

# High-resolution dispersion modelling of $PM_{2.5}$ , $PM_{10}$ , $NO_x$ and $NO_2$ exposure in metropolitan areas in Sweden 2000–2018 – large health gains due to decreased population exposure

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# Abstract

Ambient air pollution remains the major environmental cause of disease. Accurate assessment of population exposure and small-scale spatial exposure variations over long time periods is essential for epidemiological studies. We estimated annual exposure to fine and coarse particulate matter ( $PM_{2.5}$ ,  $PM_{10}$ ), and nitrogen oxides ( $NO_x$ ,  $NO_2$ ) with high spatial resolution to examine time trends 2000–2018, compliance with the WHO Air Quality Guidelines, and assess the health impact. The modelling area covered six metropolitan areas in Sweden with a combined population of 5.5 million. Long-range transported air pollutants were modelled using a chemical transport model with bias correction, and locally emitted air pollutants using source-specific Gaussian-type dispersion models at resolutions up to  $50 \times 50$  m. The modelled concentrations were validated using quality-controlled monitoring data. Lastly, we estimated the reduction in mortality associated with the decrease in population exposure. The validity of modelled air pollutant concentrations was good ( $R^2$  for  $PM_{2.5}$  0.84,  $PM_{10}$  0.61, and  $NO_x$  0.87). Air pollution exposure decreased substantially, from a population weighted mean exposure to  $PM_{2.5}$  of 12.2 µg m<sup>-3</sup> in 2000 to 5.4 µg m<sup>-3</sup> in 2018. We estimated that the decreased exposure was associated with a reduction of 2719 (95% CI 2046–3055) premature deaths annually. However, in 2018, 65%, 8%, and 42% of residents in the modelled areas were still exposed to  $PM_{2.5}$ ,  $PM_{10}$ , or  $NO_2$  levels, respectively, that exceeded the current WHO Air Quality Guidelines for annual average exposure. This emphasises the potential public health benefits of reductions in air pollution emissions.

Keywords Air Pollution · Environmental Exposure · Dispersion model · Health Impact Assessment

# Introduction

Ambient air pollution is a major global health concern, estimated to cause 4.1 million premature deaths annually (Murray et al. 2020). Major adverse health consequences

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that have been linked to ambient air pollution exposure is cardiovascular disease, respiratory disease, and several types of cancer, with emerging evidence also for gestational, metabolic, renal and cognitive diseases (Thurston et al. 2017; Newman et al. 2020). Firm evidence indicates adverse health

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effects even at exposure to low concentrations of air pollution (Brunekreef et al. 2021; Strak et al. 2021; Wolf et al. 2021). Despite this, uncertainties remain regarding, e.g., the breadth of organ systems and pathologies affected by air pollution exposure, the underlying biological mechanisms, and the possibly differential toxicity of air pollutants from different sources. Given that an overwhelming majority of the world's population is exposed to air pollution concentrations exceeding the World Health Organization (WHO) recommendations, the public health implications are vast.

Assessment of long-term air pollution exposure requires accurate and precise depictions of spatial exposure contrasts. Exposure assessment that capture these variations is especially important as epidemiological studies need to account for spatially correlated confounders, such as environmental co-exposures or socioeconomic inequalities (Klompmaker et al. 2021). Although vital for air quality surveillance, monitor data mostly lack sufficient spatial resolution to adequately capture local exposure contrasts and can only indirectly disentangle different pollution sources. Land use regression (LUR) and dispersion modelling approaches have instead been used to model the spatial variation of long-term (e.g., annual) average concentrations of air pollution (Briggs et al. 1997; Hoek et al. 2008). Advantages of the dispersion strategy include higher generalisability over longer time periods, more accurate modelling of non-traffic sources, and independence of monitoring data, which can instead be used for validation (Bellander et al. 2001; Beelen et al. 2010; de Hoogh et al. 2014; Hennig et al. 2016). Dispersion modelling can also be used for source attribution and scenario calculations consistent with projected emissions, providing guidance for mitigation strategies.

Dispersion modelling-based exposure assessments in the Nordic region have been conducted for the Helsinki metropolitan area (Kukkonen et al. 2018) and for Denmark (Frohn et al. 2021; Ketzel et al. 2021). An assessment covering the whole Nordic continental region 1979-2018 was conducted by Frohn et al. (2022), but was limited to  $1 \times 1$  km<sup>2</sup> spatial resolution. Recently, two dispersion modelling-based assessments of population exposure to PM2.5, PM10, and NO2 during 2019, with estimates of health and economic impacts, have been reported for Sweden (Gustafsson et al. 2022; Alpfjord Wylde et al. 2023). Europe-wide concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, NO<sub>2</sub>, and O<sub>3</sub> in 2000–2019 were recently estimated using LUR at a resolution of 25×25 m (Shen et al. 2022). However, no longitudinal, high-resolution dispersion model has so far been developed covering a large portion of the Swedish population while allowing for the separation of different sources of air pollution.

Exposure for the Stockholm, Gothenburg, and Umeå metropolitan areas has previously been modelled for 1990–2011 (Segersson et al. 2017) and for the Malmö metropolitan area 1992–2011 (Hasslöf et al. 2020; Rittner et al. 2020). The current study extends this work both temporally and geographically to cover all study centres of the Swedish CArdioPulmonary bioImage Study (SCAPIS), a large general population cohort from six Swedish cities designed to investigate cardiovascular risk factors at the subclinical disease stage and other adverse health outcomes (Bergström et al. 2015). Our objectives were to (1) create high-resolution longitudinal models of air pollutant concentrations for the period between 2000 and 2018 covering the SCAPIS sites, to be used in future epidemiological studies of environmental exposure and health; (2) examine exposure time trends; (3) evaluate spatial differences and compliance with the WHO Air Quality Guidelines; and (4) perform a health risk assessment to estimate the impact of the change in PM<sub>2.5</sub> exposure during the model period.

# **Material and methods**

We modelled concentrations of fine particulate matter  $(PM_{25})$ , coarse particulate matter  $(PM_{10})$  and nitrogen oxides  $(NO_x)$  for the areas presented in Fig. 1 for the years 2000, 2011, and 2018. We included six modelling areas, corresponding to and named after the SCAPIS sites, but including substantial rural areas surrounding the cities. Concentrations were estimated using a combination of dispersion modelling at regional and local scale. Modelling for the Umeå, Linköping and Gothenburg areas was performed by the Swedish Meteorological and Hydrological Institute (SMHI), while modelling for the Stockholm and Uppsala areas was performed by Stockholms Luft- och Bulleranalys (SLB-analys) and for the Malmö area by the Environmental Department of the City of Malmö. Input data and methodology were harmonised between the modelling areas, with remaining significant differences described below.

# Local contributions

#### Dispersion modelling at local scale

Gaussian-type dispersion models were used to model the local contribution, with dynamic resolutions up to  $50 \times 50$  m depending on proximity to emission sources. Emissions were represented as line, point, or area sources. The Next Generation Gaussian Model (NG2M) was used for the Gothenburg, Linköping, and Umeå modelling areas. NG2M is a Gaussian-type model based on Olesen et al. (2007), Omstedt (2007), and Segersson (2021). The Airviro Gauss model (Apertum IT AB, Linköping, Sweden) was used for the Stockholm and Uppsala modelling areas, while Aermod within the Enviman system (Rittner et al. 2020) was used for the Malmö area. Source-specific contributions were estimated for emissions from road traffic exhaust, road traffic non-exhaust, small scale residential heating (mainly wood combustion), domestic and international shipping, and other **Fig. 1** Geographic location of the modelling areas within Sweden, covering major metropolitan areas



sources (diffuse emissions from mobile machinery; industrial, power, and district heating facilities; product usage; waste management; and agriculture).

Road traffic volumes, fleet composition, and time variations were described using municipal data, where available, supplemented with national statistics and traffic modelling from the Swedish Transport Administration. Emission factors for vehicle exhaust were obtained from the Handbook of Emission Factors for Road Transport (HBEFA) 4.1 (www. hbefa.net). Non-exhaust emissions from traffic, including road, break and tyre wear as well as resuspension of previously deposited dust were described using the NORTRIP model (Denby et al. 2013a, b). The NORTRIP model was applied to describe the emissions from all roads with significant traffic. The model utilises a large number of input parameters, including road pavement, traffic conditions, and road maintenance. The parameter values applied used model defaults and the reference runs included in NOR-TRIP as starting point. Road configurations were determined based on the national road database of the Swedish Transport Administration and road maintenance activities (e.g., ploughing and salting) were modelled using the builtin routines in the NORTRIP model. A simplified approach was applied for Stockholm and Uppsala, where NORTRIP reference runs were used to obtain emission factors for road traffic non-exhaust particles based on road speed and the percentage of studded tyres at each road.

Emissions from residential heating were derived from municipal inventories of heating appliances provided by chimney sweepers, containing information on location and type of appliance. Assumptions regarding fuel consumption and firing habits were based on surveys available for some areas (Omstedt et al. 2014). For municipalities where chimney sweeper data could not be obtained (some municipalities on the outskirts of the Gothenburg and Linköping modelling areas, as well as for all municipalities within the Stockholm and Uppsala modelling areas), proxy data (buildings and building types) were used. The relation between proxy data and stoves and boilers was assumed to be the same as for nearby municipalities with available chimney sweeper data. Energy balances from Statistics Sweden were used to describe trends over time.

Major industrial and power production facilities were represented as point sources. Emissions from smaller facilities were either represented as point-sources or, in absence of exact coordinates, gridded with a resolution of  $1 \times 1$  km<sup>2</sup>.

# Meteorological data

For the Linköping, Umeå, and Gothenburg modelling areas, a gridded re-analysis of the meteorological conditions was used as input to the local scale dispersion modelling. Hourly wind speed, wind direction at 10 m above ground and air temperature at 2 m above ground were retrieved from the SMHI meteorological analysis system MESAN (Häggmark et al. 1997), while global radiation from the SMHI radiation analysis system STRÅNG (strang.smhi.se). For the Stockholm and Uppsala modelling areas, a climatological model based on meteorological measurements was used as input to the dispersion model. These measurements included horizontal and vertical wind speed, wind direction, temperature, temperature difference between the three levels and solar radiation, from a 50-m-high mast in Högdalen in Stockholm as well as measurements from a 24-m-high mast in Marsta (north of Uppsala) from the period 1998 to 2019. The meteorological measurements were used as input for a simplified wind model based on Danard (1977) integrated in the Airviro system, which carries out a mesoscale interpolation of wind conditions and takes local surface roughness into consideration. In Malmö, calculations were based on hourly meteorological data from a 24 m high mast next to a large open area (Heleneholm meteorological monitoring station, south-eastern Malmö), which represented the whole modelling area.

#### Inter-annual variation

For the years between the dispersion modelling years (2000, 2011, and 2018), the local contribution was interpolated. A linear interpolation was first performed to reflect a gradual change in emissions. This interpolation was adjusted for inter-annual variation due to local meteorological conditions by multiplication with a meteorological ventilation index. For all modelling areas this meteorological index was created using time-series modelling of the period 2000–2018 at selected locations. For Linköping, Umeå and Gothenburg, a timeseries of gridded yearly ventilation indices was constructed, based on modelling at 40 selected locations distributed across Sweden and then interpolated using ordinary kriging. For Malmö, Stockholm and Uppsala, the same ventilation indices were applied for the cities' whole model domains.

#### Long-range transport

## **Chemical transport modelling**

To estimate regional background concentrations of PM and  $NO_x$ , the MATCH model, covering Europe with 44 × 44 km<sup>2</sup> resolution, was applied for all years and nested to a domain at 5×5 km<sup>2</sup> resolution covering Sweden. For meteorological forcing, the HIRLAM reanalysis EURO4M (Dahlgren et al. 2016) and operational weather data for 2013–2015 (11×11 km<sup>2</sup> resolution), as well as operational weather data from the SMHI forecast model HARMONIE

for years 2016–2018 were used. Details on the model configuration and input data have been described by Ciarelli et al. (2019).

#### **Bias correction**

Data from regional background stations were used to bias-correct the modelled results. Data for Sweden were obtained from the national air quality data host (smhi.se/ data/miljo/luftmiljodata), while the EBAS service (ebas. nilu.no) was used for neighbouring countries. At each monitoring station, the bias correction was calculated as the difference between modelled and measured concentrations. To avoid amplification of short, local episodes, the corrections were calculated based on daily average concentrations instead of the hourly measurements. The corrections were then interpolated to hourly values before application. For the areas between monitoring stations the correction value was interpolated onto the model grid using the ordinary Kriging method. The interpolated correction was added to the modelled results yielding biascorrected regional concentrations.

Lack of regional background  $PM_{2.5}$  and  $PM_{10}$  measurements in the Umeå modelling area resulted in overestimation of the long-range contribution for parts of the time-period. In order to reduce inconsistencies over time, measurements 2002–2008 at Vindeln, 50 km west of Umeå, were projected back and forth in time following Segersson et al. (2017).

# Emissions

A comprehensive compilation of emissions in all Nordic countries from the Nordic WelfAir project was used (Geels et al. 2020; Paunu et al. 2020, 2021). Large point-sources were described individually, while other sources were gridded with a spatial resolution of  $1 \times 1$  km<sup>2</sup>. In general, the emissions were consistent with the official emissions reported according to the United Nations Economic Commission for Europe (UNECE) convention on Long-range Transboundary Air Pollution (LRTAP), but the reported emissions have a spatial resolution limited to  $0.1^{\circ} \times 0.1^{\circ}$ (4-6 km (East-West)×c. 11 km (North-South)). Exceptions are emissions from small scale residential heating, where the spatial proxy-data used in the gridding was improved by including information from chimney-sweeper registers (Paunu et al. 2021) and shipping emissions, which were modelled separately using STEAM II (Jalkanen et al. 2012) for 2000-2014 and extrapolated forward to 2018. The inventory describes emission every 5 years up to 2010, and then every 2 years, requiring interpolation for unrepresented years.

## The non-local contribution

A new scheme, Back-trace Upwind Diffuse Downwind (BUDD), was applied to remove the contribution of local sources from the regional background concentration fields. BUDD has been described in Segersson (2021). Briefly, BUDD uses hourly fields of meteorological parameters and concentrations and operates over a rolling window covering an area of  $15 \times 15$  km<sup>2</sup>. Starting from the grid-cell at the centre of the rolling window, a trajectory is followed backwards in the wind field until reaching the boundary of the window. At the point and time where the boundary is reached, a vertical profile is interpolated from the background concentration fields. A vertical diffusion equation is then solved for the vertical profile, describing how the vertical profile would have developed if no local emissions would have been added along the trajectory. The resulting concentration field does not include contribution from sources within 15 km.

In the calculations for the Stockholm and Uppsala modelling areas, the local contribution was not limited to 15 km, but instead included the combined modelling areas. A simplified horizontal distribution of the long-range transport was therefore applied to avoid local maxima resulting from the emissions within the modelling area. It was constructed based on spatial mean values from a region south and north of the calculation area and assuming a north-south linear gradient in between.

# **Model validation**

The results of the air pollution modelling (i.e.,  $PM_{2.5}$ ,  $PM_{10}$ , and  $NO_x$ ) were validated against data from monitoring stations. All monitoring stations located within the modelling areas using continuous monitoring of sufficient data quality to capture urban or regional background concentrations were included. Street-level monitoring stations were excluded from the validation as these are not comparable to the modelled urban background concentrations. Regional measurements that were used for bias-correction of the longrange contribution were also excluded. In total, we used data from 20 monitoring stations across the modelling areas for the model validation. Measurement data from the stations were downloaded from the Swedish national database for air quality measurements (SMHI 2023), where data have been validated and reviewed.

Measurement series for the entire modelled period (2000–2018) were available in Stockholm, Uppsala, Gothenburg, and Malmö, while measurements in Linköping and Umeå have been partially conducted over the 18 years. All monitoring stations used for validation and the years for which data were available are presented in Table S1. Measurement data were most abundant for  $NO_x$ , to a somewhat lesser extent for  $PM_{10}$ , and to an even lesser extent for  $PM_{2.5}$ . Agreement

between measured and modelled concentrations was assessed using the coefficient of determination ( $\mathbb{R}^2$ ) and root mean square error ( $\mathbb{R}MSE$ ). Deming regression was used to characterise the relationship between measured and modelled concentrations, to account for errors in both values.

# Conversion of NO<sub>x</sub> to NO<sub>2</sub>

The modelled and validated  $NO_x$  levels were converted to  $NO_2$ . Since long-range transported  $NO_x$  consists almost solely of  $NO_2$ , long-range transported  $NO_2$  was set equal to  $NO_x$ . Concentrations of locally produced  $NO_2$  were converted from the total of local  $NO_x$  using the formula in Eq. 1, which was empirically derived from the best fit of the relationship between measured  $NO_2$  and  $NO_x$  for all modelling areas and for all years.

$$NO_2 = NO_x \times \left(\frac{30}{36 + NO_x} + 0.173\right)$$
 (1)

# Estimation of population and cohort exposure

We attributed modelled exposure concentrations to the entire population of the study areas to calculate population weighted exposure for each site and year. Population data on a  $100 \times 100$  m<sup>2</sup> grid were obtained from Statistics Sweden for 2018 and assumed to be stable throughout the modelling period. Population weighted mean exposure and standard deviations were calculated for each year and modelling area, for total exposure levels and for each source contribution, using Eq. 2.

$$PWME = \left. \frac{\sum (E_i \times P_i)}{\sum P_i} \right|$$
(2)

where PMWE is the population weighted mean exposure,  $E_i$  is the exposure for grid square *i*, and  $P_i$  is the population of grid square *i*. For each pollutant we assessed the number of individuals exposed to annual mean concentrations above the current WHO Air Quality Guidelines (5 µg m<sup>-3</sup> for PM<sub>2.5</sub>, 15 µg m<sup>-3</sup> for PM<sub>10</sub>, and 10 µg m<sup>-3</sup> for NO<sub>2</sub>).

For participants in the SCAPIS cohort, yearly address data for 2000–2018 were obtained from the Swedish Tax Agency. Addresses were automatically geocoded by Metria AB (Stockholm, Sweden) and manually checked and corrected for ambiguities, and coordinates used to assign yearly average exposure. Average time trends in exposure were assessed with simple linear regression models.

# Health impact assessment

We performed a quantitative health impact assessment (HIA) for each site, estimating the number of prevented deaths and years of life lost (YLL) in 2018 due to the decreased in

air pollution exposure compared to 2000, within the adult population (30–90 years) in the modelling areas. We used data on population age and gender distribution as well as age- and gender-specific expected remaining years of life on county level for the year 2018, obtained from Statistics Sweden. County-level data on total non-accidental mortality for each gender and five-year age group were obtained from the National Board of Health and Welfare and linearly imputed for each age-year. The number of prevented premature deaths due to non-accidental causes were calculated separately for each site and gender, using Eq. 3.

$$ED = MR \times p \times (RR^{\Delta x} - 1)$$
(3)

where *ED* signifies excess deaths, *MR* the mortality rate for men and women in each county, *p* the population in each age and gender group, *RR* the risk ratio, and  $\Delta x$  the population weighted change in exposure between 2000 and 2018. We used the exposure–response function for PM<sub>2.5</sub> and non-accidental mortality (RR 1.08, 95% CI 1.06, 1.09 per 10 µg/m<sup>3</sup>) from the recent revision of the WHO Air Quality Guidelines (Chen and Hoek 2020). Confidence ranges were derived from the 95% confidence intervals of the exposure-response functions. The number of YLL was calculated analogously to the number of excess deaths, multiplying the number of excess deaths for each age, gender, and modelling area with the number of expected remaining years of life for the corresponding age, gender, and county.

# Results

#### Population weighted exposure and time trends

In 2018, the modelling areas had a total population of 5.46 million individuals (53% of the Swedish population). The population weighted median exposure to PM2.5, PM10, and NO<sub>2</sub> in the modelling areas in 2000 was 12.2, 16.2, and 14.6  $\mu$ g m<sup>-3</sup>, respectively. In 2018, corresponding figures were 4.8, 11.0, and 4.9  $\mu$ g m<sup>-3</sup> (Figs. 2 and S1). PM<sub>2.5</sub> and PM<sub>10</sub> were primarily of non-local origin, with local emissions accounting only for 13.6% and 15.8%, respectively, while 58% of NO<sub>2</sub> was of local origin in 2018. Population weighted mean exposure, intra-area standard deviations, and percentage local contribution for 2018 are presented in Table 1 (corresponding figures for 2000 are presented in Table S2). The  $PM_{2.5}$  and  $PM_{10}$  exposure for the SCAPIS population closely resembled that of the general population in the modelling areas, while the SCAPIS participants had substantially higher NO<sub>2</sub> exposure.

There were substantial contrasts in exposure levels between the modelling areas, with the highest exposures generally found in Malmö. Exposures within each modelling



Fig. 2 Maps of the modelling areas with concentrations of  $PM_{2.5}$  in 2018

area were, however, relatively homogenous. Air pollutant exposures exhibited a decreasing trend in all modelling areas (Fig. 3). The mean annual decrease in population weighted PM<sub>2.5</sub> exposure ranged from 0.09  $\mu$ g m<sup>-3</sup> in the Umeå modelling area to 0.37  $\mu$ g m<sup>-3</sup> in the Stockholm and Gothenburg modelling areas. Corresponding annual mean decreases in PM<sub>10</sub> ranged from 0.12  $\mu$ g m<sup>-3</sup> in Gothenburg

to 0.29  $\mu g~m^{-3}$  in Stockholm, and in NO\_2 from 0.11  $\mu g~m^{-3}$  in Umeå to 0.50  $\mu g~m^{-3}$  in Malmö.

Residential heating was the largest local contributor to  $PM_{2.5}$  exposure in all modelling areas except Malmö (Fig. S2, Table S3). However, exposure to  $PM_{2.5}$  from local residential heating decreased at all sites during the modelling period, as did exposure from local traffic exhaust, Table 1 Population weighted mean exposure to  $PM_{10}$ ,  $PM_{2.5}$ , and  $NO_2$  and percentage local contribution in 2018

Modelling area	Total population	Mean exposure (standard deviation) in 2018 $[\mu g m^{-3}]$			Mean percentage local contribution in 2018 [%]		
		PM <sub>2.5</sub>	PM <sub>10</sub>	NO <sub>2</sub>	PM <sub>2.5</sub>	PM <sub>10</sub>	$NO_2$
Umeå	153 888	4.0 (0.3)	8.9 (1.5)	6.0 (2.5)	11.0	19.3	56.4
Uppsala	376 163	5.2 (0.3)	10.3 (0.8)	5.1 (2.5)	7.5	8.9	45.5
Stockholm	2 339 543	5.9 (0.4)	12.1 (1.1)	10.4 (3.5)	14.6	17.4	65.9
Linköping	436 912	4.9 (0.4)	12.6 (2.2)	6.5 (3.1)	10.7	19.3	56.7
Gothenburg	1 406 118	4.0 (0.5)	13.0 (2.1)	9.7 (5.4)	15.1	16.4	50.6
Malmö	744 145	7.3 (0.3)	14.4 (0.8)	13.2 (3.5)	12.5	10.8	54.6
All areas	5 456 769	5.4 (1.2)	12.5 (1.9)	9.8 (4.5)	13.6	15.8	58.0



Fig. 3 Time trends in  $PM_{2.5}$ ,  $PM_{10}$ , and  $NO_2$  exposure in the general population in each modelling area, compared to the SCAPIS population at each SCAPIS site

shipping, and other sources, while exposure to  $PM_{2.5}$  from traffic non-exhaust increased in all areas. In Malmö, the 'Other' category was the largest local source of  $PM_{2.5}$  exposure, where it included all pollution originating in the nearby Copenhagen region. Because of the large proportion of rural areas included in the Gothenburg modelling area, median exposures to traffic and shipping emissions were low compared to the other more populous and urbanised modelling areas (i.e., Stockholm and Malmö).

Consequently, the composition of the local contribution differed between modelling areas and changed over the modelling period. Notably, the proportion of  $PM_{2.5}$ attributable to traffic non-exhaust at least doubled between 2000 and 2018 in all modelling areas, while the proportion of  $PM_{2.5}$  from traffic exhaust increased in the Gothenburg, Linköping, and Umeå areas and decreased in the Malmö, Stockholm, and Uppsala areas. The proportion of  $PM_{2.5}$ exposure coming from local residential heating decreased in Umeå, while it increased in the Gothenburg, Linköping, Stockholm, and Uppsala areas.

#### **Exceedances of the WHO Air Quality Guidelines**

# In 2018, almost two thirds (64.5%) of the population were exposed to $PM_{2.5}$ concentrations above the current WHO guidelines for annual average exposure, while almost half (42.3%) were exposed to $NO_2$ and fewer than one in ten (7.8%) was exposed to $PM_{10}$ concentrations above the guidelines (Table 2). For $PM_{2.5}$ and $PM_{10}$ , there was substantial variation between the modelling areas. There was less between-site variation in $NO_2$ exposure, presumably because of the larger proportion of local contribution. In the Gothenburg, Umeå, and Linköping modelling areas individuals exposed to $PM_{2.5}$ levels above the WHO guidelines primarily resided close to or in the urban centres, while $PM_{2.5}$ concentrations exceeded the guidelines in almost the entire Stockholm and Malmö modelling areas (Figs. 4, S3 and S4).

# **Model validation**

The modelled annual average air pollutant concentrations agreed well with annual average concentrations measured at urban background stations (Fig. 5, results from the Deming regression analyses are presented in Table S4). The coefficient of determination  $(R^2)$  when comparing modelled and measured PM<sub>2.5</sub>, PM<sub>10</sub>, and NO<sub>x</sub> was 0.84, 0.61, and 0.87, respectively. The RMSE for  $PM_{2.5}$ ,  $PM_{10}$ , and  $NO_x$ was 1.5, 2.6, and 5.4  $\mu$ g m<sup>-3</sup>, respectively. The lower agreement for  $PM_{10}$  than  $PM_{2.5}$  was caused by a tendency for the model to slightly overestimate measured PM<sub>10</sub> concentrations at the Gothenburg Femman station and to slightly underestimate measured PM<sub>10</sub> concentrations at the Stockholm Torkel Knutssonsgatan station. For NO<sub>x</sub> we observed greater difference in the Umeå modelling area, where severe inversions during the winter caused substantial underestimations compared to measured concentrations. The Stockholm Norr Malma station had very low NO<sub>x</sub> concentrations as it is located in a rural part of the modelling area. For stations with complete data series 2000-2018, agreement was high across calendar years (Fig. S5), with the overall decreasing trend observed both in modelled and measured PM2.5 and NO<sub>x</sub> concentrations.

Health impact assessment

We estimated that, if  $PM_{2.5}$  exposure in 2018 had remained at the same levels as in 2000, this would have caused an additional 2719 (95% CI 2046–3055) premature non-accidental deaths annually among individuals between the ages 30 and 90 years and residing within the modelling areas. This corresponds to a 5.3% (95% CI 4.0–6.0) higher nonaccidental mortality, compared to the observed mortality in 2018. The largest estimated number of prevented deaths was in the Stockholm modelling area, while the largest fraction of deaths prevented by the decreased  $PM_{2.5}$  exposure was found in the Malmö modelling area. Prevented fractions, number of excess deaths and YLL for each modelling area are presented in Table 3.

# Discussion

We developed and validated a high-resolution dispersion model of ambient air pollution covering six metropolitan areas, representing more than half of the Swedish population, between 2000 and 2018. Population weighted average concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, and NO<sub>2</sub> in 2018 across all six modelling areas were low (5.4, 12.5, and 9.8  $\mu$ g m<sup>-3</sup>, respectively) in comparison to most countries, but a large proportion of the population was still exposed to air pollutant concentrations above the updated WHO guidelines. For example, a majority of the population (64.5%) in the modelling area was exposed to PM2.5 concentrations above the WHO guidelines. We observed a clear decreasing trend in population weighted exposures in all modelling areas. The decreasing trend was strongest for  $PM_{25}$  and  $NO_2$ and weaker for PM<sub>10</sub> exposure, and primarily caused by decreased long-range and regional contributions. We estimated that this decrease in PM<sub>2.5</sub> exposure prevented 2719 (95% CI 2046-3055) premature non-accidental deaths annually by comparing PM<sub>2.5</sub> exposure during 2018 with 2000. The validity of the modelled air pollutant concentrations was generally high, with  $R^2$  for  $PM_{2.5}$  0.84,  $PM_{10}$  0.61, and NO<sub>x</sub> 0.87.

		Population exceeding WHO 2021 guidelines				
Modelling area	Total population	$PM_{2.5} > 5 \ \mu g \ m^{-3}$	$PM_{10} > 15 \ \mu g \ m^{-3}$	$NO_2 > 10 \ \mu g \ m^{-3}$		
Umeå	153 888	0.2%	0.4%	3.8%		
Uppsala	376 163	67.6%	0.0%	6.4%		
Stockholm	2 339 543	98.6%	0.9%	47.5%		
Linköping	436 912	30.6%	15.0%	16.8%		
Gothenburg	1 406 118	5.9%	17.5%	37.8%		
Malmö	744 145	100.0%	12.5%	75.5%		
All areas	5 456 769	64.5%	7.8%	42.3%		

Table 2Population exposedto air pollutant concentrationsexceeding the 2021 WHOguidelines for annual averageexposure in 2018



# Density of population exceeding World Health Organization air quality guidelines for PM<sub>2.5</sub>

Fig. 4 Density of population exceeding the WHO 2021 guidelines for annual  $PM_{2.5}$  exposure in 2018

The modelled exposure levels from this study are in line with results from previous studies. A recent estimation of air pollution across Sweden for 2019 reported a population weighted exposure to  $PM_{2.5}$ ,  $PM_{10}$ , and  $NO_2$  of 5.21, 9.95,

and 5.08  $\mu$ g m<sup>-3</sup>, respectively (Alpfjord Wylde et al. 2023). These figures are similar but slightly lower than our estimates, especially for NO<sub>2</sub>, which is expected as Alpfjord Wylde et al. included the entire Swedish population (i.e., not



Fig. 5 Scatter plot of measured (horizontal axis) and modelled (vertical axis) yearly average air pollutant concentrations. The solid line is a reference equality line; the dashed line corresponds to the Deming regression

Table 3 Estimated number of annually prevented all-cause non-accidental deaths and years of life lost (YLL) because of the lower $PM_{2.5}$ exposure in 2018 compared to 2000 (95% confidence intervals in parentheses)	Modelling area	Prevented fraction, %	Prevented deaths	Prevented YLL
	Umeå	1.7% (1.3-1.9)	32 (24-35)	365 (276-409)
	Uppsala	5.0% (3.8-5.6)	174 (131–195)	2170 (1633-2437)
	Stockholm	5.5% (4.1-6.1)	1034 (778-1162)	13 256 (9971-14 891)
	Linköping	4.9% (3.7-5.5)	231 (174-260)	2737 (2060-3073)
	Gothenburg	5.8% (4.3-6.5)	842 (633–946)	10 275 (7726-11 545)
	Malmö	5.2% (3.9-5.9)	406 (306-456)	4906 (3691-5510)
	Total	5.3% (4.0-6.0)	2719 (2046-3055)	33 708 (25 357-37 865)

focusing on metropolitan areas) and used a coarser spatial resolution. Gustafsson et al. (2022) also calculated population weighted mean exposures to PM<sub>2.5</sub>, PM<sub>10</sub>, and NO<sub>2</sub> in Sweden 2019 of 7.2, 10.9, and 5.9  $\mu$ g m<sup>-3</sup>, respectively. Their estimate for PM2.5 is notably higher than ours, possibly because PM<sub>2.5</sub> levels were derived from PM<sub>10</sub>.

Our results are similar to the previous modelling performed within the SCAC project, covering three of the metropolitan areas (Gothenburg, Stockholm, and Umeå) 1990-2015 (Segersson et al. 2017). For 2012, population weighted mean exposures to PM2.5 and PM10 were estimated to 6.5 and 15  $\mu$ g m<sup>-3</sup>, respectively in Gothenburg, 6.5 and 14  $\mu$ g m<sup>-3</sup> in Stockholm, and 5.2 and 8.5  $\mu$ g m<sup>-3</sup> in Umeå in the SCAC models, compared to 5.6 and 13  $\mu g m^{-3}$  in Gothenburg, 5.7 and 9.4  $\mu$ g m<sup>-3</sup> in Stockholm, and 4.2 and  $8.0 \ \mu g \ m^{-3}$  in Umeå from the current modelling of the same year. The slightly lower figures from the current modelling can be attributed to the larger modelling areas, including more rural areas, as well as to methodological improvements, especially in the description of non-exhaust road traffic emissions and in the precision of emissions data for small scale residential heating. These two sources of pollution are common sources of uncertainty when assessing air pollution concentrations in the Nordic countries and in regions with similar climate. (A more detailed description of methodological differences compared to the SCAC model is provided in the supplement.)

Residential heating, traffic non-exhaust emissions, and other sources (including non-road vehicle exhaust, industry, power and district heating facilities, waste management, and agriculture, as well as, in the Malmö modelling area, crossborder transport of local emissions from Copenhagen) were the largest sources of locally emitted PM25 and increased as a percentage of PM<sub>2.5</sub> exposure in the majority of the modelled areas. This is in line with findings by Gustafsson et al. (2022). Notably, the percentage of PM<sub>2.5</sub> exposure originating from local non-exhaust emissions more than doubled in all modelling areas, while traffic exhaust decreased in absolute terms as well as in proportion of PM2 5 exposure in most areas. This is also in line with previous source-specific exposure assessments performed in Nordic cities (Molnár and Sallsten 2013; Kukkonen et al. 2020; Orru et al. 2022). Our population weighted estimate of PM<sub>2.5</sub> exposure from residential heating in Umeå in 2018 (0.20  $\mu$ g m<sup>-3</sup>) is, however, substantially lower than the corresponding estimate reported by Orru et al. for 2019 (0.93  $\mu$ g m<sup>-3</sup>), presumably because our estimate only includes residential heating emissions within 15 km from the residential address. Compared to total or exhaust emissions, these sources have received less attention in health studies, which highlights the need for further research focusing on the health effects of particles from these sources.

The clear decreasing trends in  $PM_{2.5}$ ,  $PM_{10}$ , and  $NO_2$  exposure corroborates similar findings from previous studies in Sweden (Olstrup et al. 2018; Gustafsson et al. 2022) and at the European level (European Environment Agency et al. 2020; Shen et al. 2022). Investigating time trends in urban background levels using monitoring data from Stockholm, Gothenburg, and Malmö, Olstrup et al. (2018) found substantive decreasing trends in PM<sub>2.5</sub> and NO<sub>2</sub>, but not PM<sub>10</sub> or O<sub>3</sub>, between 1990 and 2015. Notably, they identified that the decreasing trend in NO<sub>2</sub> levels was broken around 2008 in Stockholm, but not in Göteborg or Malmö, which they attributed to increasing sale of diesel cars in Stockholm. This change of NO<sub>2</sub> trends was not apparent in our models, neither in Stockholm, nor in the other modelling areas (Fig. 3).

We estimated that 2719 (95% CI 2046–3055) premature non-accidental deaths attributable to  $PM_{2.5}$  were prevented by the lower  $PM_{2.5}$  exposure in 2018, compared to 2000. Importantly, since population data for 2018 were used for the entire period, this figure does not consider population growth (i.e., is naturally age-standardised). Despite this, the figure is likely to be an underestimation, as the calculations were based on the global exposure response function (i.e., assuming a constant risk ratio 1.08 per 10 µg m<sup>-3</sup> across exposure levels) derived by the WHO (Chen and Hoek 2020), while epidemiological evidence points towards a nonlinear exposure-response relationship with stronger associations at low exposure levels (Vodonos et al. 2018; Burnett et al. 2018; Christidis et al. 2019).

While reflective of the decreasing trend in exposure, the number of prevented deaths should be put in the context of persisting health consequences of air pollution exposure in Sweden. Alpfjord Wylde et al. (2023) estimated that  $PM_{2.5}$  was still associated with 4264 premature deaths across the entire Swedish population in 2019, assuming a supralinear exposure-response relationship without a lower threshold

and a stronger association with near-source exposure. Similarly, including also PM<sub>10</sub> and NO<sub>2</sub>, Gustafsson et al. (2022) estimated 6740 premature deaths in Sweden in 2019, based on their higher estimates of population exposure. The difference in health impact between Alpfjord Wylde et al. and Gustafsson et al. can be attributed to differences in estimated exposure levels, as discussed above, as well as different exposure-response functions applied. The differences in figures from our study compared to both Alpfjord Wylde et al. and Gustafsson et al. is that they estimated the remaining health impact of exposure in 2019, whereas we focus on the change in health impact due to decreasing exposure levels, i.e., difference between exposure in 2000 and 2018. Although the decreasing trends in air pollutant concentrations show that implemented efforts to reduce exposure have been effective and that the health benefits have been substantive, these figures also indicate that there are considerable health benefits to be derived from further reductions.

The main strengths of this study include the large area covered, including all major metropolitan areas in Sweden and more than half of the country's population, the long time period, the high spatial resolution, the inclusion of multiple important emission sources, and the high validity when compared to data from quality-controlled monitoring stations representing urban background concentrations. The correlations between modelled and measured concentrations were comparable to or higher than previous studies (Rittner et al. 2020; Ketzel et al. 2021; Shen et al. 2022). Another strength is that our modelling includes large portions of rural areas, which are underrepresented in air pollution studies and increases exposure contrasts in epidemiological studies. However, the current modelling areas were selected to cover the six SCAPIS sites, which are all located in metropolitan areas, and the population weighted mean exposures for the modelling areas are thus not fully representative of the entire Swedish population. While large efforts were made to reduce differences between model areas, some differences remain. Although the consequences of these differences are likely to be small, if using the data for epidemiological studies it remains advisable to adjust statistically for the modelling area.

High-resolution exposure modelling at the scale required to cover the modelling areas over multiple years necessitated some methodological simplifications. Most importantly, buildings were not explicitly represented in the models, meaning that elevated concentrations in streetcanyons are not resolved. This is likely to have resulted in underestimation of the exposure in the major urban centres, and consequently underestimated the number of individuals exposed above the WHO guidelines. Similarly, we did not consider inter-annual variation in road traffic non-exhaust emissions due to variations in rainfall or road surface humidity. The chemical transport model included all relevant pollutants and associated chemical reactions; however, it is possible that sources for some sectors are missing in the emission representation in the model, or the time-variations of emissions may fail to capture the real situation. Similarly, substantial uncertainty remains in the estimate of residential heating emissions due to the limited information available on types of appliances, firing habits, and fuel consumption. There are also significant uncertainties in the meteorological data as well as in parameterisations related to the description of the atmospheric boundary layer. These uncertainties were reduced by the bias-correction scheme, which combines the model results with monitoring data. The high correlations with measurement data also corroborate a high validity of the resulting exposure models. Since the different pollutants are correlated, we performed the health impact assessment only for PM<sub>2.5</sub> to avoid double-counting. However, this means that the total mortality prevented by decreased levels of all air pollutants is likely to be larger, especially considering local interventions which have a greater effect on NO<sub>2</sub>.

# Conclusions

We developed a high-resolution dispersion model to assess ambient concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, NO<sub>x</sub>, and NO<sub>2</sub> in six metropolitan areas across Sweden, covering 5.5 million inhabitants. The data generated by the model are useful for future epidemiological studies and health impact assessments. The model validity was high when compared to independent measurement data. Our results show a strong decreasing trend in population exposure with tangible positive effects on population mortality. However, despite the comparatively low concentrations, a majority of the population was still exposed to pollutant concentrations above the current WHO guidelines in 2018. This indicates that ambitious reduction targets for air pollution can provide tangible public health benefits, also in areas with relatively clean air. Our findings highlight the public health importance of continued efforts to reduce ambient air pollution emissions.

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Author contributions Peter Molnár, Leo Stockfelt, Petter Ljungman, Göran Pershagen, and Martin Tondel contributed to the study conception and design. Air pollution modelling and validation were performed by Christian Asker, David Segersson, Cecilia Bennet, Mårten Spanne, Susanna Gustafsson, Jenny Lindvall, and Kristina Eneroth. The health impact assessment was performed by Karl Kilbo Edlund. Illustrations and tables were prepared by Peter Molnár, Karl Kilbo Edlund, and Mårten Spanne. The first draft of the manuscript was written by Karl Kilbo Edlund, David Segersson, and Marta Kisiel, and all authors commented on subsequent versions of the manuscript. All authors read and approved the final manuscript. **Funding** Open access funding provided by University of Gothenburg. This study was funded by the Swedish Research Council for Health, Working Life and Welfare (FORTE, grants 2019-00169, 2020-01044).

**Data availability** Concentration maps for total  $PM_{2.5}$ ,  $PM_{10}$ ,  $NO_x$ , and  $NO_2$  are publicly available via the Swedish National Data Service (Molnár and Ögren 2024). Total and source-specific exposure data for the SCAPIS participants are available from the SCAPIS database (https://www.scapis.org), subject to data access regulations. Source-specific concentration maps are available from the authors upon reasonable request.

#### Declarations

**Ethical approval** SCAPIS was given ethical approval by the Regional Ethical Review Board in Umeå (Dnr 2010–228-31 M).

**Consent to participate** All SCAPIS participants provided written informed consent to participate.

Consent to publish This article contains no identifiable personal data.

Competing interests All authors declare no competing interests.

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