four considered Nordic countries, as these are based on the same EU regulations and directives. However, the national governance systems and the procedures of RWC are in some cases substantially different across the countries, which is reflected in varying national policies and regulations. The policies and regulation that manage air pollution originated from RWC are linked to regulation of clean air, urban planning, urban environment and the heating of houses in the Nordic countries.

In the following, the relevant EU regulations are first briefly reviewed. Second, we discuss the specific regulations in each country, their implementation and measures to abate pollution from RWC.

### E1. The EU regulations

Emissions from woodstoves are regulated in the EU by the Clean Air Directive and its successors and associated directives. In particular, the Ecodesign Directive, i.e., EU 2015/1185 and the directive EU 2015/1189 address the regulations of harmful emissions attributed to RWC. Recognition of the health and environmental costs of air pollution has encouraged the establishment of a common strategy on air pollution. This has stimulated a growing number of directives since the 1990's.

The main directives were merged into the Clean Air Directive, 2008/50/EC on Ambient Air Quality and Cleaner Air for Europe. This directive aimed to reduce air pollution to a level, which could be considered to be not harmful to both human health and nature. This directive included also the reduction of $PM_{2.5}$ emissions attributed to RWC. The Clean Air Directive specifically targets air quality in cities and requires the member states to lower the level of exposure to $PM_{2.5}$ by 20 % by 2020, relative to the corresponding levels in 2010. The emissions from woodstoves in residential areas are therefore regulated by the overall Clean Air Directive, although the EU has not issued any specific directive on RWC.

### E2. The RWC regulations in four Nordic countries

#### E2.1 Finland

In Finland, RWC for heat production in private homes has contributed substantially to the national $PM_{2.5}$ emissions within the 1990's, 2000's and 2010's (Kukkonen et al., 2018). The Ministry of Health and Social Welfare has stressed out the importance of emissions from wood stoves and the need to limit the associated emissions.

Finland issued its first national programme for the protection of air quality in 2002, complying with the EU Clean Air Directive of 2001. Emissions originated from several source categories of air pollution, such as industry, large-scale energy production and most modes of transport, have since that time been regulated and reduced. However, the national emissions from RWC have not substantially decreased. A new national programme for the protection of air quality has been proposed in 2018, as part of the implementation of the Emissions Ceiling Directive by the EU in 2016 (Ministry of the Environment, 2019).

The 2018 national programme assesses the compliance with the national emission ceiling (NEC) directive and gives recommendations for further actions to reduce health impacts and environmental damage caused by air pollution. It also includes policy instruments for limiting emissions in general terms; however, the emissions from RWC are not directly targeted. In addition, this programme reviews the previous information and awareness raising campaigns (Savolahti et al., 2015), which have been linked to national eco-label for wood-heated sauna stoves.

The 2018 national programme enables local authorities to intervene in RWC smoke issues. The EcoDesign Directives of boilers (EU 2015/1189) and space heating appliances using solid fuels (EU 2015/1185) regulate most of the RWC combustion appliances used in Finland. However, the stoves in saunas have been excluded from these regulations. The EcoDesign regulations aim at improved energy efficiency and lower emissions of such wood combustion appliances, which will be sold after 2022. However, the regulations have neither extensively addressed wood burning habits nor older appliances that are already in operation.

The authorities in some cities, such as the Helsinki Metropolitan Area, have also called for improved control of wood stoves (Kaski et al., 2016). Concrete actions include information campaigns for proper storage of fuels and use of wood stoves in the Helsinki Metropolitan Area.

### E2.2 Denmark

Policies and policy actions that target emissions from wood stoves in accordance with the EU guidelines emerged on Danish political agendas in the early 2000's. The Environmental Protection Agency issued the Woodstove Regulation in 2001, which allocated the authority to local governments to regulate the use of woodstoves in cases of severe pollution. The most recent Woodstove Regulation in 2018 revised the regulation of use and emissions from woodstoves. Implementation of major changes in combustion plants are required prior to installation, in order to document compliance with emission standards, and thus fulfill the monitoring requirements of the EU's Clean Air Directive. The use of petroleum coke has not been allowed in private households since 2019.

Policy instruments to implement the Danish clean air policy are based on various measures to meet the standards regarding clean burning in woodstoves, as outlined in the Clean Air Directive. As a key strategic tool, the monitoring of compliance is divided between the Environmental Protection Agency and local governments; this involves using networks of monitoring stations.

Based on the Danish Planning Act, local governments have the option of regulating the use of wood stoves in specified urban zones; although only to a limited extent. The Woodstove Regulation in 2018 widens the legal options for local governments to issue plans and regulations for establishment and use of combustion plants. Over the past few years, local governments have called for a change of law, which would enable them to ban woodstoves in specified residential areas. In early 2018, more than a tenth (12 of 98) of the Danish Local Governments had issued regulations for the use of woodstoves, recommending ways and types of fuels suitable for use in domestic woodstoves, including also details on the operation of fuels and stoves.

Regulation of RWC has also included economical policy instruments. Since the first Woodstove Regulation in the early 2000's included a subsidy scheme, which targeted at renewing woodburning technologies to cleaner stoves. This scheme rewarded woodstove owners, if they changed woodstoves that were older than 1990. This scheme has been effective; most of the older than 1990 implemented wood stoves have been substituted by more recent models.

In addition, incentive-based instruments have been used to change behaviour and reduce emissions. These instruments have relied on information campaigns, in which professional chimney sweepers were involved. The chimney sweepers have encouraged clean practices for the use of RWCs during the compulsory annual inspections of woodstoves.

Woodstoves have been almost exclusively used as a supplementary heating source in Denmark, due to an extensive infrastructure of district heating. Household heating is therefore less dependent on woodstoves, compared with the situation in Norway and Sweden. In Denmark, household heating is commonly associated with the convenience use of woodstoves.

There has been an emerging public awareness of the harmful effects of the pollution attributed to RWC in Denmark during the last few years, especially regarding the effects in densely populated urban areas. The health effects of RWC have received attention both at national and local city levels. The Danish policy agendas are therefore gradually recognizing better the pollution from RWC.

### E2.3 Norway

As an associated country to the EU, Norway implements the EU directives and regulations. The Norwegian ambient air quality policy therefore at least complies with the EU's Ambient Air Quality Directive of 2016. Woodstoves play a significant role as heating sources in Norwegian households. The RWC pollution is targeted both nationally and locally. The secondary homes in Norway are commonly heated by wood stoves; however, these are commonly located in sparsely populated areas. The exposure to emissions from such wood stoves is therefore limited.

At local level, air quality is regulated by three separate legal mechanisms: (i) the Pollution Act, (ii) the national air quality objectives specified by the government and (iii) the air quality standards. The compliance is monitored using a network of monitoring stations dispersed across Norway. This system is in accordance with the Clean Air Directive's standards and procedures for monitoring.

In addition, the regulation of emissions originating from RWC is based on economic instruments. In several municipalities, there are economic incentives to replace older wood stoves with new installations based on cleaner wood burning technologies. For instance, in Oslo municipality since 1998, there has been a payment plan for this purpose. It has been estimated that from 1998 to 2015, approximately 8700 wood stoves had been replaced using the granted support. Incentive-based policy instruments are also applied to motivate a change of woodburning behaviour. These are focused on information campaigns.

### E2.4 Sweden

The Swedish regulation of RWC is based on the EU Clean Air Directive. It aims at regulating multiple uses of woodstoves, especially including households. Heating of a large fraction of the residential houses and workplaces is based on the use of woodstoves in Sweden. The $PM_{2.5}$ concentration levels are the highest in the capital city of Stockholm and in other major cities. This is reflected in policies and regulations. The Government New Strategy for Clean Air in the 2010's recognized the severe health risks associated with the $PM_{2.5}$ exposure and identified wood stoves as a significant source of the $PM_{2.5}$ emissions.

The National Board of Housing Building and Planning is responsible for the administration and implementation of The Planning and Building Act. The Act specifies spatial zoning and allows local governments to limit the use of RWC and ban the wood burning practices with the highest emissions. A range of Swedish cities, towns and local governments use this regulatory tool to reduce emissions from RWC, especially in densely populated areas. For instance, the authorities in the city of Malmø have extensively applied zoning to limit the air pollution from woodstoves. The city has prohibited the so-called convenience use of woodstoves during the warmer months from April to September.

Moreover, there is a national scheme for promoting research and development for cleaner RWC technology, known as the Eco-design scheme. The scheme subsidies innovation and promotion of more efficient and cleaner wood stoves, under the administrative authority of the Energy Agency. In addition, local governments disseminate information on public websites to encourage awareness of cleaner wood burnıng practices, as well as to disseminate knowledge of specified local zoning regulations.

## E3. Discussion on RWC regulations and measures in four Nordic countries

Emissions from woodstoves in all the considered Nordic countries are regulated based on the EU Clean Air Directive. The maximum allowed concentration levels therefore need to comply with the same values. The regulation concerning the monitoring networks and systems is also the same in these countries, as outlined in the Clean Air Directive. In addition, the Nordic countries target RWC pollution by a range of national regulations.

The Nordic regulation of emissions from RWC includes three types of policy instruments: regulatory, economic and incentive-based. The regulatory policy instruments in the Nordic countries include (i) national requirements for local governments to consider spatial zoning that includes air quality, and (ii) ban of wood stoves in high- to medium-risk urban areas, especially residential areas. This may imply amendments to National Planning Acts at an urban level. The economic policy instruments include (i) subsidies for substitution to wood stoves based on cleaner burning technology at household level, (ii) integration the social costs (including health costs) associated with wood burning in the price of RWC technology, maintenance and fuels, and (iii) financial support for research and development. The incentive-based measures include (i) research and innovation to develop cleaner wood burning technology and to improve assessment of RWC exposures, and (ii) information campaigns to raise public awareness and develop skills on cleaner wood burning practices. The campaigns are addressed to citizens and households, sellers of wood stoves, and chimney cleaners.

Despite the use of the above-mentioned policy instruments, there are challenges in achieving the required lower levels of pollution. In particular, experience has shown that the spatial zoning regulations, combined with a direct regulation on the use of woodstoves, are an efficient regulatory tool in urban areas. For instance, the authorities in Sweden have used this method in urban areas in combination with detailed specification of when, how and how often wood stoves can be used. However, spatial zoning regulations are limited by other national and local regulations and plans. Regulations to ban of the use of wood stoves in new residential or urban areas have also been suggested; however, this is currently outside the legal framework for spatial planning.

The use of more advanced and cleaner wood burning technologies has been successfully supported in some Nordic countries by using subsidies to households that voluntarily replace older stoves with new ones. Information campaigns have been organized in all the four Nordic countries regarding the proper storage of fuels and the use of wood stoves. The use of stoves with cleaner burning technology and improved wood burning habits will also be useful in view of the economic factors.

Information campaigns have also targeted to promote cleaner burning practices of households. However, the habits related to the collection, storage and combustion of wood are closely related to the long-term historical and cultural aspects in all these four Nordic countries. Wood combustion has played an important role for residential heating in Nordic countries for centuries. A more current trend is the use of RWC as a supplementary heating method, and its convenience use.

## 8. Author contributions

Jaakko Kukkonen has coordinated the analyses, compiled together the information and written a substantial fraction of the article. David Segersson, Gunnar Omstedt, Camilla Andersson and Bertil Forsberg have provided the relevant information and analyses on the measurements and modelling in Umeå. Camilla Geels, Ulas Im, Jesper H. Christensen, Ole-Kenneth Nielsen, Marlene S. Plejdrup, Jacob Klenø Nøjgaard and Jørgen Brandt have provided the relevant information regarding Copenhagen. Leena Kangas, Mari Kauhaniemi, Ari Karppinen, Mikhail Sofiev and Heidi Hellén have provided the relevant information regarding Helsinki. Susana López-Aparicio, Gabriela Sousa Santos and Ingrid Sundvor have provided the relevant information regarding Oslo. Kari Riikonen, Juha Nikmo and Androniki Maragkidou have worked on the processing, analysis and harmonisation of the datasets for all the target cities. Anne Jensen, Timo Assmuth and Niko Karvosenoja have written the section regarding regulatory frameworks in four Nordic countries. In addition, Niko Karvosenoja has analysed the emission data for all the four cities. Anu Kousa and Jarkko V. Niemi have compiled the RWC emission inventory in the Helsinki Metropolitan Area.

## 9. Competing interests

The authors declare that they have no conflict of interest.