Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-968
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 3 January 2018

Atmospheric Chemistry and Physics Discussions

Modelling of the urban concentrations of $PM_{2.5}$ on a high resolution for a period of 35 years, for the assessment of lifetime exposure and health effects

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Abstract. Reliable and self-consistent data on air quality is needed for an extensive period of time for conducting long-term, or even lifetime health impact assessments. We have modelled the urban scale concentrations of fine particulate matter ($PM_{2.5}$) in the Helsinki Metropolitan Area for a period of 35 years, from 1980 to 2014. These high resolution computations included both the emissions originated from vehicular traffic (separately exhaust and suspension emissions) and those from small-scale combustion, and were conducted using the road network dispersion model CAR-FMI and the multiple source Gaussian dispersion model UDM-FMI. The regional background concentrations were evaluated based on reanalyses of the atmospheric composition on global and European scales, using the SILAM model. The modelled concentrations of $PM_{2.5}$ agreed fairly well or well with the measured data at a regional background station and at four urban measurement stations, during 1999 – 2014. There was no systematic deterioration of the agreement of predictions and data for earlier years (the 1980's and 1990's), compared with the results for more recent years (2000's and early 2010's). The local vehicular emissions were about five-folds higher in the 1980's, compared with the emissions during the latest considered years. However, the local small-scale combustion emissions increased slightly over time. The highest urban concentrations of $PM_{2.5}$ occurred in the 1980's; these have since decreased to about to a half of the highest values. However, there is only a very slight decreasing trend of the $PM_{2.5}$ concentrations during the last decade. Regional background is the largest contribution in this area. Vehicular exhaust has been the most important local source, but the relative shares of both small-scale combustion and vehicular suspension emissions have increased. The study provides long-term, high-resolution concentration databases on regional and urban scales that have been used for the assessment of health effects.

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1 Introduction

This study has been part of the multidisciplinary research programs entitled "The Influence of Air Pollution, Pollen and Ambient Temperature on Asthma and Allergies in Changing Climate, APTA" (http://www.oulu.fi/apta-en/) and "Understanding the link between Air pollution and Distribution of related Health Impacts and Welfare in the Nordic countries, NordicWelfAir" (http://projects.au.dk/nordicwelfair/). The overall objective of the APTA project was to assess the influence of environmental exposures related to changing climate on asthma and allergies, and on cause-specific mortality. One of the aims of the NordicWelfAir project was to further understand the link between air pollution levels, the chemical composition of pollution and the related health effects.

A special focus of the present study was on health effects of air pollution, allergenic pollen and extreme climatic factors, especially ambient temperatures. Asthma is globally the most common chronic disease in children (WHO, 2011) and the prevalence of asthma in adult populations varies from 5 to 25 %. Allergies are even more common, with prevalence of 30 – 40 %. Accurate information on the long-term concentrations of air pollutants and pollen species on a high spatial and temporal resolution was required for conducting life-time health impacts assessments.

Only few studies have estimated the past levels air pollution over decades. However, some studies have been conducted, e.g., for the northern hemisphere (Xing et al., 2015) and for specific continents, regions or countries (Guerreiro et al., 2014, Colette et al., 2011, Cho et al., 2011, Astitha et al., 2017). All of these studies have addressed a relatively short time period, commonly from a few years up to two decades. The evaluations of air quality for urban areas over longer periods have been especially rare (Gupta and Christopher, 2008, Wise and Comrie, 2005). Previous investigations have not addressed the modelling of spatially and temporally high resolution concentrations in cities or urban agglomerations over a period of several decades.

Xing et al. (2015) evaluated air quality trends during a 21-year period, 1990–2010, in the Northern Hemisphere, using both observations and modelling. They used the Community Multiscale Air Quality (CMAQ) model driven by meteorology from Weather Research and Forecasting (WRF) simulations and the emission inventories from EDGAR. They concluded that the model successfully reproduced the observed decreasing trends in SO_2, NO_2, 8 h O_3 maxima, SO_4^{2-} and elemental carbon (EC) in the US and Europe. However, the model also failed to reproduce part of the trends, such as that of NO_3^- in the US, and some trends for China. Due to the coarse spatial resolution, the modelling underestimated most concentrations in urban networks.

Guerreiro et al. (2014) presented an analysis of air quality in Europe from 2002 to 2011. The evaluation was based on ambient air measurements and data on anthropogenic emissions. They reported that the emissions of the main air pollutants in Europe declined in the considered period, resulting in improved air quality across the region, for some of the pollutants. The trends of PM10 were on average decreasing across Europe. Most of the registered significant trends of the measured PM10 concentrations were decreasing in monitoring stations that were situated mostly in France and Germany. However, the data at most of the stations did not show significant trends. In case of the $PM_{2.5}$ concentrations, most of the stations did not register significant trends in the period 2006 - 2011.

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Colette et al. (2011) analyzed air quality trends in Europe during a decade, from 1998 to 2007, using both measurements and six regional and global chemistry-transport models. They presented the trends of NO_2, O_3 and PM_{10}, based on both the measured data and modelled results. However, this article was focused on the evaluation of the performance of the selected models.

Astitha et al. (2017) evaluated the trends of ozone concentrations over the contiguous United States from 1990 to 2010, using the coupled WRF-CMAQ system. They addressed various O_3 measures, such as, e.g., the daily maximum 8-hr ozone concentrations, during the warm seasons. During the earlier decade (from 1990 to 2000), the simulated and observed trends were not statistically significant. During the more recent decade, all observed trends were decreasing statistically significantly. Wise and Comrie (2005) analyzed meteorologically adjusted ozone and PM data from 1990 to 2003 for five major metropolitan areas in the Southwestern U.S. Long-term ozone trends indicated increasing concentrations over the considered decade; the long-term PM concentrations did not show any significant trends.

Gupta and Christopher (2008) analysed particulate matter concentrations in Birmingham, U.S., during a seven year period, 2002 – 2006, based on both surface and satellite measurements. They used the aerosol optical thickness (AOT) values determined by the Moderate Resolution Imaging Spectroradiometer (MODIS), and ground measurements of particulate matter mass. They concluded that the $PM_{2.5}$ mass concentrations over Northern Birmingham were decreased by about 23 %. However, the MODIS-Terra AOT data was available only about 50 % of the time, due to cloud cover and unfavorable surface conditions.

Cho et al. (2011) reviewed studies that addressed the concentrations and size distributions of ambient airborne particulate matter containing Pb, from 1980 to 2008. The maximum three month average airborne Pb concentrations in total suspended particulate matter decreased 92 % at 19 national sites in the U.S. during the considered period. However, the relevant size distribution data were very scarce.

The first systematic review of urban air quality in Finland was presented by Kukkonen et al. (1999). They considered the data from 42 Finnish cities and towns, in 1990-1993. The results showed that the national air quality guidelines were fairly often exceeded in the early 1990's in urban areas, most commonly for particulate matter. Some exceedances also occurred for NO_2 and CO, at sites with high traffic densities. However, the European Union air quality limit values were only exceeded at one monitoring station for all of the years considered. More recently, Anttila and Tuovinen (2010) evaluated the trends in the atmospheric concentrations of the main gaseous and particulate pollutants in Finland for the period of 1994–2007, based on measured data at both regional background and urban stations. They found that the concentrations of SO_2, CO and NO_x declined considerably during the considered period, whereas the concentrations of O3 increased in the urban data. For PM_{10}, five of the studied 12 trends were decreasing at the urban locations.

The main empirical framework of the present study addressed the Helsinki Metropolitan Area (HMA), from 1980 to 2014. The Espoo Cohort Study (ECS), which is a major prospective, population-based epidemiologic study, formed the basis for investigating the health impacts, in addition to registry-based hospitalization and mortality data. The ECS includes detailed information on the lifetime history of the residences and work places of the cohort members, as well as extensive

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information on health, environmental exposures and behavioral factors collected both prospectively and by linkage to available health and environmental information registries (Jaakkola et al., 2010, Paaso et al., 2014). However, for the assessment of lifetime environmental exposure, a database would first be required on the concentrations of the key pollutants, allergenic pollen species and meteorological factors over a time period of 35 years.

The first aim of this study was to provide an accurate and reliable high-resolution database on the urban scale concentrations of $PM_{2.5}$, to be used in the subsequent health impact assessments. This article therefore presents the emission and dispersion modelling in the Helsinki Metropolitan Area, for the three and a half decades that are relevant for the assessment of long-term exposures on health. A new, comprehensive emission inventory is presented for $PM_{2.5}$ originating from small-scale combustion in this area. We have also extended the urban emission inventories regarding the exhaust and suspension emissions of vehicular traffic, and those for small-scale combustion, for a multi-decadal period. The second aim was to evaluate the reliability and accuracy of the modelled concentrations, by comparing those with the corresponding available measured concentrations. The third aim was to present selected results regarding the long-term temporal evolution of the concentrations of $PM_{2.5}$.

2 Materials and methods

2.1. Overview of the domain and the set-up of the computations

Helsinki Metropolitan Area and its surrounding regions are situated on a fairly flat coastal area by the Baltic Sea. The climate is relatively milder, compared with most other areas in the same latitudes, caused by the warming effect of the Gulf Stream and the prevailing global atmospheric circulation. The Helsinki Metropolitan Area (HMA) comprises four cities; Helsinki, Espoo, Vantaa and Kauniainen. The total population in the Helsinki Metropolitan Area is currently approximately 1.13 million, while the population of Helsinki is about 0.63 million inhabitants. The cities in this area, and the selected measurement stations have been presented in Fig. 1.

The most important local source categories for both the concentrations of the PM mass fractions and the PM number concentrations are vehicular traffic and small-scale combustion, with smaller contributions from shipping and harbour operations, industrial sources, and aviation (Soares et al., 2014, Kukkonen et al., 2016 and Aarnio et al., 2016). The aim of this study was to evaluate the concentrations of $PM_{2.5}$ for a multi-decade period on a high temporal and spatial resolution. It was therefore necessary to simplify the computations, compared to corresponding assessments for shorter periods.

We have therefore addressed in this study explicitly only long-range transport, and local vehicular traffic and small-scale combustion. The information on the temporal evolution of shipping, the technical specifications of ships, and the engines and emissions of ships is very scarce for the earlier parts of the target period.

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Soares et al. (2014) evaluated the emissions from shipping and major stationary sources in this area for 2008. Emissions from ship traffic were modelled using the Ship Traffic Emissions Assessment Model (STEAM) presented by Jalkanen et al. (2016) and Johansson (2017). The geographical domain was selected to include all the major harbours in Helsinki. However, the secondary particulate matter from shipping was not included. They also could not include the influence of small-scale combustion. They found that over all receptor grid locations, long range transport, vehicular traffic and shipping contributed 86, 11 and 3 % to the $PM_{2.5}$ concentrations, respectively. However, the contribution of shipping to the total PM2.5 concentrations can be higher than 20 % within a distance of one kilometre of the harbours.

We have used a roadside dispersion model and a multiple source Gaussian model for evaluating the atmospheric dispersion on an urban scale; however, we have not modelled the dispersion in street canyons in detail. The computations were performed on a high temporal resolution, one hour, and on a high spatial resolution, ranging from 10 m to 200 m within the domain.

2.2 Meteorological pre-processing

Measured meteorological data was analyzed using the meteorological pre-processing model MPP-FMI that has been adapted for an urban environment (Karppinen et al., 2000a). The MPP-FMI model is based on the energy budget method of van Ulden and Holtslag (1985). The model utilises meteorological synoptic and sounding observations. The output of the model contains an hourly time series the meteorological data that is needed for the local dispersion modelling, including temperature, wind speed, wind direction, Monin-Obukhov length, friction velocity and boundary layer height. The pre-processed meteorological parameters were evaluated for all hours during the target period. However, the predicted meteorological dataset is not spatially variable; i.e., the same meteorological parameters were used for the whole of the Helsinki Metropolitan Area in the dispersion calculations.

We used the synoptic weather observations at the station of Helsinki-Vantaa airport (18 km north of Helsinki city center) and at marine stations south of Helsinki (Katajaluoto, Isosaari and Harmaja), radiation observations at Helsinki-Vantaa, and sounding observations at the station of Jokioinen (90 km northwest of Helsinki) for the whole target period. We had to use a compilation of the wind data measured at several stations located south of Helsinki in marine environments, as the time series of data at any of these stations was not complete for the whole of the target period. All three marine stations were located on islands in the Gulf of Finland.

The synoptic data used in the model was based primarily on observations at Helsinki-Vantaa. At any specific time, the predicted wind values were, however, obtained as a combination of observational data at two stations. This procedure yields a better spatial representativity of the wind values for the considered domain, compared with using only the data at one station. The sounding data included observed vertical temperature and pressure profiles.

However, after the procedure described above, the time-series of all the relevant observations was still not totally complete as input for the meteorological pre-processor. Single (hourly) missing

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observations or short intervals of observations were therefore completed, by interpolating, or replacing the missing values by the corresponding values at each previous measured time instant. There was one longer period with missing data at the station of Helsinki-Vantaa (the month of April in 1986) that was replaced by (i) the corresponding synoptic data at a station in the city Turku, and (ii) the radiation data at the station of Jokioinen.

2.3 Evaluation of the emissions in the Helsinki Metropolitan Area

2.3.1 Evaluation of the traffic flows and vehicular emissions

The emission inventory included exhaust and suspension emissions originated from vehicular traffic for the network of roads and streets in the HMA.

Evaluation of traffic flows and vehicular exhaust emissions in 2012

We have used the detailed geographical information on the line source network in the Helsinki Metropolitan Area that was provided by the Helsinki Region Transport. We also conducted a thorough checking of the accuracy of this data, based on independent information on the location of streets and roads reported by (i) Google Earth, (ii) National Land Survey of Finland and (iii) Helsinki Region Environmental Services Authority. The locations of the reported line sources were in a number of cases found to be substantially inaccurate and were therefore revised. The final 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 2008 (Helsinki Region Transport, 2011). The traffic flow data included geographical information and vehicle mileage for the above mentioned line sources within the Helsinki Metropolitan Area, for three selected hours of each day. The traffic flow data also included average driving speeds for personal cars. The hourly traffic volumes were computed using a set of regression-based factors for the diurnal variation of hourly traffic flows, separately for weekdays, Saturdays and Sundays. These factors were estimated by the Helsinki Region Transport.

We also used the data of the national calculation system for traffic exhaust emissions and energy consumption in Finland, called LIPASTO (Mäkelä and Auvinen, 2009; lipasto.vtt.fi). The system comprises two separate units, the emission inventory part and the unit emission database part. The LIPASTO system also includes data on mileages. The database contained city-level data for years 2012-2014, and the trends of emissions and mileages since 1980. The emission factors of vehicles are mainly based on national laboratory measurements. The LIPASTO system is similar, e.g., to the corresponding system that is used in the U.K. (AEA, 2009). The mileage trend data was used to scale the 2008 traffic volume data to evaluate traffic flows in 2012.

Separate emission factors were used in this study for various classes of vehicle types, including personal vehicles, vans, buses, and a combination of lorries and trucks. However, the emissions from motorcycles and mopeds were not included. In case of personal vehicles, we also allowed for differing

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emissions for (i) the streets within the city centers and other highly populated areas, and (ii) the roads in suburban areas and in the outskirts of the most populated urban areas.

Evaluation of traffic flows and vehicular exhaust emissions for the whole target period

We have used the inventory of traffic flows and traffic exhaust emissions in 2012 as a starting point for evaluating the traffic flows and emissions for the whole target period. The emissions were scaled for the other years using the data on the annual values of the emissions within the Helsinki Metropolitan Area contained in the LIPASTO system.

The emission inventory for 2012 was scaled for the whole target period, by scaling the emission in each road link. The scaling was done by multiplying the emissions in 2012 by the ratio of the total exhaust emission values in the Helsinki Metropolitan Area in any selected year to that in 2012. The total exhaust emissions in the area were extracted from the LIPASTO system. However, when completing this procedure, one needs to assume that the spatial distribution of the traffic exhaust emissions does not substantially change from year to year.

Evaluation of the emissions of suspended dust for the target period

It was not possible to use detailed suspension emission models, such as the NORTRIP or FORE models (Kauhaniemi et al., 2014) for such a long period. The input values for such models regarding, e.g., road sanding and the use of studded tyres were not available for the whole target period. We have therefore used a simpler semi-empirical modelling approach for evaluating the emissions of suspended dust. First, we evaluated the fractions of suspended particle emissions based on model computations for a couple of recent years, for which the detailed input information was available. Second, we evaluated the evolution of these fractions for the whole period.

We computed both the $PM_{2.5}$ concentrations originated from traffic exhausts, using the CAR-FMI model, and the suspended $PM_{2.5}$ concentrations, using the FORE model, for the whole of the Helsinki Metropolitan Area in 2008 and 2010. We have subsequently computed the monthly ratios of the concentrations, C_{susp}/C_{exh}, in which C_{susp} is the suspended $PM_{2.5}$ concentration and C_{exh} is the corresponding concentration originated from the traffic exhausts. These ratios were computed at four stations. The monthly average ratios were then used to evaluate the traffic suspension emissions. The annual average values of the ratio C_{susp}/C_{exh} for $PM_{2.5}$ were 0.33 and 0.36 in 2008 and in 2010, respectively.

We evaluated the deviations of the above described simpler semi-empirical method, in comparison with the computations by the FORE model for three years, 2012-2014. The predicted annual average suspended $PM_{2.5}$ concentrations at four measurement stations (Mannerheimintie, Kallio, Leppävaara and Tikkurila) computed with the semi-empirical method ranged from 67 to 98 % of the predictions by the FORE model. The monthly averaged variation of the suspended concentrations was slightly more moderate, using the simpler model.

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The computations using the FORE model allow for the suspended dust originated from sanding, studded tyres and the wear of the road surfaces (including especially the wear caused by the studded tyres). However, the emissions of brake and clutch wear are not explicitly included. Kupiainen et al. (2015) have estimated that the particulate matter emissions for tyre, brake and pavement wear in the Helsinki Metropolitan Area were less than a third of the total vehicular non-exhaust emissions. The model computations include part of this fraction, i.e., the wear due to studded tyres. In summary, the resulting inaccuracy in the predicted vehicular non-exhaust emissions can therefore be estimated to be smaller than a couple of tens of per cent.

The estimation of suspended dust emissions for the other years was done using the emissions and traffic volumes, obtained from the LIPASTO system. The ratio of vehicular suspension and exhaust emissions during year y is by definition

$$R(y) = \frac{e_{susp}(y)}{e_{exh}(y)} \qquad (1),$$

where e_{susp} and e_{exh} are the suspension and exhaust emissions, respectively.

For simplicity, we assume that the total emission from vehicular suspension sources is directly proportional to the total traffic volume, both evaluated within the considered domain for each year. The annual vehicular suspension emission in the domain is also dependent on other factors, especially on the fraction of studded tyres in cars, characteristic traffic speeds and the types of street surfaces used. However, sufficiently detailed information on these factors for the whole of the target period was not available.

Equation (1) can therefore be written as

$$R(y) = \frac{\frac{V(y)}{V(T)} e_{susp}(T)}{e_{exh}(y)} \qquad (2),$$

Where V(y) and V(T) are the traffic volumes during the years y and any other year T. T can be selected to be the target year, 2012.

Using equation (1) to rewrite $e_{susp}(T)$ yields

$$R(y) = \frac{V(y)}{V(T)} R(T) \frac{e_{exh}(T)}{e_{exh}(y)} \qquad (3).$$

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In equation (3), the ratios of the traffic volumes V(y)/V(2012) and those of the exhaust emissions $e_{susp}(2012)/e_{exh}(y)$ can be computed. The ratio of vehicular suspension and exhaust emissions during the target year R(2012) is also known, based on the above mentioned evaluation.

The vehicular suspension emissions can therefore be computed for any historical year (y) based on equation (3), if the exhaust emissions and the total traffic volume during that year are known. We have used this approach for evaluating the suspension emissions for all the other years, except for 2012.

2.3.2 Evaluation of the emissions from small-scale combustion

A novel emission inventory was compiled for 2014 for the following pollutants: particles (PM_1, $PM_{2.5}$ and PM_{10}), nitrogen oxides (NOx), non-methane volatile organic compounds (NMVOC), carbon monoxide (CO), black carbon (BC) and benzo(a)pyrene (BaP). For a more detailed description of this inventory, the reader is referred to Hellen et al. (2016) and Kaski et al. (2016).

The procedures and habits of combustion, and the amount of combusted wood were estimated by a questionnaire (Kaski et al., 2016a). Wood combustion was seldom used as a primary heating method (only in approximately 2 % of the houses); it was much more common both as a supplementary heating method and as a fuel in the stoves used in saunas. In total, wood combustion was used as either a primary or secondary heating method in approximately 90 % of the detached and semidetached houses in the area.

The small-scale combustion that is not using oil or gas in this domain uses almost solely wood as a fuel. The average amount of wood burned per house was annually 1.52 solid cubic meters. Most of the wood was used in heat-storing masonry heaters (0.72 solid-m^3/house) and sauna stoves (0.31 solid-m^3/house); only a minor amount was burned in heating boilers (0.09 solid-m^3/house) and other devices.

We used the wood combustion emissions in 2014 as a starting point. The total emission values for the other years were estimated by using previous available emission surveys and various statistical data. In addition to the above mentioned survey for 2014, the Helsinki Region Environmental Services Authority has estimated the amount of wood combustion in the area for 2002. This survey was conducted using the results of the previous corresponding studies in this domain, and various statistical data. In addition, the emissions were evaluated for 2007 by a questionnaire.

For estimating the amount of wood combustion, it is essential to know the number of detached and semidetached houses, the amount of firewood used, the shares of primary heating sources, and the numbers of boilers and sauna stoves. The numbers of detached and semidetached houses were extracted from the Helsinki Region Trends statistical database, and from the statistical data reported by the urban municipalities in this area. The estimate on the amount of firewood used is based both on the above mentioned questionnaire survey for the year 2014, and on a survey by the Forest Research Institute in Finland for the years 1992 and 1993, and 2000 - 2001. The shares of primary

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heating sources are based on statistical data and the above-mentioned surveys by the Helsinki Region Environmental Services Authority.

The historical fractions of sauna stoves in detached and semidetached houses during previous years was evaluated partly based on Luoma (1997). The estimates in this inventory are based on a limited amount of data, regarding 300 detached and semidetached houses. According to Luoma (1997), there was a sauna stove in approximately 50 % of the detached and semi-detached houses in this area in 1980. According to more recent statistics by the Helsinki Region Environmental Services Authority, this share has decreased to 28 % in 2013. The amount of firewood used for sauna heating was evaluated based on the survey by the Helsinki Region Environmental Services Authority. The amount of heating boilers was analyzed based on Luoma (1997) and the surveys by the Helsinki Region Environmental Services Authority. For boilers, the number of heating days per year was also taken into account.

The emission distributions for the other years were evaluated by multiplying the emission values in 2014 by the ratio of the total emissions for each year and that for the year 2014. This scaling was done separately for each of the above mentioned fireplace categories (masonry heaters, sauna stoves and boilers), as their temporal trends were substantially different.

There are several challenges in modelling accurately the emissions from small-scale combustion for such a long period. The temporal changes of the locations and magnitudes of the emission sources were not known in sufficient detail to be included in the modelling. We also could not allow for the detailed changes in the types of burning appliances in time.

2.4 Atmospheric dispersion modelling

2.4.1 Urban scale dispersion modelling

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 computes an hourly time-series of the pollutant dispersion from a network of line sources. The dispersion equation is based on a semi-analytical solution of the Gaussian diffusion equation for a finite line source. The dispersion parameters are modelled as a function of the Monin-Obukhov length, the friction velocity and the mixing height. Traffic-originated turbulence is modelled with a semi-empirical treatment.

The modelling system containing the CAR-FMI model has been evaluated against the measured data of urban measurement networks for gaseous pollutants (e.g., Karppinen et al., 2000c and Kousa et al., 2001) and for $PM_{2.5}$, PM_{10} and particle number concentrations in the Helsinki Metropolitan Area (Kauhaniemi et al., 2008, Aarnio et al., 2016 and Kukkonen et al., 2016). The model has also been evaluated both against gaseous and particulate pollutant measurements in London (Sokhi et al., 2008, and Singh et al., 2013 and Srimath et al., 2016) and in Birmingham, U.K. (Srimath et al., 2016). The performance of the CAR-FMI model has also been evaluated for gaseous pollutants against the results

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of field measurement campaigns and inter-compared with other models (Kukkonen et al., 2001, Oettl et al., 2001, Levitin et al., 2005, Srimath et al., 2016).

The dispersion from small-scale combustion sources was evaluated using the UDM-FMI model (Urban Dispersion Model – Finnish Meteorological Institute; Karppinen et al., 2000b). The model is based on multiple source Gaussian plume equations for various stationary source categories (point, area and volume sources). The modelling system including the UDM-FMI model has been evaluated against the measured data of urban measurement networks for gaseous pollutants (e.g., Karppinen et al., 2000b and Kousa et al., 2001) and for $PM_{2.5}$ (Kauhaniemi et al., 2008).

In this study, the emissions from domestic wood combustion were uniformly distributed in area sources of size 100 m x 100 m. The altitude of the releases for domestic wood combustion was assumed to be equal to 7.5 m, including the initial plume rise. This altitude value is based on the average heights of the detached and semidetached houses and their chimneys within the study domain, and an estimated plume rise. The dispersion was evaluated separately for three different emission source categories: sauna stoves, boilers, and other fireplaces. The diurnal, weekly, and monthly variations within the emission inventory were applied for each source category.

Both for the dispersion of vehicular and small-scale combustion pollutants, $PM_{2.5}$ was treated as inert, i.e., no chemical or physical transformation was assumed to take place within the urban time scales.

2.4.2 Regional scale dispersion modelling

The air quality measurements that are representative for the regional background of this area have been conducted at the station of Luukki. However, these measurements were started only after more than a half of the target period had passed, in 1999 and 2004 for PM_{10} and $PM_{2.5}$, respectively. It was therefore not feasible to use the measured regional background concentrations for the whole period. Instead, we used modelled regional background concentrations, which constituted a self-consistent time series for the whole target period.

SILAM is a global-to-meso-scale dispersion model developed for evaluating atmospheric composition and air quality, and for emergency decision support applications, as well as for solving inverse dispersion problems (e.g., Sofiev et al, 2006). The model incorporates both Eulerian and Lagrangian transport routines. There are eight chemico-physical transformation modules (basic acid chemistry and secondary aerosol formation, ozone formation in the troposphere and the stratosphere, radioactive decay, aerosol dynamics in the air and transformations of pollen) and modules for three- and four-dimensional variational data assimilation. The source term descriptions of the SILAM model include point- and area- source inventories, sea salt, wind-blown dust, natural pollen, natural volatile organic compounds, nuclear explosion, as well as interfaces to the ship emission system STEAM and the fire information system IS4FIRES.(http://silam.fmi.fi/).

In the present study, we have conducted reanalyses of the atmospheric composition and air quality for the period 1980-2014 on global, European, and northern European scales, using the SILAM model (version 5.5). The simulations were conducted in three spatial domains, i.e., global (the troposphere and the stratosphere), European (troposphere) and Northern European ones (troposphere). However,

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as a combination of meteorological inputs was used for computing the concentrations in the Northern European domain, these computations did not provide a completely homogeneous time series. We therefore used the corresponding results for the European domain as boundary conditions for the urban scale computations.

An overview of the global and regional computations is presented in the following. For a more detailed description, the reader is referred to Sofiev et al. (2018).

The global reanalysis was done using a 1.44°×1.44° longitude-latitude resolution, for both the troposphere and the stratosphere, for the period 1980-2014. The meteorological data was the ERA-Interim archive taken at full resolution, 0.72°×0.72°, and the vertical coverage included 61 hybrid levels (Dee et al., 2011; Simmons et al., 2010). For the global and European reanalysis, we have used the following emission data: MACCity and EDGAR (EDGAR 2014) for the anthropogenic PM emissions (Janssens-Maenhout et al., 2017), ACCMIP for wild-land fires (Lamarque et al., 2010, Granier et al., 2011), MEGAN for biogenic emissions (Guenther et al., 2006), and GEIA for the emissions from lightning (Price et al, 1997) and aircrafts. The emissions of sea salt (Sofiev et al., 2011), wind-blown dust, and biogenic VOC's (for the European and Northern European domains; Poupkou et al., 2010) were evaluated using the embedded modules in SILAM.

The model output on global scale contains hourly surface and column-integrated concentration, and column aerosol optical depth (AOD), as three-hourly values at 18 model levels. There were 50 chemical species in the model output.

The European reanalysis was done using a longitude-latitude resolution of 0.5°×0.5°, for the troposphere, for the period 1980-2014. The meteorological input data for the dispersion modelling was the ERA-Interim for the global and European domains (Dee et al., 2011, Simmons et al., 2010). The zoomed re-analysis for Northern Europe at a resolution of 0.1°×0.1° used BaltAn meteorological reanalysis by the Estonian Meteorological Institute. As the values included in the BaltAn reanalysis extend only up to the year 2005, this dataset was expanded by using the data from the ECMWF operational archives for 2006-2014.

The model output contained hourly three-dimensional surface, and column-integrated concentration and column AOD, at 13 modelled vertical levels. The model output included 40 chemical species.

In order to obtain the time series of background concentrations, we first selected four grid points of the SILAM computations that were closest to the 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. This method is less sensitive to potential occasional local influence of the metropolitan area on the regional background concentrations, compared with using the predicted value only at one specific grid point. 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.

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3. Results and discussion

First, the predicted concentrations are evaluated against the measured values. Second, selected predicted results are presented, especially on the temporal evolution of the local emissions and urban concentrations. Third, the concentrations are analyzed in terms of the contributions originated from the various pollution source categories.

3.1 Evaluation of the predicted results against the measured concentrations

3.1.1 Statistical parameters for the comparison of predictions and measurements

Four statistical parameters were computed: the index of agreement (IA), the factor-of–two (F2), the coefficient of determination (R^2) and the fractional bias (FB). The parameters IA, R^2 and F2 are measures of the correlation of the predicted and observed time series of concentrations, whereas FB is a measure of the agreement of the predicted and observed mean concentrations.

The original form of the index of agreement (IA) is calculated as (Wilmott, 1981)

$$IA = 1 - \left[\frac{\sum_{i=1}^{n} (x_i - y_i)^2}{\sum_{i=1}^{n} \left[\left| x_i^1 \right| + \left| y_i^1 \right| \right]^2} \right]$$

where n is the number of data points, and x and y refer to the predicted and measured pollutant concentrations, respectively. The symbols with the superscript '1' are defined as:

$$x_i^1 = x_i - \bar{y} \text{; and}$$

$$y_i^1 = y_i - \bar{y} \quad ,$$

in which the overbar refers to an average value. Also other forms of IA have been presented in the literature.

The IA, as defined above, ranges from 0.0 (theoretical minimum) to 1.0 (perfect agreement). However, even for a random predicted distribution, the IA value will be larger than the theoretical minimum. Karppinen et al. (2000b) found that for a random predicted distribution varying from zero to twice the measured average concentration, the IA value was approximately 0.40.

F2 is a measure of how many predictions are within a factor of two compared with the observations. Fractional bias (FB) ranges from - 2.0 to + 2.0 for extreme under- and over-prediction, respectively. The FB values equal to - 0.67 and + 0.67 are equivalent to under-and over-prediction by a factor of two, respectively.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-968
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3.1.2 Statistical analysis of the agreement of the predicted and measured concentrations

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Regional background concentrations at one station

We first compared the predictions of the SILAM model for the regional background at the station of Luukki with the available measurements. The above mentioned statistical parameters were computed for both the hourly and daily concentrations of $PM_{2.5}$, separately for each year. Selected results for
480 the daily concentrations are presented in Table 1. The corresponding scatter plots of the predicted and measured concentrations have been presented in Figs. 2a-k.

The IA values range from 0.56 to 0.73. These fairly high or high IA values indicate that the temporal variation of the predicted daily $PM_{2.5}$ concentrations agrees well or fairly well with the observed data. There are also no evident temporal trends in the statistical parameters during this period, i.e., the
485 performance of modelling does not deteriorate for the earlier years. The fractional bias values in Table 1 indicate a slight under-prediction in most cases, the bias values ranging from – 14 to + 11 %.

Urban concentrations at four stations

We have selected the data of four urban measurement stations for the evaluation of the predicted
490 results. These stations and their classifications are as follows: Kallio2, urban background, Mannerheimintie, urban traffic, Leppävaara, suburban traffic and Tikkurila, suburban traffic. The stations of Kallio2 and Mannerheimintie are situated in central Helsinki, the station of Leppävaara is located in the city of Espoo and the station of Tikkurila in the city of Vantaa. The stations of Leppävaara and Tikkurila are also influenced by small-scale combustion from the surrounding
495 residential areas. For simplicity, these stations are referred to in the following as KAL, MAN, LEP and TIK.

The above mentioned statistical parameters were computed for both the hourly and daily concentrations of $PM_{2.5}$ at the four selected measurement stations, separately for each year. Selected results for the daily concentrations are presented in Table 2. The corresponding scatter plots of the
500 predicted and measured concentrations for one of the stations (KAL) have been presented in Figs. 3a-p.

The IA values range from 0.64 to 0.75, from 0.61 to 0.84, from 0.70 to 0.75 and from 0.71 to 0.83 at the stations of KAL, MAN, LEP and TIK, respectively. These fairly high or high IA values indicate that the temporal variation of the predicted daily $PM_{2.5}$ concentrations agrees well or fairly well with
505 the observed data. The other considered statistical values can also be considered to indicate a good or fairly good agreement. In particular, there is no deterioration of the agreement of predictions and data for earlier years, compared with the corresponding results for more recent years. The scatter plots presented in Figs. 3a-p do not show any systematic under- or over-prediction of pollutant concentrations for the majority of the predicted concentrations.

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The scatter plot of the predicted and measured annual averages is illustrated in Fig. 4. The results show that the differences of the modelled and measured annual average concentrations are less than 20 % in all the cases, except for two (Fig. 4, MAN 2010 and 2011). For three stations (KAL, LEP and TIK), there are no notable biases; however, for the station of MAN, there is a systematic under-prediction. The under-prediction could be caused by the reduced dilution caused by buildings, as this site is located in the center area of the city. The MAN site is also close to a junction of two heavily trafficked streets, and there is frequently a congestion of traffic. In particular, a substantial road re-construction work was in progress during 2010 – 2011 in the vicinity of this site; this could possibly have caused the relatively higher measured concentrations.

The indices of agreement and the fractional biases for both hourly and daily values have been presented graphically in Figs. 5a-b. As expected, the IA values are lower for the hourly values, compared with the daily values, caused by the larger scatter of hourly values, compared with the daily ones.

The agreement of the temporal variations of the predictions and measured data (as measured, e.g., by the index of agreement) is on the average slightly better for the urban stations, compared with that at the regional background station of Luukki. This could be caused by the fact that the modelling of the the temporal variations of the emissions from traffic and local small-scale combustion, compared with the emissions, transport and transformation on a regional scale.

3.2 The trends of urban emissions and concentrations

The predicted emissions originated from vehicular traffic and small-scale combustion in the Helsinki Metropolitan Area are presented in Fig. 6. The vehicular emissions were clearly higher in the 1980's and early 1990's, approximately five-fold, compared with the corresponding values during the latest years. The vehicular emissions were highest from 1986 to 1988. The relative share of suspension emissions (compared with the total vehicular emissions) is clearly higher during the most recent years.

During the considered period, the total mileage (as kilometers travelled per annum) in this area has grown continuously, except for a slight plateau during the early 1990's, from approximately 28 000 to 58 000 millions of km/a (Mäkelä and Auvinen, 2009). The mileage has also been projected to continue increasing in the foreseeable future. In summary, although the mileage has almost doubled, at the same time the vehicular emissions have decreased approximately to a fifth of their original value.

There is a slight increasing trend in the emissions from small-scale combustion. During recent years, the mass-based emissions from small-scale combustion have been only slightly smaller than those from vehicular traffic.

The temporal evolution of the modelled annual average $PM_{2.5}$ concentrations has been presented in Fig. 7 at four stations in the HMA, together with the modelled regional background concentrations. According to these computations, the highest concentrations have occurred in the early and middle years of the 1980's. In 2014, the concentrations have decreased to about a half of the highest values

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during the considered period. This has been caused both by the decreasing regional background and the decreasing local contributions.

The regional background is clearly the largest fraction of the total concentrations. The concentrations were systematically highest at the stations with the most dense traffic in the vicinity of the station (MAN and LEP), and relatively lowest at the urban background station of Kallio. However, during the last decade, there has been only a very slight decreasing trend.

3.3 The contributions originated from various source categories

The modelled urban source contributions (vehicular exhaust, vehicular suspension and small-scale combustion) have been presented in Figs. 8a-d. For clarity, the regional background values have not been presented in this figure. The local contributions range from 0.5 to 4.5 μg/m^3. Vehicular exhaust has commonly been the largest urban contribution. However, at the stations that are most directly influenced by small-scale combustion (TIK and LEP), that contribution is also comparable with that of vehicular sources, during the most recent years.

4. Conclusions

We have modelled the concentrations of fine particulate matter globally, in Europe and in the Helsinki Metropolitan Area for a period of 35 years, from 1980 to 2014. The present study addresses the results in case of $PM_{2.5}$ for the urban area. Results on urban concentrations have not previously been presented in the literature on such a high spatial (tens or a couple of hundreds of meters) and temporal (hourly) resolution for several decades. These long-term results and trends were also thoroughly evaluated against the measured data at one regional background station and at four urban stations. The results were subsequently post-processed to provide for the life-time concentrations of $PM_{2.5}$ for the members of the Espoo Cohort Study. We have allowed for the changes of air quality, caused by the changes of the residences, separately for each cohort member.

Clearly, there are numerous challenges in modelling accurately the emissions and atmospheric dispersion for such an extended period. Various factors affecting the relevant emissions, the emission coefficients and the absolute amounts of emissions are known less accurately for the earlier period, i.e., especially for the 1980's and also for the 1990's. This is the case regarding both the global and European scale emission inventories, and the urban scale inventories.

We have examined the available quantitative information on the changes of the street and road network in the considered area, but that information was not sufficiently detailed to be included in the emission modelling. The fractions and types of studded tires used in vehicles in the 1980's and 1990's were not known sufficiently accurately to be included in the modelling. It was also not possible to use more detailed models, such as those for evaluating the vehicular suspension emissions, and those for street canyon dispersion, due to missing input data (in the former case) and practical

limitations on the amount of computations (in the latter case). The regional scale computations rely on the data of several previously compiled emission inventories; the accuracy of these datasets could be worse for earlier decades.

The modelled concentrations of $PM_{2.5}$ agreed fairly well or well with the measured data at four measurement stations, during 1999 – 2014. These stations represented regional background, urban background, urban traffic and suburban traffic environments. Two of these stations were also influenced by small-scale combustion from the surrounding residential areas. However, we did not find any systematic deterioration of the agreement of predictions and data for earlier years (the 1980's and 1990's), compared with the corresponding results for more recent years (2000's and early 2010's). This gives more confidence that the emission inventories used for the earlier years were not substantially more inaccurate, compared with those for the more recent years.

The measured concentrations of $PM_{2.5}$ were slightly under-predicted for most years at the regional background station. However, these were not systematically under- or over-predicted for most of the urban stations and years. There was a systematic under-prediction of concentrations for one station, the Mannerheim St. in central Helsinki. Part of this under-prediction was probably caused by the reduced dilution caused by buildings and the increased vehicular emissions resulting from the frequently congested traffic conditions. The road re-construction works during a couple of years could also have caused an additional contribution to the measured values.

The local vehicular emissions of $PM_{2.5}$ were about five-fold higher in the 1980's, compared with the emissions during the latest considered years. This substantial decrease of vehicular emissions has been achieved, although during the same period, the total mileage (as kilometers driven per annum) in the area has almost doubled. During the same period, the emissions originated from small-scale combustion have slightly increased in time. The highest concentrations of $PM_{2.5}$ have occurred in the early and middle years of the 1980's. The urban concentrations of $PM_{2.5}$ have decreased to about a half of their highest values during the considered period. However, during the last decade, there has been only a very slightly decreasing trend.

The regional background constitutes clearly the largest fraction of the total concentrations. Vehicular exhausts have commonly been the most important local source, but the relative share of small-scale combustion has continuously increased.

The study has provided high-resolution concentration databases on global, European and urban scales. The global and regional scale data is available to be used for similar smaller domain, high-resolution assessments worldwide. The urban scale data has already been used for the assessment of individual-level life-time exposure to air pollution and its effects, especially with respect to the risk of developing asthma during the first three decades of life in the Espoo Cohort Study (ECS).

5. Acknowledgements

This study has been part of the research projects Global health risks related to atmospheric composition and weather (GLORIA), funded by the Academy of Finland, "Understanding the link

between Air pollution and Distribution of related Health Impacts and Welfare in the Nordic countries", project #75007 (NordicWelfAir), funded by Nordforsk, and "Environmental impact assessment of airborne particulate matter: the effects of abatement and management strategies" (BATMAN), funded by the Academy of Finland. The funding from these two agencies is gratefully acknowledged. We also wish to thank Helsinki Region Transport (HSL) for the traffic flow data.

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Table 1. Selected statistical parameters associated with the agreement of the measured and predicted daily averaged concentrations of $PM_{2.5}$, at the regional background station of Luukki, from 2004 to 2014. The average values of the measurements and predictions, and the numbers of data points have also been presented.

	Luukki (daily values)	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014
IA	Index of agreement	0.67		0.58		0.73	0.67	0.57	0.63	0.68	0.56	0.59
F2	Factor-of-two	61		52		54	61	57	54	64	55	57
R2	Correlation coefficient squared	0.26		0.12		0.44	0.24	0.10	0.16	0.26	0.11	0.18
FB	Fractional bias	-0.06		-0.03		-0.06	-0.01	-0.14	-0.10	-0.04	0.03	0.11
AvgCp	Average of predicted data	7.6		8.1		7.0	7.1	7.4	6.9	6.7	6.5	8.2
AvgCo	Average of observed data	8.1		8.3		7.4	7.2	8.5	7.6	7.0	6.3	7.3
N	Number of datapoints	365		359		364	364	359	357	348	364	363

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Table 2. Three selected statistical parameters associated with the agreement of the measured and predicted daily averaged concentrations of $PM_{2.5}$, at four urban measurement stations, from 1999 to 2014. The average values of the measurements and predictions, and the numbers of data points have also been presented.

Kallio2		**1999**	**2000**	**2001**	**2002**	**2003**	**2004**	**2005**	**2006**	**2007**	**2008**	**2009**	**2010**	**2011**	**2012**	**2013**	**2014**
IA	Index of agreement	0.75	0.70	0.72	0.71	0.71	0.66	0.81	0.69	0.75	0.77	0.75	0.65	0.75	0.69	0.64	0.71
F2	Factor-of-two	80	81	78	72	71	70	73	64	62	56	73	67	63	66	66	65
FB	Fractional bias	-0.01	0.12	0.01	0.02	0.14	0.06	0.07	-0.12	-0.12	-0.11	-0.07	-0.08	-0.03	-0.06	0.01	0.07
AvgCp	Average of predicted data	10.2	9.2	8.5	9.2	10.6	8.6	9.6	9.0	8.0	7.7	7.9	8.3	7.5	7.0	7.1	8.7
AvgCo	Average of observed data	10.3	8.2	8.4	9.0	9.2	8.1	8.9	10.0	9.0	8.6	8.4	9.0	7.8	7.4	7.0	8.1
N	Number of datapoints	347	366	365	365	365	366	360	365	365	361	365	363	358	363	364	365
Mannerheimintie								**2005**	**2006**	**2007**	**2008**	**2009**	**2010**	**2011**	**2012**	**2013**	**2014**
IA	Index of agreement							0.84	0.66	0.73	0.77	0.75	0.63	0.73	0.71	0.61	0.72
F2	Factor-of-two							74	67	68	64	71	61	60	69	65	63
FB	Fractional bias							-0.07	-0.15	-0.12	-0.14	-0.18	-0.24	-0.22	-0.14	-0.15	-0.12
AvgCp	Average of predicted data							10.1	9.5	8.6	8.2	8.3	8.7	7.9	7.5	7.4	9.0
AvgCo	Average of observed data							10.8	11.1	9.7	9.4	10.0	11.0	9.9	8.6	8.6	10.1
N	Number of datapoints							360	365	365	365	365	365	363	356	363	364
Leppävaara4													**2010**	**2011**	**2012**	**2013**	**2014**
IA	Index of agreement												0.75	0.77	0.70	0.70	0.71
F2	Factor-of-two												77	72	74	75	68
FB	Fractional bias												0.03	-0.04	0.02	0.08	0.15
AvgCp	Average of predicted data												9.0	8.2	7.7	7.7	9.3
AvgCo	Average of observed data												8.8	8.6	7.6	7.1	8.0
N	Number of datapoints												355	364	366	363	363
Tikkurila3												**2009**	**2010**	**2011**	**2012**	**2013**	**2014**
IA	Index of agreement											0.71	0.81	0.83	0.77	0.75	0.74
F2	Factor-of-two											78	77	75	75	75	69
FB	Fractional bias											0.03	-0.09	-0.02	0.05	0.03	0.07
AvgCp	Average of predicted data											8.2	8.6	7.9	7.5	7.4	9.0
AvgCo	Average of observed data											7.9	9.4	8.0	7.1	7.2	8.4
N	Number of datapoints											365	365	365	366	363	365

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Fig. 1. The cities in the Helsinki Metropolitan Area, and the selected four measurement stations. The geographical extent of the area is approximately 35 x 30 km.

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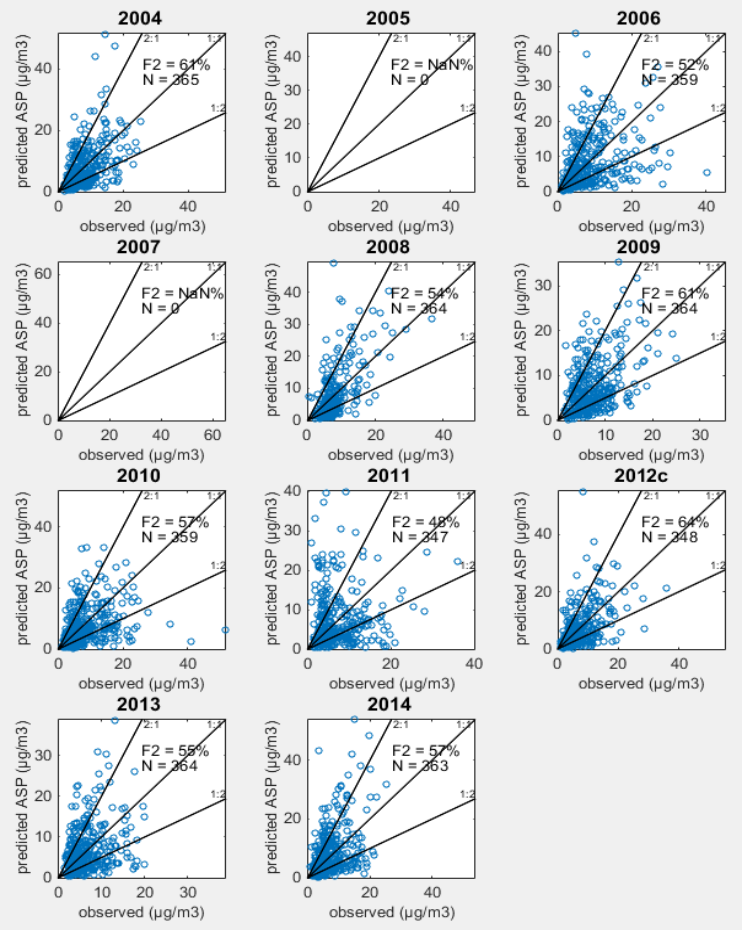

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Figs. 2a-k. The scatter plots of measured and predicted daily average concentrations of $PM_{2.5}$ at the station of Luukki, for the period 2004 – 2014. The factor of two –values and the numbers of measured days have also been presented in the panels.

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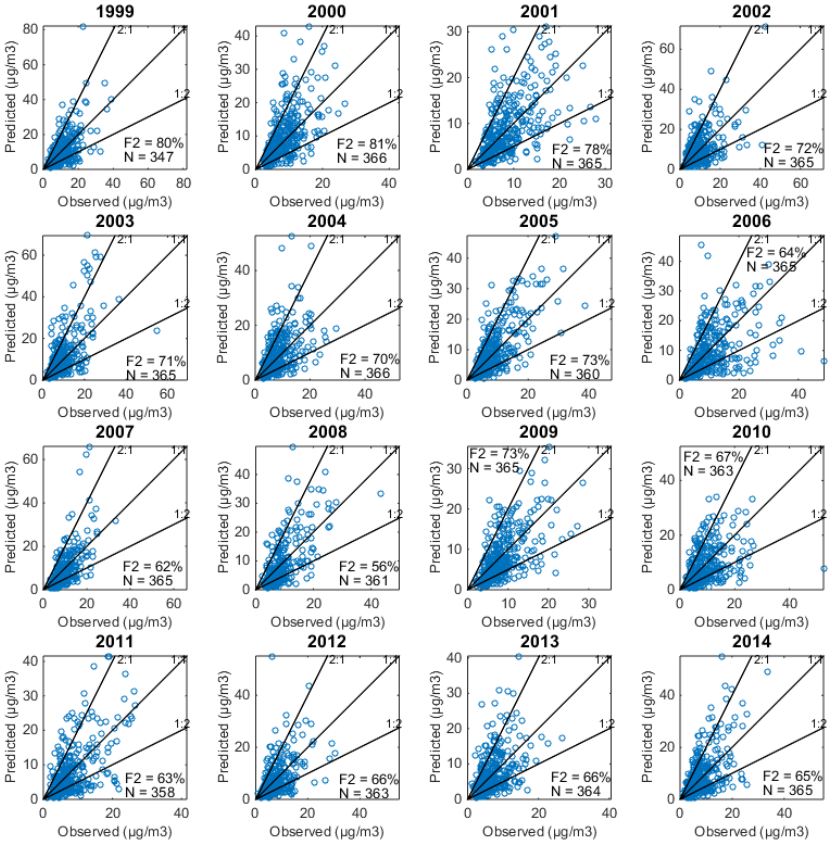

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Figs. 3a-p. The scatter plots of measured and predicted daily average concentrations of $PM_{2.5}$ at the station of Kallio2, for the period 1999 – 2014. The factor of two –values and the numbers of measured days have also been presented in the panels.

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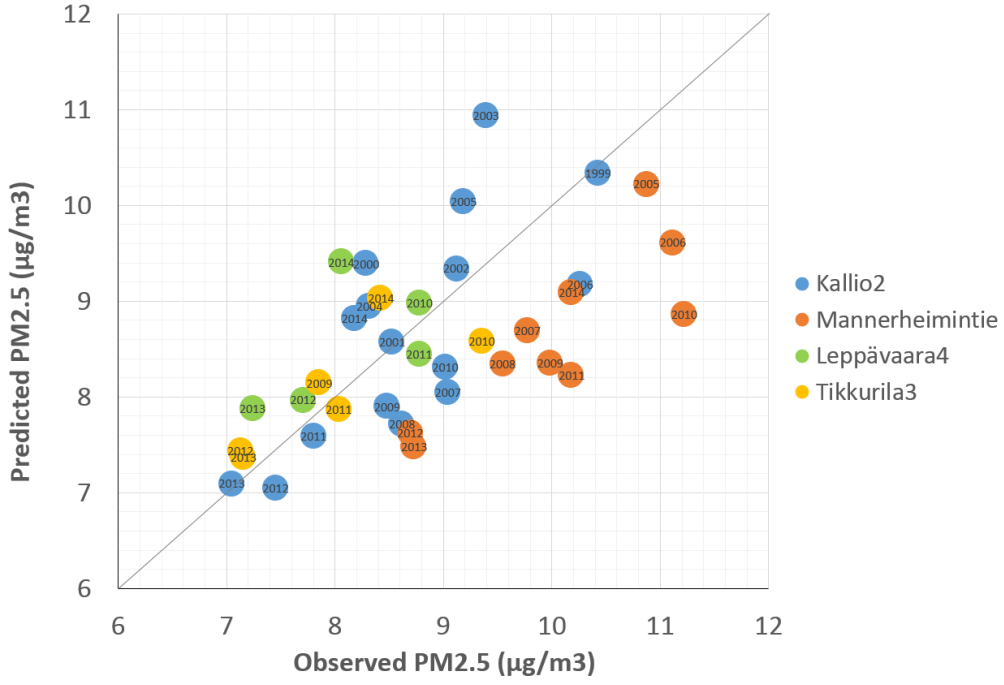

Fig. 4.The scatter plot of measured and predicted annual average concentrations of $PM_{2.5}$ at four measurement stations. The lowest values of both axes have been selected to be 6 µg/m^3.

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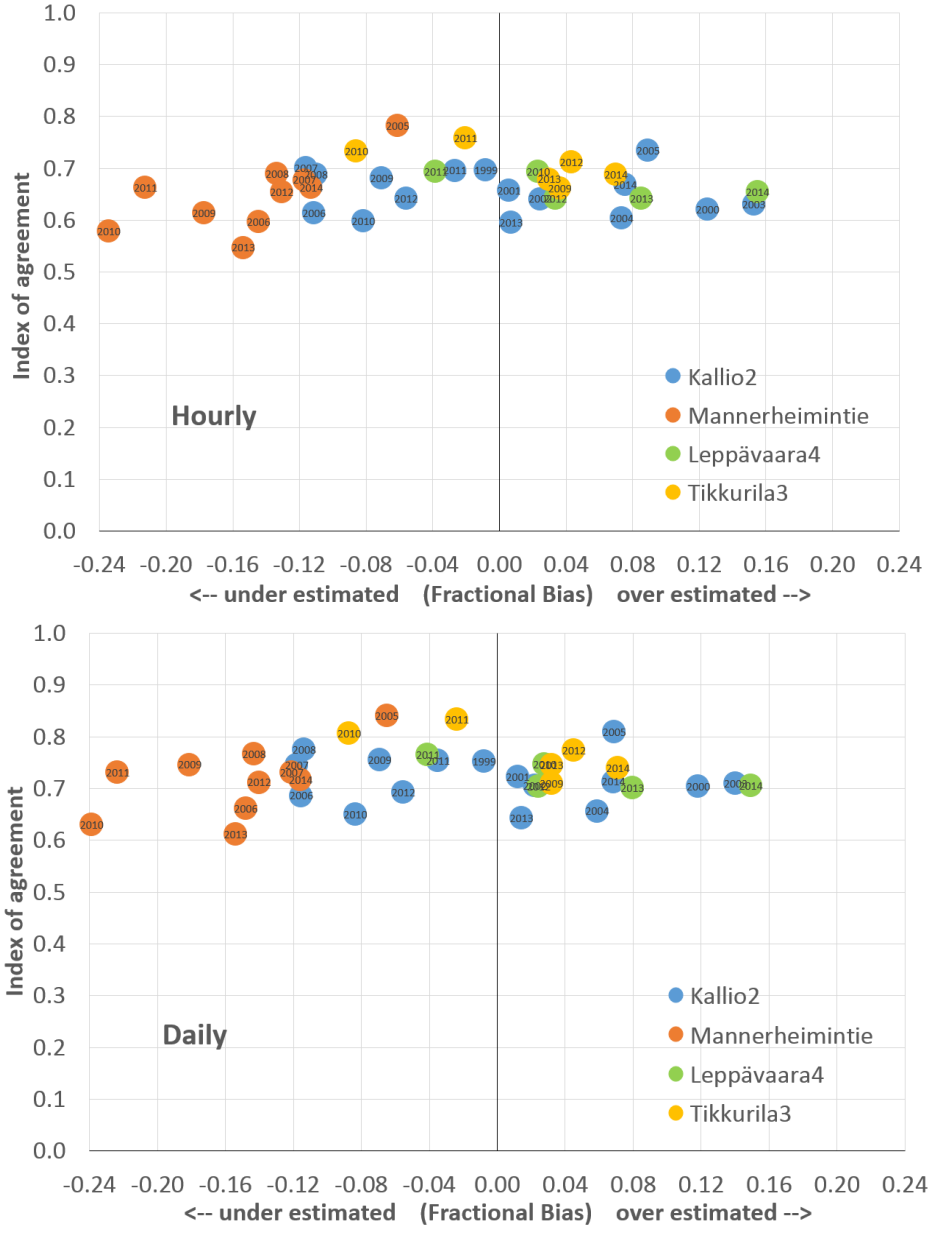

870 Figs. 5a-b. The annually evaluated indices of agreement and fractional biases, shown separately at four measurement stations and for each year, for hourly (upper panel) and daily (lower panel) concentrations of $PM_{2.5}$. The stations have been indicated by the various colours, and the years by small text inside each dot.

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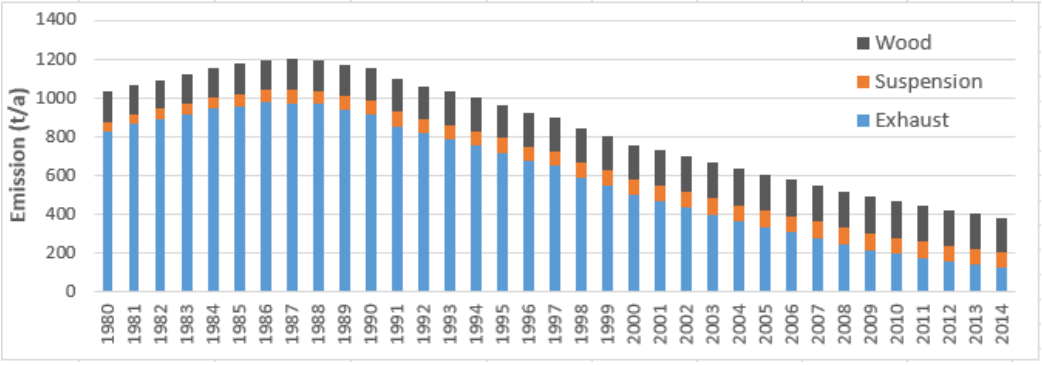

Fig. 6. The predicted emissions originated from vehicular sources and from small-scale combustion (indicated by 'Wood') in the Helsinki Metropolitan Area from 1980 to 2014. The vehicular emissions have been presented separately for suspended matter and exhaust sources. The values for the vehicular exhaust emissions have been extracted from the LIPASTO system (Mäkelä and Auvinen, 2009).

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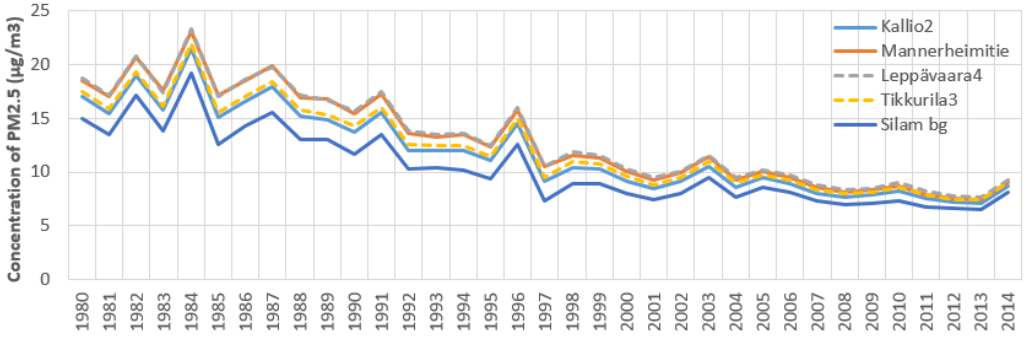

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Fig. 7. The predicted annual average concentrations of $PM_{2.5}$ at four stations in the Helsinki Metropolitan Area from 1980 to 2014, and the predicted regional background concentrations.

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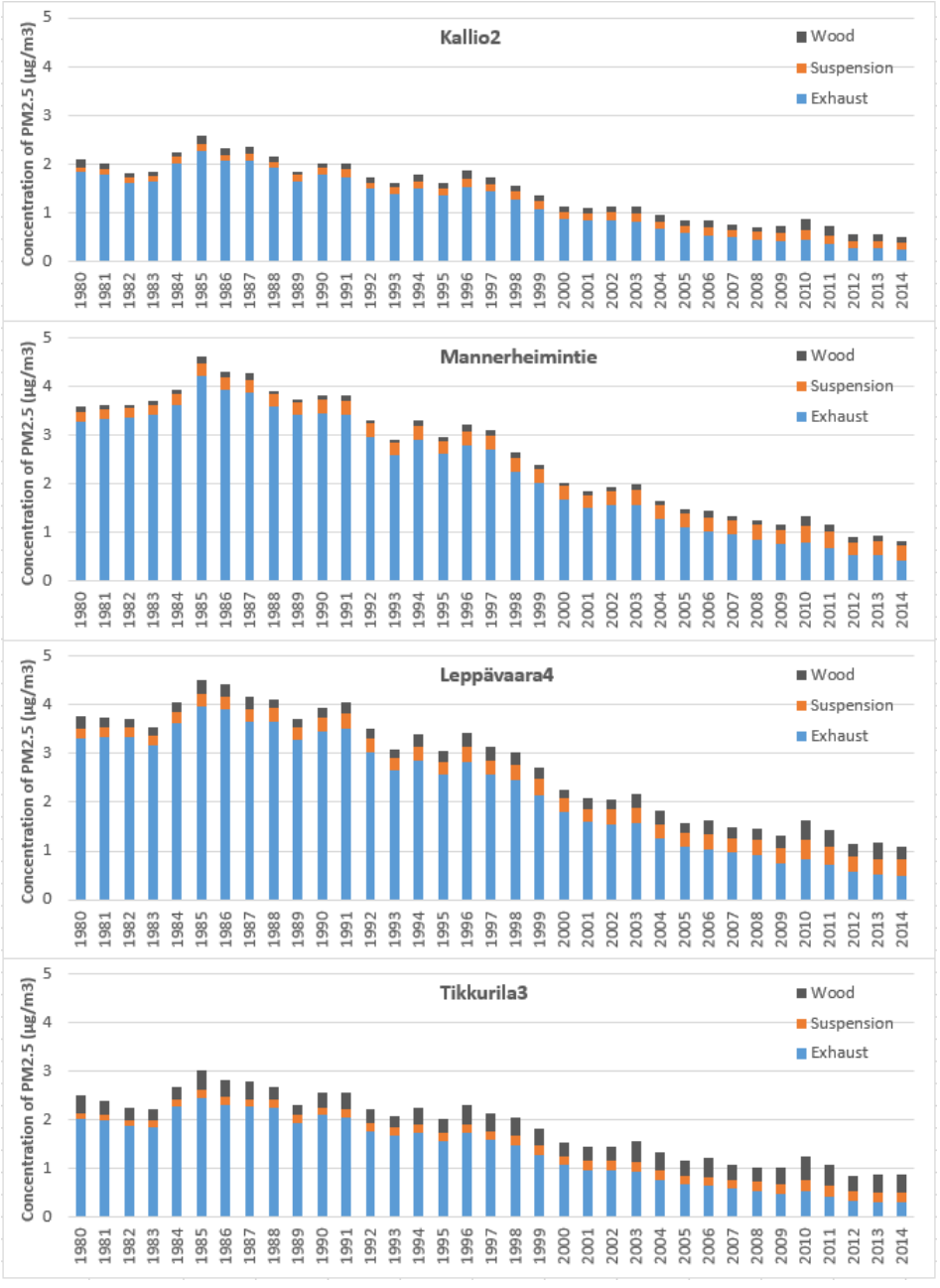

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Figs. 8a-d. The predicted annual average concentrations of $PM_{2.5}$ originated from the three most important local source categories, i.e., small-scale combustion, vehicular suspended matter and vehicular exhausts, at four measurement stations.