Trends in air pollutants and health impacts in three Swedish cities over the past three decades

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Abstract. Air pollution concentrations have been decreasing in many cities in the developed countries. We have estimated time trends and health effects associated with exposure to NO_x, NO_2, O_3, and PM_{10} in the Swedish cities Stockholm, Gothenburg, and Malmo from the 1990's to 2015. Trend analyses of concentrations have been performed by using the Mann-Kendall test and the Theil-Sen method. Measured concentrations are from central monitoring stations representing urban background levels, and they are assumed to indicate changes in long-term exposure to the population. However, corrections for population exposure have been performed for NO_x, O_3, and PM_{10} in Stockholm, and for NO_x in Gothenburg. For NO_x and PM_{10}, the concentrations at the central monitoring stations are shown to overestimate exposure when compared to dispersion model calculations of spatially resolved population-weighted exposure concentrations, while the reverse applies to O_3. The trends are very different for the pollutants that are studied; NO_x and NO_2 have been decreasing in all cities, O_3 exhibits an increasing trend in all cities, and for PM_{10}, there is a slowly decreasing trend in Stockholm, a slowly increasing trend in Gothenburg, and no significant trend in Malmo. Trends associated with NO_x and NO_2 are mainly attributed to local emission reductions from traffic. Long-range transport and local emissions from road traffic (non-exhaust PM emissions) and residential wood combustion are the main sources of PM_{10}. For O_3, the trends are affected by long-range transport, and there is a net removal of O_3 in the cities. The increasing trends are attributed to decreased net removal, as NO_x emissions have been reduced.

Health effects in terms of changes in life expectancy are calculated based on the trends in exposure to NO_x, NO_2, O_3, and PM_{10}, and the relative risks associated with exposure to these pollutants. The decreased levels of NO_x are estimated to increase the life expectancy by up to 11 months for Stockholm and 12 months for Gothenburg. This corresponds to up to one fifth of the total increase in life expectancy (54–70 months) in the cities during the period 1990–2015. Since the increased

concentrations in O_3 have relatively small impact on the changes in life expectancy, the overall net effect is increased life expectancies in the cities that have been studied.

1. Introduction

Air pollution exposure is clearly recognised as an important global risk factor for the development of a large number of diseases and disabilities (Cohen et al., 2017). In 2015, exposure to $PM_{2.5}$ (particles with an aerodynamic diameter smaller than 2.5 micrometer) was ranked number five among all risk factors for premature deaths, where smoking, diet, and high blood pressure are included. It is estimated to have caused 4.2 million premature deaths among the world's population. This can be compared to smoking, with a corresponding value of 6.4 million premature deaths for the year 2015 (State of Global Air, 2017).

In view of the apparent major health impact associated with exposure to air pollutants, it is of great importance to analyse the trends in air pollution concentrations, and how these affect the public health.

Worldwide, many cities, especially in high-income countries, show substantial decreasing trends in air pollution concentrations (e.g. WHO, 2016a; Geddes et al., 2016; Colette et al., 2011). Considering the changes in the total emissions in Europe and in the U.S., respectively, there are different conditions. In Europe (EU–27), during the period 2002–2011, the emissions of nitrogen oxides (NO_x = sum of NO and NO_2) have decreased by 27 %, and the emissions of primary PM_{10} (particles with an aerodynamic diameter smaller than 10 micrometer) have decreased by 14 % (Guerreiro et al., 2014). In the U.S., the NO_x emissions in eight cities have decreased between 25 % and 48 % during the period 2005–2012 (Tong et al., 2015). Considering the trends in the U.S. regarding all sector-specific emissions during the period 1990–2010, NO_x exhibits a decrease of 48 %, while PM_{10} exhibits a decrease of 50 % (Xing et al., 2013). So, the NO_x and PM_{10} emissions have decreased almost equally in the U.S., while in Europe, the emissions regarding NO_x exhibit a sharper decline compared to PM_{10} (Guerreiro et al., 2014). However, it should also be noted that there are large between-country variations in NO_x emission trends, partly reflecting that some countries have had problems meeting the original National Emission Ceilings and the Air Quality directives (EEA, 2017).

For ozone (O_3) in Europe, the concentrations tend to increase at urban sites, especially during the cold season, as observed by e.g. Sicard et al. (2016) for cities in France, and for most cities in Europe by Colette et al. (2011). This rise can be explained by increases in imported O_3 by long-range transport, and also by a decreased titration by nitrogen monoxide (NO), due to the reduction in local NO_x emissions (Colette et al., 2011; Sicard et al., 2016). However, trends in O_3 are also different for summer and winter, with mainly decreasing trends in the summer and increasing trends in the winter, and there are also some variations between cities in the EU (EEA, 2016). Furthermore, considering different statistical metrics, the trends for O_3 at both urban and rural sites in the UK are different during the period 1993–2011, depending on the metric that is used; mean and median trends are positive, while the maximum trend is negative (Munir et al., 2013). In Northern Alberta in Canada, where measurement results from four urban locations were analysed, the mean concentrations of O_3 have increased at most stations during the period 1998–2014 (Bari and Kindziersky, 2016).

This means that the general population exposure, and presumably also associated health effects of the urban populations, have been reduced for NO_x and PM_{10}, but increased for the mean O_3 concentrations. So far, there are, however, rather few studies that have assessed the net health gain or health loss associated with the trends. Henschel et al. (2012) has examined intervention studies focusing on improvements in air quality, and associated health benefits for the assessed population. Some studies have focused on trends and effects associated with particulate matter (Tang et al., 2014; Keuken et al., 2011; Correria et al., 2013), and some studies have also included ozone (Fann and Risley, 2013; Gramsch et al., 2006). Correria et al. (2013) estimated that for the most urban communities in the USA, as much as 18 % of the increase in life expectancy from 2000 to 2007 was attributable to the reduction in $PM_{2.5}$. When health improvements associated with $PM_{2.5}$ trends are compared to those for O_3, the health benefits associated with $PM_{2.5}$ are much greater (Fann and Risley, 2013).

In a few studies, air quality dispersion models together with gridded population data and exposure-response functions have been used to assess health impacts of changing air pollutant emissions. E.g. for Rotterdam (The Netherlands), the health benefits associated with decreasing trends for EC (elemental carbon) and PM_{10} have been calculated for the period 1985–2008, and the average gain in life expectancy was 13 and 12 months per person for PM_{10} and EC, respectively (Keuken et al., 2011). Fann and Risley, (2013) estimated changes in ozone and $PM_{2.5}$ concentrations in the U.S. during the period 2000–2007, and they estimated the impact on the number of premature deaths by using health-impact functions based on short-term relative risk estimates for O_3 and long-term relative risk estimates for $PM_{2.5}$. Data from monitoring stations were spatially interpolated. Overall, they found net benefits in the number of premature deaths ranging from 22 000 to 60 000 for $PM_{2.5}$, and from 880 to 4 100 for ozone, but interestingly, they found opposing trends in premature mortality associated with $PM_{2.5}$ and ozone at some locations, and a considerable year to year variation in the number of premature deaths. Tang et al. (2014) evaluated the health benefits associated with coal-burning factory shutdowns and accompanied decreasing levels of PM_{10} in Taiyuan in China during the period 2001–2010. They used PM_{10} measurements from monitoring stations, but without considering spatial variations in the exposures, and they used the Chinese national standard of 40 µg m^{-3} as a threshold level. The number of premature deaths associated with PM_{10} levels dropped from 4 948 in 2001 to 2 138 in 2010, and they did not include other pollutants in their analysis.

The studies that have been referred above have used different methodologies of estimating the exposure; by simply taking the concentration at the monitoring stations, and making spatial interpolations between several monitoring stations, or by using dispersion modelling to estimate the spatial distribution of the exposure concentrations. They have also used different relative risks for mortality, and different ways to apply the baseline mortality and the health-effect exposure threshold.

The objective of this study is to quantify and compare the changes in life expectancy, resulting from changes in different pollutants, namely NO_x, NO_2, O_3, and PM_{10}, based on measurements during 25 years in the three largest cities in Sweden. We show that the health impacts related to change in different pollutants are different, and we aim to discuss how different trends in different air pollutants affect the life expectancy assessment.

2. Methods

2.1 The choice of air pollutants for trend analysis

Our main trend analyses are based on simultaneous, continuous measurements of NO_x, NO_2, O_3, and PM_{10}, during the period 1990 to 2015 in Stockholm, Gothenburg, and Malmo; the three largest cities in Sweden. Based on these trends, we calculate the health impacts associated with changes in the population exposure. The changes in life expectancy are calculated with the mean values and the 95 % confidence intervals of the relative risks, while for the trends and the population-weighted exposure concentrations, only the median and the mean values, respectively, have been used, but without considering their confidence intervals. This means that the ranges of the confidence intervals are narrower than they would have been if the confidence intervals of the trend-lines and the population-weighted exposure concentrations were also included in the calculations. NO_2, O_3, and PM_{10} have been regulated in the EU directives, and these regulations also set methods, quality assurance, and control (EU, 2008), making these pollutants relevant to analyse in our study.

2.2 Measuring sites and instrumentation

In all analysed cities, the measuring site is located in the city centre and represents the urban background (Fig. 1). Stockholm is the capital and the largest city in Sweden and has a temperate climate with four distinct seasons. Gothenburg is the second largest city located on the west coast of Sweden. It is like Stockholm located within the west-wind belt, and the proximity to the Atlantic Ocean means a slightly milder climate compared to Stockholm. Malmo is located in the southernmost part of Sweden. General information about the three cities regarding population structure, life expectancy at birth, and baseline mortality is presented in Table A1.

The measuring station in Stockholm is located at Torkel Knutssonsgatan on a roof 20 m above ground level, in Gothenburg it is located on a roof 30 m above ground level in the neighbourhood Östra Nordstan, and in Malmo it is located on a roof 20 m above ground level at the city hall (Rådhuset) in the city center (Fig. 1). They are all regulatory monitoring urban background stations using reference methods. In all stations, PM_{10} has been measured by using Tapered Element Oscillating Microbalance (TEOM 1400A, Thermo Fisher Scientific, USA). However, in Gothenburg, the Continuous Dichotomous Ambient Air Monitor (Thermo Scientific 1405-DF TEOM) has also been used. Nitrogen oxides are measured in all stations by using chemiluminescense (AC 32M, Environnement SA., France). In Stockholm, ozone is measured by using UV-absorption (O342M, Environnement S.A, France). In Gothenburg, ozone has been measured by using non-dispersive UV photometry (EC9811, Ecotech Pty Ltd, Australia), and also by using CLD 700 AL, Ecophysics, Switzerland, and in recent years by using T200, Teledyne, USA. In Malmo, ozone has been measured by using UV-absorption (Thermo Environmental Instruments Model 49C, USA).

Figure 1: Stockholm, Gothenburg, and Malmo; the three largest cities in Sweden, and their surroundings. The black dot in each city shows the location of the measuring station. Each map represents an area of 35 x 35 km.

5 **2.3 Statistical analysis of the trends**

The changes in air pollution concentrations during the period 1990–2015 have been calculated by using the Openair package (Carslaw and Ropkins, 2012). For the trend analyses, the Mann-Kendall test and the Theil-Sen method have been used. The Mann Kendall test is a non-parametric trend test, which is based on the ranking of observations (Hirsch et al. 1982). The Theil-Sen method is used to calculate the median slope of all possible slopes that may occur between the data points (Theil, 1992;
10 Sen, 1968). In our calculations, the trends are based on monthly averages, and they are adjusted for seasonal variations, as these can have a significant effect on monthly data. The Theil-Sen method is regarded as more suitable than the linear-regression method, as it gives more accurate confidence intervals with non-normal distributed data, and it is not affected as much by outliers.

2.4 Health impact and life expectancy calculations

15 Trends in urban background concentrations reflect the change in population exposure over time (see further below), and they are used to calculate changes in health impacts, presented as changes in life expectancy. As basis for the calculations, we use the population size, age distribution, and mortality rate in Stockholm, Gothenburg, and Malmo according to the year 1997, where data in different age groups have been taken from Statistics Sweden (SCB, 2017), and from the Swedish National Board of Health and Welfare (Socialstyrelsen, 2017). Mortality data before 1997 are not available. However, our own test runs have
20 shown that the calculations in life expectancy give very similar results regardless of the year (1997–2015) in which the population structure and mortality statistics are based on. To illustrate the health benefits associated with decreasing trends, the increase in life expectancy at birth has been calculated in AirQ+ according to a log-linear function (Eq. 1). Similarly, the decrease in life expectancy at birth associated with increasing trends has been calculated in AirQ+ according to the same log-linear function (Eq. 1) (WHO, 2016b). We have applied relative risks obtained from previous epidemiological studies, where

the relationships between mortality and exposure to NO_x, NO_2, O_3, and PM_{10} have been analysed. The concept relative risk (RR) represents a log-linear exposure-response function, where the ratio of the incidence in an exposed group is compared to the incidence in a non-exposed, or a less exposed, group. The mortality rate associated with a change in exposure to pollutant x is calculated as:

$$RR(x) = e^{\beta(X-X0)} \qquad (1)$$

where the beta-coefficient (β) indicates the linear relationship between the health impact and the change ($x - x_0$) in exposure.

For NO_x, we apply the RR 1.06 (95 % CI 1.03–1.09) per 10 µg m^{-3} increase, based on the results from Stockfelt et al. (2015), representing all-cause mortality associated with long-term exposure to NO_x in a cohort with men in Gothenburg.

For NO_2, we apply the RR 1.066 (95 % CI 1.029–1.104) per 10 µg m^{-3} increase. This RR is based on pooled estimates of mortality associated with long-term exposure to NO_2 (Faustini et al., 2014).

For O_3, we apply the RR 1.02 (95 % CI 1.01–1.04) per 10 ppb increase, corresponding to 1.01 (95 % CI 1.005–1.02) per 10 µg m^{-3} at 25 ºC and 1 atm, based on a large prospective study examining the associations between long-term ozone exposure, and all-cause and cause-specific mortality (Turner et al., 2016).

For PM_{10}, we apply the RR 1.04 (95 % CI 1.00–1.09) per 10 µg m^{-3} increase, which is based on a meta-analysis of 22 European cohorts (Beelen et al., 2014). This value is also in line with the RR estimate of 1.043 (95 % CI 1.026–1.061) per 10 µg m^{-3} increase, since many years used in impact assessments (Künzli et al., 2000), based on cohort studies in the U.S.

All RRs described above are based on calculations for the population aged 30 years and over, and therefore, the changes in life expectancy, calculated in AirQ+, are also based on the age group 30 years old and over. The calculations have also been performed by assuming no threshold under which no effect occurs.

2.5 Relationship between urban-background concentrations and population-weighted exposure concentrations

The trends measured at urban background monitoring stations may not be representative for the trends in exposure to the entire population in those areas, due to the position of the urban-background measuring stations, the spatial variation in air pollution concentrations, and the variability in the population density within the cities. In order to assess the health effects of the population, associated with changes in air pollution exposure in each metropolitan area, we estimated the relations between the concentrations at the urban-background monitoring stations, and the population-weighted exposure concentrations. This is assessed by comparing model-calculated annual population-weighted exposure concentrations with annual-mean urban-background concentrations. We have done this for NO_x in Stockholm and Gothenburg, and for O_3 and PM_{10} in Stockholm, but due to lack of data, these relations have not been possible to calculate other than for the above-mentioned pollutants and cities.

Geographically resolved annual-mean concentrations of NO_x in Stockholm were calculated by using a wind model and a Gaussian air-quality dispersion model as a part of the Airviro system (Airviro, 2017). Details on the modelling and emission data are described in Johansson et al. (2017). The same modelling of NO_x was done for Gothenburg by using a Gaussian model (Aermod, US EPA) as a part of the EnviMan AQ Planner (OPSIS, Furulund, Sweden) as described in Molnár et al. (2015).

Spatially resolved O_3 concentrations in Stockholm are calculated from a combination of measurements and dispersion modelling of NO_x concentrations. The modelled NO_x concentrations are converted to O_3 based on the measured NO_x concentrations at an urban background site in Stockholm, and the difference between O_3 at an urban background site and at a rural background site. The basic chemistry behind this is that the O_3 concentration within a city is controlled by the transport from the surrounding areas into the city, and by the removal of O_3 due to the reaction with NO (nitrogen monoxide). It is further set that if O_3 at the urban background is higher compared to the rural background, then it is set to the same value as the urban background, and if it becomes less than zero, it is set to zero. More details of this method is described in Olsson et al. (2016).

Population-weighted exposure concentrations (C_{pop}) are obtained by multiplying the calculated concentration (C_i) in each grid-square cell with the number of people in the corresponding grid-square cell (P_i), and summing all products, and dividing the sum by the total population (Eq. 2). This procedure has been used in several previous studies (e.g. Johansson et al., 2009; Orru et al., 2015).

$$Cpop = \frac{\sum Ci\,Pi}{\sum Pi} \qquad\qquad (2)$$

The relationships between urban-background levels and population-weighted exposure concentrations are presented in Appendix A Fig. A1–A4.

3. Results

3.1 Overview of trends

Fig. 2–4 show the trends in concentrations of NO_x, NO_2, O_3, and PM_{10} measured at urban background sites in Stockholm, Gothenburg, and Malmo during the period 1990–2015. During the given time periods, NO_x and NO_2 exhibit decreasing trends in all cities, whereas O_3 exhibit increasing trends in all cities, and for PM_{10}, the trends are less clear and consistent. For PM_{10} in Stockholm and Malmo, the data from the measuring stations only include the period 1997–2015 and 1996–2015, respectively. In several cases, the trends are not perfectly linear throughout the periods (Fig. 2–4).

For NO_x, NO_2, and PM_{10}, the trends are based on the monthly average concentrations, and for O_3, they are based on rolling eight-hour daily maximum concentrations. The reason for this division is that the relative risk of O_3, used in our health-impact calculations, is based on eight-hour daily maximum values. In Fig. 2–4, the median slopes are calculated according to the Theil-Sen method (Theil, 1992; Sen, 1968).
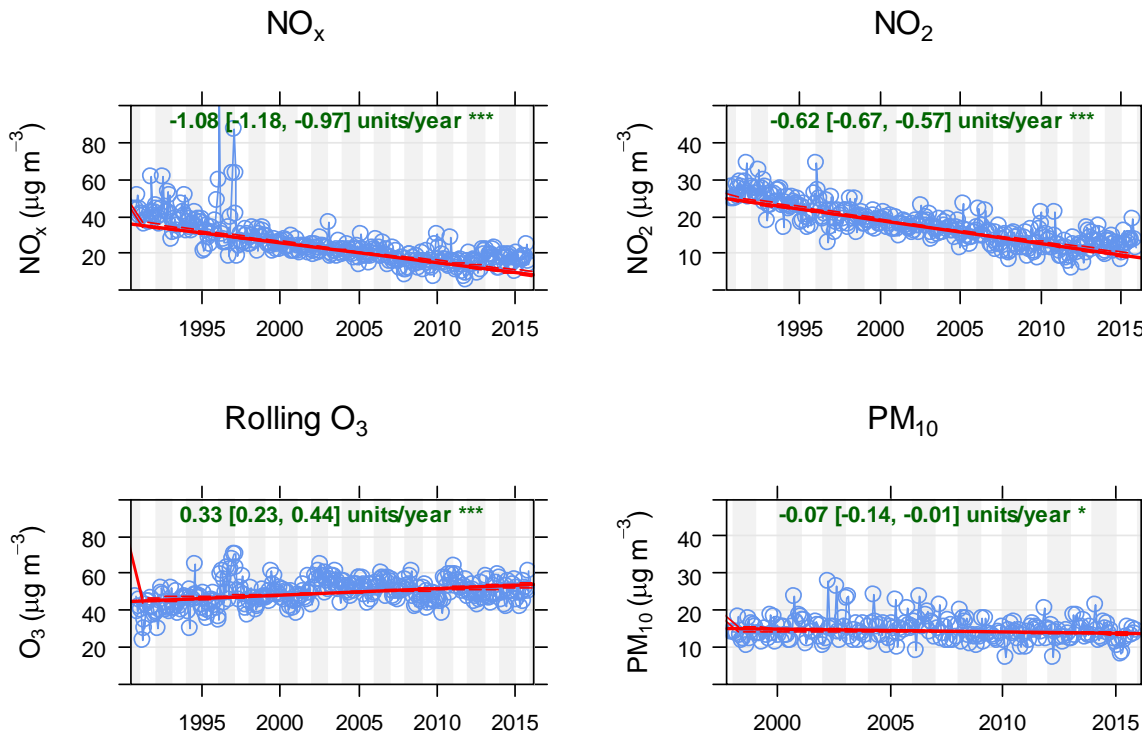
Figure 2. Stockholm: Trends in NO_x, NO_2, O_3, and PM_{10}, measured from 1990–2015. For PM_{10}, data are only for the period 1997–2015. The blue rings are monthly averages, and the calculated deseasonalised trends using the Theil-Sen method are shown as the red thick lines. Unit for the trends is $\mu g\ m^{-3}\ year^{-1}$, and the values in parentheses are 95 % confidence intervals. The stars (*) represent the significance level of the trend-lines, where three stars means that $p < 0.001$, and one star means that $0.01 < p < 0.05$.
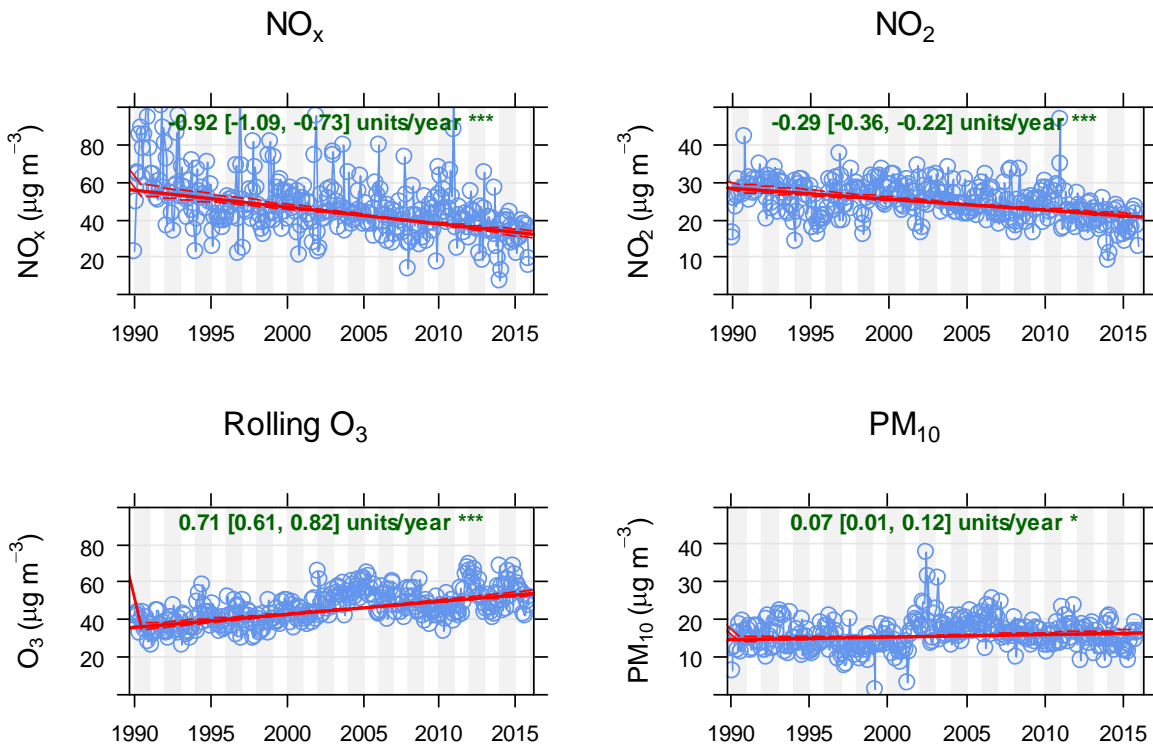
Figure 3. Gothenburg: Trends in NO_x, NO_2, O_3, and PM_{10}, measured from 1990–2015. The blue rings are monthly averages, and the calculated deseasonalised trends using the Theil-Sen method are shown as the red thick lines. Unit for the trends is µg m^{-3} year^{-1}, and the values in parentheses are 95 % confidence intervals. The stars (*) represent the significance level of the trend-lines, where three stars means that $p < 0.001$, and one star means that $0.01 < p < 0.05$.
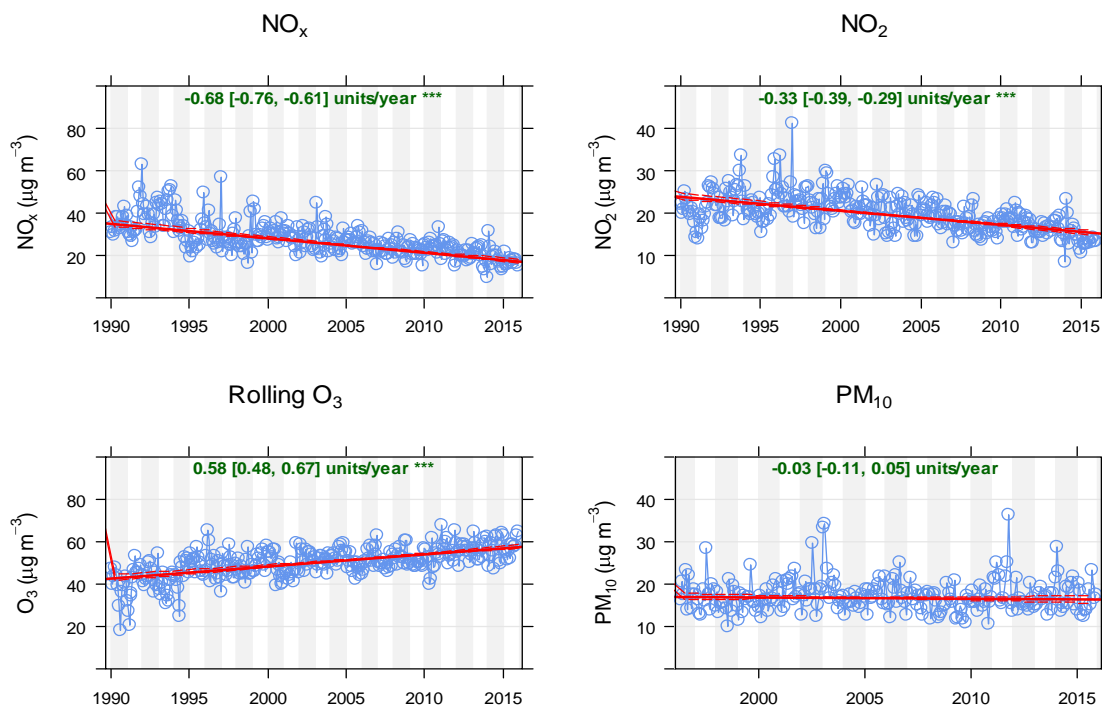
Figure 4. Malmo: Trends in NO_x, NO_2, O_3, and PM_{10}, measured from 1990–2015. For PM_{10}, data are only for the period 1996–2015. The blue rings are monthly averages, and the calculated deseasonalised trends using the Theil-Sen method are shown as the red thick lines. Unit for the trends is $\mu g\ m^{-3}\ year^{-1}$, and the values in parentheses are 95 % confidence intervals. The stars (*) represent the significance level of the trend-lines, where three stars means that $p < 0.001$, and one star means that $0.01 < p < 0.05$.

In Figure 5, we show the trends for the first period 1990–2007, and for the second period 2008–2015 separately (except for PM_{10} which for Stockholm is from 1997–2007 and from 2008–2015, and for Malmo from 1996–2007 and from 2008–2015). In Stockholm, the NO_x and NO_2 concentrations have decreased significantly from 1990–2007, but for 2008–2015, there is even a tendency for increasing concentrations. The patterns are somewhat different in Gothenburg and Malmo.
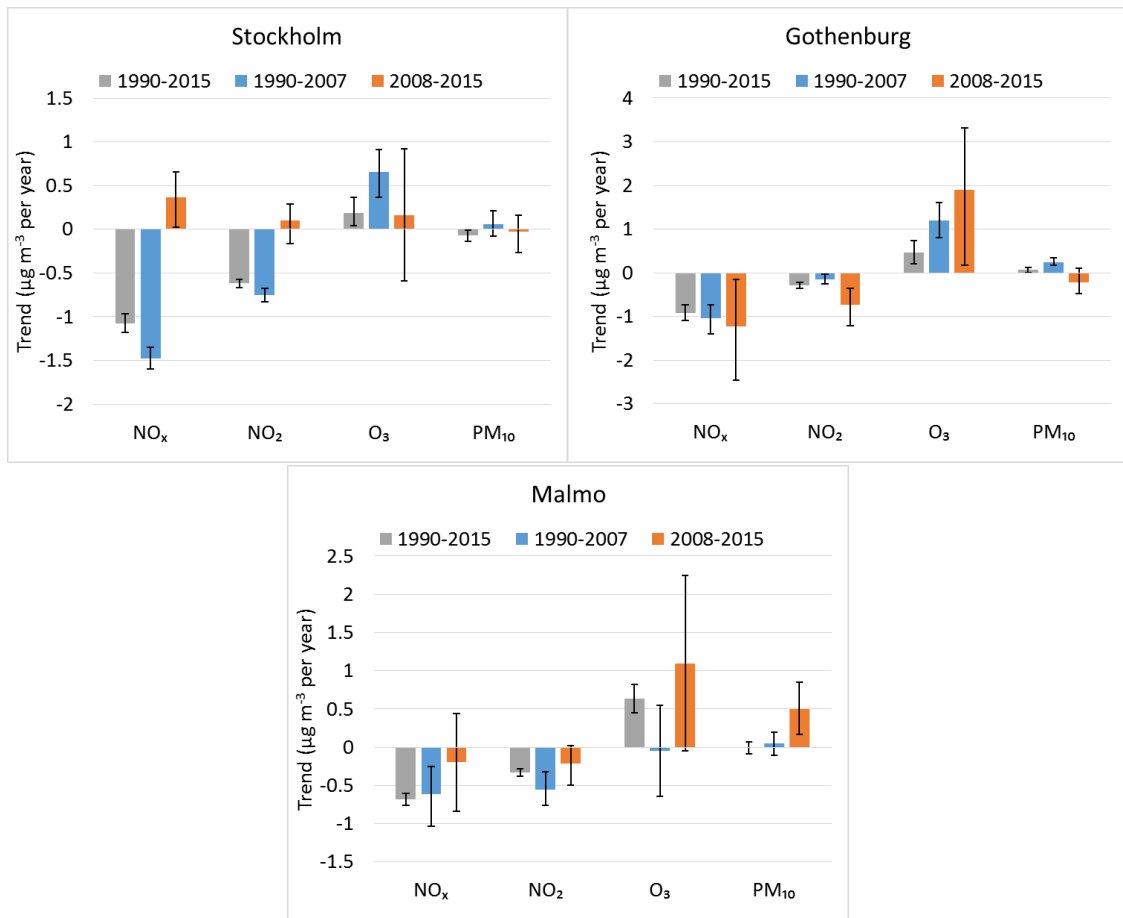
Figure 5: The trends for the measured pollutants in µg m^{-3} year^{-1} (95 % CI) for the whole period, and divided into two different periods.

There are also significantly different trends during weekdays and weekends in the three cities. In Fig. 6–8, the trends have been divided into weekdays and weekdays, reflecting the importance of local emissions compared to non-local emissions, where local emissions from traffic are more prominent during weekdays compared to weekends. Fig. 6–8 are otherwise designed in the same manner as Fig 2–4. For NO_x and NO_2, the downward trends in all cities are more prominent during weekdays compared to weekends, indicating that local emission reductions, mainly from traffic, have had the greatest impact. For O_3, the increasing trends in Gothenburg and Malmo are slightly higher during the weekends compared to the weekdays, while this does not apply to Stockholm. The trends related to PM_{10} are less clear, and they are also in many cases not statistically significant. Possible reasons for the appearances of the trends are further analysed in the discussion section.
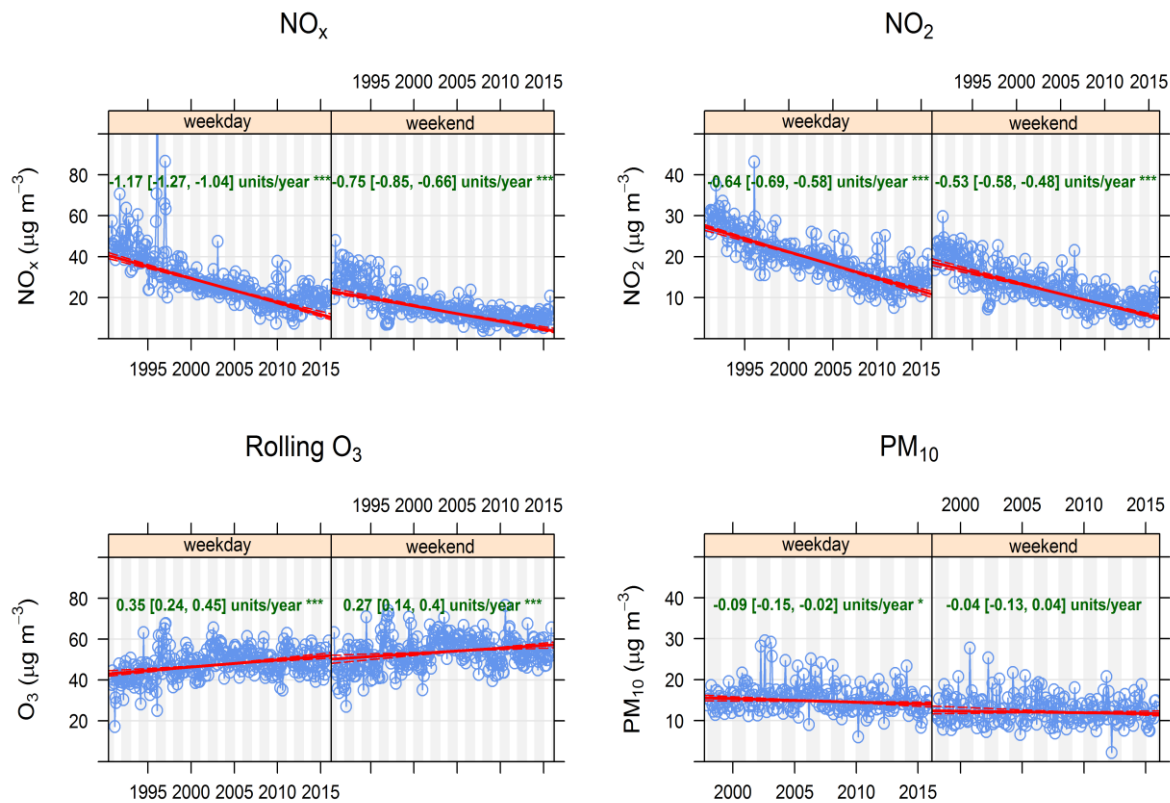
Figure 6. Stockholm: The trends for NO$_x$, NO$_2$, O$_3$, and PM$_{10}$, divided into weekdays and weekends.
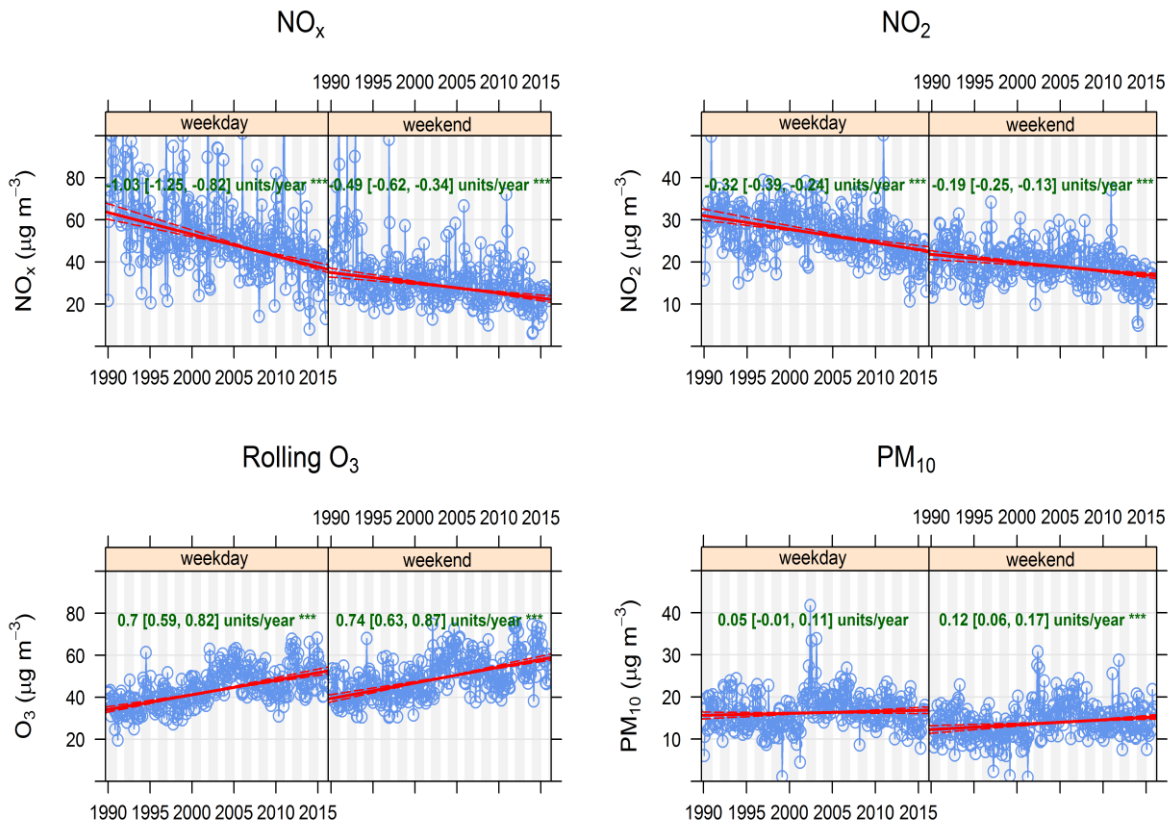
Figure 7. Gothenburg: The trends for NO_x, NO_2, O_3, and PM_{10}, divided into weekdays and weekends.
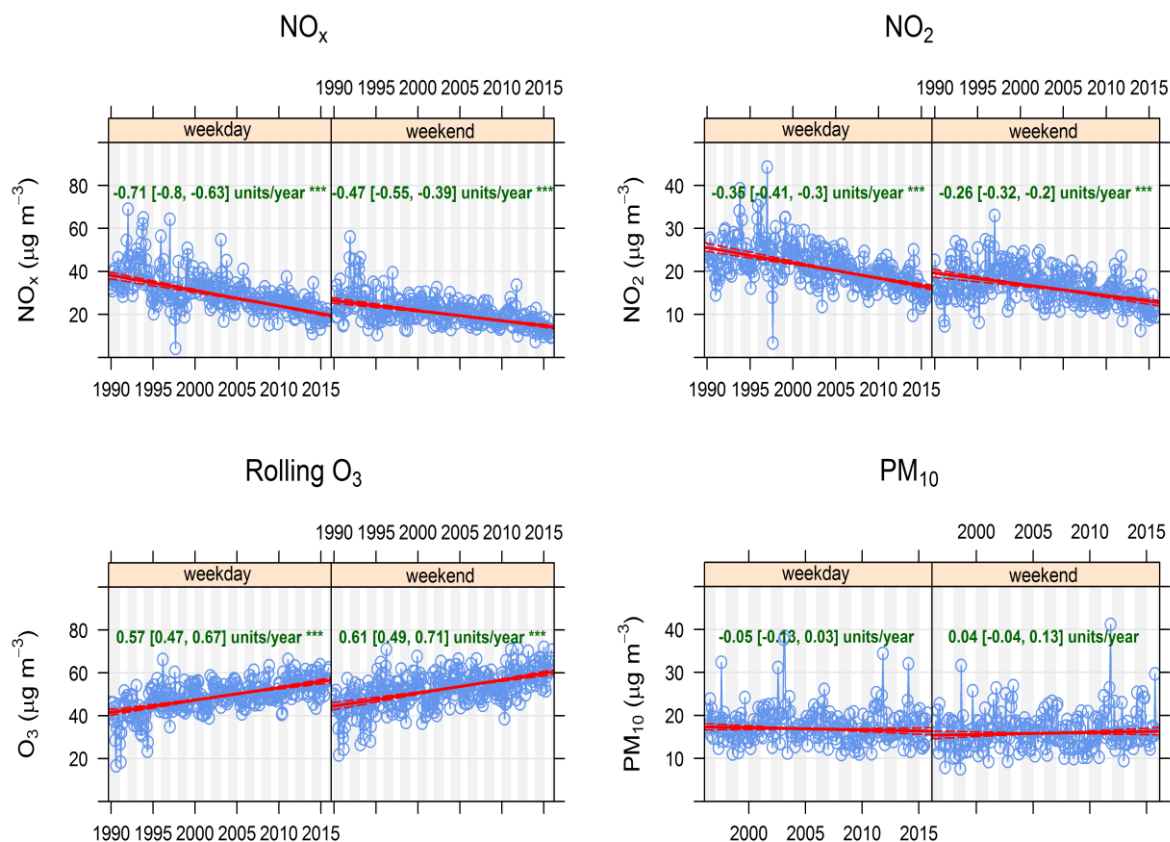
Figure 8. Malmo: The trends for NO_x, NO_2, O_3, and PM_{10}, divided into weekdays and weekends.

All the trends, except for PM_{10} in Malmo, are statistically significant within a 95 % CI. It can be noted that the rate of decreasing NO_x is highest in Stockholm, and then comes Gothenburg and last Malmo, whereas the opposite order is for the rate of increasing O_3 concentrations. If the change in O_3 was only associated with local NO titration, the opposite would be expected, so this indicates that other factors are also important for the O_3 trends, like the O_3 concentrations at the regional background sites outside the cities.

3.2 Health impact assessment associated with the trends in air pollution concentrations

3.2.1 Change in life expectancy

In order to estimate the health impacts associated with changing concentrations of NO_x, NO_2, O_3, and PM_{10} in Stockholm Gothenburg, and Malmo, the concentration changes during the given time periods for the pollutants, presented in Table 1, and the relative risks, presented in section 2.4, have been used. Adjustments for population-weighted exposure concentrations have also been performed in the cases where data are available (Fig. A1–A4). The changes in life expectancy are calculated with

the mean values and the 95 % confidence intervals of the relative risks, while for the trends and the population-weighted exposure concentrations, only the median and the mean values, respectively, have been used, but without considering their confidence intervals. NO_x and NO_2 exhibit decreasing trends in all cities, which means an increase in life expectancy (positive values). The opposite applies to O_3, with increasing trends in all cities. For PM_{10}, there is a decreasing trend in Stockholm,

5   with an increase in life expectancy, and an increasing trend in Gothenburg, with a decrease in life expectancy. For PM_{10} in Malmo, however, no life expectancy change has been possible to calculate, due to the lack of a significant trend (see Fig. 4). The detailed results are presented in Table 1.

The largest increase in life expectancy is expected due to the reduction in NO_x, and somewhat less due to the reduction in NO_2 concentrations. The decrease in NO_x levels corresponds to up to about 20 % of the total population life expectancy increase

10   of between 54 and 70 months during the period 1990–2015 (Table A1.). In opposite, the increase in ozone exposure might have caused the life expectancy to decrease with one to two months. For PM_{10}, the effects are very mixed; in Stockholm, there might have been a small increase in life expectancy, whereas in Gothenburg, a small decrease has appeared, and for Malmo, no life expectancy change has been possible to calculate.

15   **Table 1:** Change in life expectancy in months with 95 % CI in brackets, caused by change in exposure during the measured periods. Decreasing trends are associated with an increase in life expectancy, and increasing trends are associated with a decrease in life expectancy (minus signs). The change in life expectancy, adjusted for population-weighted exposure concentrations (see Section 2.5 and Fig. A1–A4), are presented in bold below the ordinary values. Note that the trend of PM_{10} in Stockholm is only for the period 1997–2015.

Pollutant	Stockholm	Gothenburg	Malmo
NO_x	21 (11–32)	17 (9–26)	13 (7–19)
	10 (5–15)	**11 (5–16)**	
NO_2	13 (6–21)	6 (3–9)	7 (3–11)
O_3	-1 (-0.5 – -2)	-2 (-1 – -4)	-2 (-1 – -4)
	-1 (-0.6 – -3)		
PM_{10}	0.6 (0–1)	-1 (0 – -2)	No significant change
	0.2 (0.0–0.5)		

4 Discussion

4.1 Trends in concentrations

4.1.1 NO_x and NO_2

25   In all the three cities, the NO_x and NO_2 trends for urban background concentrations are decreasing. The emissions of NO_x and PM from different sectors during 1990–2015 have been quantified for Stockholm and Gothenburg as part of a Swedish research programme called SCAC (www.scac.se) (Segersson et al., 2017; Stockfelt et al., 2017). These studies show that the emissions of NO_x in both Stockholm and Gothenburg are dominated by the contribution from road traffic. The emissions dropped by 60

to 70 % from 1990 to 2011 in Gothenburg (inferred from Figure 3a in Stockfelt et al., 2017). A similar trend in road traffic emissions is calculated for Stockholm based on reported emission data for 1993 and 2017 (SLB, 1995 and SLB, 2018). This is mainly due to decreased vehicle emissions associated with the renewal of the vehicle fleet. The effect of changes in the number of vehicle kilometres is relatively small compared to the effect of lower emissions per kilometre. Other local sources

5   have minor impact on NO_x, as shown for Stockholm in (Johansson et al., 2008). The rural background concentrations of NO_x outside the cities are low (2–8 µg m^{-3} or between 6 % and 14 % of the urban background concentrations), and not very important for the overall trend (Figure 9 in IVL, 2016). The importance of road traffic is also consistent with the clear difference between the trends measured during weekdays and weekends (Fig. 6–8).

In the case of Stockholm, the downward trend diminish almost entirely from 2008 onwards, and exhibit rather an increasing

10   trend (Fig. 5). Based on statistics on the share of new registrations of diesel vehicles in the three cities during the period 2008–2015, the proportion of new registrations of diesel vehicles has been greatest in the Stockholm area, which possibly can explain these slightly increasing NO_x and NO_2 trends seen in Stockholm from 2008 onwards, but not in Gothenburg and Malmo (BilSweden, 2018).

Comparing NO_x and NO_2 in the three cities, the decreasing trends for NO_x are much more pronounced compared to NO_2.

15   The proportion of NO_x that is NO_2 has been shown to be increasing at kerbside sites in many cities, and this has partly been attributed to more directly emitted NO_2 due to the increase in diesel vehicles equipped with oxidation catalysts (Carslaw, 2005) and particle filters (Grice et al., 2009). However, for urban background sites, the observed increased share of NO_2, and associated slower decline in concentrations compared to NO_x, are mainly related to the change in O_3/NO_x equilibrium, caused by decreased NO_x and increased O_3 concentrations (Keuken et al., 2009) and not to the direct NO_2 emissions.

20   **4.1.2 Ozone**

The 8-hour daily maximum O_3 levels exhibit increasing trends in all the three cities during the period 1990–2015. Since the concentrations of O_3 are lower at central urban-background sites in the cities compared to outside the cities, the net effect of the photochemistry is that ozone is consumed, mainly due to the titration involving its reaction with NO. Reduced emissions of NO mean that less O_3 is consumed, due to a reduction of the NO titration, and thereby arises an increasing trend, consistent

25   with many other cities in Europe (Sicard et al., 2016).

The increasing trend of O_3 in Stockholm, based on 8-hour daily maximum values, is weaker compared to Gothenburg and Malmo, even though the NO_x trend in Stockholm exhibits a sharper decline compared to Gothenburg and Malmo. This may, however, be explained by considering the trends in the regional background concentrations of O_3. The regional background concentrations of O_3 are measured at Aspvreten 60 km SW of Stockholm, at Råö 45 km S of Gothenburg, and at Vavihill 50

30   km N of Malmo. The trend for O_3 at Aspvreten is more sharply decreasing in comparison with the trends at Råö and Vavihill (SEPA, 2017a). This means that the site transport of O_3 into Stockholm during 1990–2015 decreases more strongly in relation to Gothenburg and Malmo, which explains the weakest increasing O_3 trend in Stockholm, despite the sharpest decreasing NO_x

trend. The differences between the regional background trends in O_3 may explain the absence of perfect anti-correlations between NO_x and O_3 in the cities.

4.1.3 PM$_{10}$

The trends for PM_{10} are not as clear as those for NO_x and O_3. Only Stockholm exhibits a significant downward trend for the entire period. PM_{10} is a mixture of locally produced particles and long-range transport of particles from emissions in other countries. The main local (primary) emissions of PM_{10} are non-exhaust particles from road traffic, ranging from 25 % to 55 %, and biomass burning ranging from 30 % to 35 % (Segersson et al. 2017; Stockfelt et al., 2017). Primary vehicle exhaust particles contribute with less than 10 % of the total mass (Johansson et al., 2007). Road wear due to the use of studded winter tyres is the main source of non-exhaust PM_{10}. The decreasing trend of PM_{10} in Stockholm can be explained by the reduced use of studded tires, and also by dust binding that has been introduced in the recent years (SLB, 2015).

Unlike Stockholm, Gothenburg exhibits increasing trends for PM_{10} during the measurement period (Fig. 3 and 7), however, not significant during weekdays. According to Fig. 5, the trend for PM_{10} in Gothenburg from 2008 onwards is negative, but not significant. During 2006, measures were established in order to reduce the levels of PM_{10}. Dissemination of dust-binding agents, prohibiting the use of studded tires, and the introduction of congestion charges during 2013 may have contributed to this changing trend (Gothenburg City, 2015; Gothenburg Annual Report, 2015).

For Malmo, the trends are not significant, neither for weekdays, weekends, or for the entire period (Fig. 4 and 8).

The trend in concentrations measured at the regional background outside Stockholm (Aspvreten) is decreasing during the period, but it is fragmented with lack of data during parts of the period. For Gothenburg (Råö) and Malmo (Vavihill), there are only data for very limited parts of the period, and firm conclusions about their impact on the urban background concentrations cannot be made (SEPA, 2017b).

4.2 Comparisons between our trends and the trends in U.S. and Europe as a whole

Regarding PM_{10}, the trends in the Swedish cities do not exhibit the same decrease as the trends in the U.S. or in Europe as a whole. An important reason for this is the large contribution of mechanically generated road-dust particles in the Swedish cities. In Stockholm, up to 90 % of the mass fraction of PM_{10} is generated from road abrasion, which is mostly caused by the use of studded tires during winter-time, and the concentration trend for PM_{10} is not significantly influenced by the reduction of exhaust emissions (Johansson et al., 2007). Similar conditions regarding the composition of PM_{10} prevail in Gothenburg (Grundström et al., 2015). In Europe as a whole, the PM_{10} concentrations exhibit a decreasing trend during the period 1990–2010, and this is mainly caused by reduced emissions of both primary PM and precursors of secondary PM within the European countries (EEA, 2017).

According to Geddes et al. (2016), where the global population-weighted annual mean concentrations of NO_2 from 1996–2012 were analysed, the average downward trend in North America (Canada and U.S.) is 4.7 % per year, which is the most heavily declining trend compared to all the other regions in the world. For Western Europe, the average downward trend per

year is, according to Geddes et al. (2016), 2.5 % per year. This downward trend is in the same magnitude as the NO_2 trends in Stockholm, Gothenburg, and Malmo (Fig. 2, 3, and 4).

For O_3, the comparison is a little bit more complicated, since the trends may differ depending on the measure that is used. In the U.K., during the period 1993–2011, the mean and median concentration trends were positive, while the maximum concentration trend was negative (Munir et al., 2013). In Europe as a whole, the O_3 concentrations are influenced by both local emissions, intercontinental inflow, and the meteorological conditions. The O_3 concentrations, and their changes in Europe during the period 1990–2010, are very different depending on time and place, and no unambiguous explanation or interpretation is possible to do (EEA, 2017). Considering the O_3 trends in Fig. 2–4, where these are based on 8-hour daily maximum values, all the trends are increasing. However, the O_3 concentrations associated with monthly averages also show increasing trends for all the three cities during 1990–2015, and as mentioned in section 4.1, the increasing O_3 concentrations in the Swedish cities are probably largely caused by a reduced titration effect, which in turn is caused by decreasing NO concentrations.

4.3 Impacts on life expectancy

In general, the impact of NO_x and NO_2 on life expectancy among the populations is much larger compared to the impact of O_3 and PM_{10}. The calculated gain in life expectancy, associated with decreasing NO_x trends (Table 1), contributes up to as much as about 20 % of the total gain in life expectancy during the period 1990–2015 (Table A1). However, since the O_3 concentrations exhibit increasing trends during the same period, and thereby giving rise to a loss in life expectancy, the summarized effects of NO_x exposure and O_3 exposure may be relevant to consider. For Stockholm, where population-weighted exposure concentrations are available for both NO_x and O_3, the mean value of the gain in life expectancy for NO_x, which is ten months, should be summed up with the mean value of the loss of life expectancy for O_3, which is one month, resulting in a net gain of 9 months. The very small corresponding gain in life expectancy of 0.2 months, associated with exposure to PM_{10}, can also be taken into account. However, the relatively weak trends associated with PM_{10}, with a significance level of $0.01 < p < 0.05$ for Stockholm and Gothenburg, and a corresponding level of $p > 0.05$ for Malmo, cannot be compared with the trends for the others pollutants, which in all cases exhibit significance levels of $p < 0.001$.

4.4 Uncertainties associated with the measurements and the relative risks

In general, the results are highly sensitive to the relative risks obtained from previous epidemiological studies. For NO_x, we use the RR 1.06 (95 % CI 1.03–1.09) per 10 µg m^{-3} increase, based on a recent cohort study in Gothenburg (Stockfelt et al., 2015). Considering the similarities between Gothenburg, Stockholm, and Malmo regarding climate, vehicle fleet, and type of city, this RR may be representative for all these three Swedish cities. The other option could have been to choose the RR 1.08 (95 % CI 1.06–1.11), which comes from on a previous long-term cohort study in Oslo (Nafstad et al., 2004), but we decided to implement the RR from the Swedish study, with exposure levels closer to the actual situation. For NO_2, we use the pooled RR estimate of 1.066 (95 % CI 1.029–1.104) per 10 µg m^{-3} increase, from a meta-analysis (Faustini et al., 2014). The other

option could have been to choose the RR 1.05 (95 % CI 1.03–1.08) from Hoek et al. (2013), which is also based on a meta-analysis from several studies in different countries and continents, but we considered the study group in Faustini et al. (2014) as more relevant for the Swedish situation, since it is based only on European studies. A RR for NO_2, based on European conditions, has also been calculated in the ESCAPE study (Beelen et al., 2014). In this study, the results for NO_2 were based on a limited passive sampling campaign for each cohort, and differ from the consistent results in the two meta-analyses (Faustini et al., 2014 and Hoek et al., 2013), where the studies with active monitoring were also included. In addition, Beelen et al. (2014) did not find any significant association with $PM_{2.5}$ absorption, possibly indicating problems to reflect exposure to vehicle exhaust. Because of this, we consider the RR in Faustini et al. (2014) as more relevant in comparison with the RR in Beelen et al. (2014).

Due to the influence of photochemistry, NO_x is a better indicator of health risks associated with vehicle exhaust emissions than NO_2. This might explain the differences in relative risks for NO_x and NO_2. For the same reason (photochemistry), the trends in NO_2 are to some extent influenced by the trends in O_3.

For O_3 exposure, the relative risk of 1.01 (95 % CI 1.005–1.02) per 10 µg m^{-3} increase, that has been used in our calculations, is based on Turner et al. (2016), which is a large prospective long-term cohort study performed in the U.S. The other option could have been to choose the RR 1.014 (95 % CI 1.005–1.024) from Jerret et al. (2009), which is very close to the RR in Turner et al. 2016, but considering the time periods during which the studies have been conducted, Turner et al. (2016) is more in line with the trend analysis period in this study. The number of epidemiological studies focusing on long-term exposure to O_3 are relatively few, and no meta-analyses are available. This means that the RR values associated with O_3 exposure are based on relatively less amount of data compared to the RR values for NO_2 and PM_{10}, which are obtained from meta-analyses.

For PM_{10}, we use the RR value of 1.04 (95 % CI 1.00–1.09) per 10 µg m^{-3} increase from Beelen et al. (2014), which is a meta-analysis based on 22 European cohorts. An important issue to consider in these Swedish cities is that the mass of PM_{10} to a relatively small percentage consists of combustion-related particles. In the Swedish cities, non-exhaust traffic generated particles are the largest contributors to the mass of PM_{10}, which is clearly shown for Stockholm by Johansson et al. (2007), and for Gothenburg by Grundström et al. (2015). In Beelen et al. (2014), there is also a RR value of 1.04 (95 % CI 0.98–1.10) per 10 µg m^{-3} increase associated with exposure to PM_{coarse}. This value is very similar to the corresponding value for PM_{10} mentioned above. Since PM_{10} in the Swedish cities to a large extent consists of mechanically generated particles in the coarse fraction, this gives additional support for the accuracy of the RR that is applied in the calculations for change in life expectancy.

This means that NO_x and NO_2, which both primarily indicate locally produced combustion-related traffic emissions, are in Sweden clearly separated from PM_{10}, which rather indicates non-exhaust emissions. From a health perspective, PM_{10} is a reflective indicator of the effects of exposure to the coarse fraction of urban aerosols. The health effects related to exposure to the coarse fraction of particles differ from exposure to the fine (combustion related) fraction. Previous studies indicate an association between exposure to the coarse fraction of PM_{10} and respiratory admissions, such as asthma and chronic obstructive pulmonary disease, while cardiovascular diseases are more closely linked to exposure to the fine fraction (Brunekreef and Forsberg, 2005).

4.5 Correlations between the different pollutants

Calculating the health effects associated with exposure to these different pollutants may be problematic, considering that exposures can occur simultaneously. When conducting health-impacts assessments, it is important to avoid double calculations when gain or loss in life expectancy are calculated as a result of decreasing or increasing trends. NO_x and NO_2 are highly correlated, and the health impacts assessments, calculated as a gain in life expectancy, cannot be summed up, but they can rather be considered as different indicators of air pollutants, mainly originated from combustion processes. On the other hand, for these three Swedish cities, the correlation between NO_x or NO_2 and PM_{10} is very low, since the mass proportion of exhaust particles in PM_{10} is very small (Johansson et al., 2007), and the health impacts associated with exposure to NO_x and PM_{10}, respectively, can therefore largely be assumed to be independent of each other. Moreover, when considering simultaneous exposure to NO_2 and $PM_{2.5}$, the relative risks associated with these pollutants are not significantly affected when included in a two-pollutant model (Faustini et al., 2014).

The relative risks associated with O_3 exposure are not significantly affected when they are included in two-pollutant models. The increased risks of circulatory and respiratory mortality associated with exposure to O_3 remained stable after adjustment for $PM_{2.5}$ and NO_2 (Turner et al., 2016). Consequently, the health effects associated with exposure to O_3 can be assumed to be separated from the health effects from the other three pollutants, and the change in life expectancy can therefore be added or subtracted from the change in life expectancy associated with NO_x, NO_2, and PM_{10}.

4.6 The differences between urban-background concentrations and population-weighted exposure concentrations

As shown in Section 2.5 and Figure A1–A4, the average population-weighted exposure concentrations may differ substantially from the concentrations measured at the measuring stations. For NO_x and PM_{10}, the average exposure concentrations for the urban population are lower compared to the measured urban-background concentrations. The reason for this is that the people who live in the outskirts of the urban area are exposed to lower concentrations compared to those living close to the measuring station in the central parts of the cities, where both exhaust emissions and the formation of mechanically generated particles are most apparent. For O_3, the average exposure concentrations are higher compared to the measured urban-background concentrations. This can be explained by the interaction between NO_x and O_3, where the O_3 reaction with NO dominates over the formation of new O_3, and this reaction is more apparent in the central part of a city, where the NO concentration is relatively higher. This explains the higher average population-exposure to O_3 compared to the measured urban-background concentrations at a central monitoring station.

The population distribution within the cities are not constant over time, and the meteorological conditions may also vary over time. Consequently, to calculate a corresponding change in population-weighted exposure, based on an air pollution trend during 25 years, is connected with uncertainties. Linear regression between the concentrations at the measuring stations, and the corresponding population-weighted exposure concentrations for the available years (Fig. A1–A4), are the best estimates

that can be performed. A disadvantage is the limited number of years of calculated relationships, but where the R^2-values in the range of 0.7–0.9 may be considered as good.

Due to lack of data, the relations between urban-background concentrations and population-weighted exposure concentrations have not been possible to obtain for NO_2 in Stockholm, and not for NO_2, O_3 and PM_{10} in Gothenburg, and not for any of the pollutants in Malmo. Without these relations, the population-weighted exposure concentrations have to be based on the data obtained directly from the measuring stations. However, based on the already established relations (Fig. A1–A4), it can be assumed that the life expectancy values associated with NO_x and NO_2 exposure, but without adjustment for population-weighted exposure, are overestimated, while the opposite applies to O_3 exposure.

4.7 Policy implications

From a policy point of view, it is important to know the health impacts of local air pollution emissions (i.e. the primary emissions in the city from e.g. wood burning and road traffic), versus long-range transport of non-local origin. The low rural compared to the urban background concentrations outside the cities shows that the trends in local primary emissions of NO_x and NO_2 are most important for the decreasing trends, and thereby also most important for the increase in life expectancy of the populations. This implies that policies addressing local emissions in the cities are important in order to improve the public health. NO_x may be regarded as an indicator of vehicle exhaust emissions, which consist of many toxic compounds (Künzli et al., 2000). For PM_{10}, long-range transport of mainly secondary PM is more important than local emissions. However, primary emissions may still be more important for the health effects associated with PM, as clearly demonstrated by Segersson et al. (2017). They calculated the sectorial contributions to PM_{10}, $PM_{2.5}$ and black carbon (BC) exposure, and the related premature mortalities in three Swedish cities, including Gothenburg and Stockholm, by using dispersion models with high spatial resolution. They showed that although the main part of the PM_{10} and $PM_{2.5}$ exposures were due to long-range-transport, the major part of the premature deaths were related to local emissions, with road traffic (dominated by non-exhaust wear emissions) and residential wood combustion having the largest impacts. For black carbon, the emissions from local road traffic (exhaust emissions) and residential wood combustion were more important both for exposures and health risks, as compared to long-range transport. This means that local policies aiming at reducing emissions from road traffic, both non-exhaust and exhaust emissions, and residential wood combustion, will be most important for reducing health risks associated with urban air pollution in these cities.

The large impact on life expectancy, which both NO_x and NO_2 represent, makes these pollutants important to consider. NO_x and NO_2 are both indicators of combustion-related air pollutants, as indicated by the high correlation between NO_x and particle number concentration at kerbside sites (Johansson et al., 2007; Gidhagen et al., 2004), and they may therefore be good indicators of population exposure to exhaust emissions from road traffic. Additionally, the change in life expectancy can be regarded as the tip of the iceberg, since reductions in NO_x concentrations are also expected to cause reductions in cardiovascular and respiratory morbidity (Johansson et al., 2009).

Our study also shows that although the reduced NO_x emissions have caused increased ozone concentrations, which contribute to adverse health effects, the net effect will be a longer life expectancy in the population (Table 2). This may not be true if other health outcomes are analysed. Our focus on increased mortality should be seen as a limitation and underestimation of the total impact. The regulated air pollutants have been associated not only with respiratory and cardiovascular morbidity and mortality, but more recently also with systemic effects and metabolic diseases, pregnancy and developmental outcomes, as well as CNS and psychiatric effects (Thurston et al., 2017). Many of these effects are poorly represented by the studies of mortality.

Health impact assessments from other studies clearly suggest that public health could largely benefit from better air quality (e.g. Künzli, 2002). However, estimates of health benefits of reducing air pollution are dependent on the indicator that is used, and on the shape of the concentration-response functions (Pope et al., 2015). In addition, Pope et al. (2015) also discusses recent evidence that the shape of the concentration-response function may be supra linear (dose-response relationship with a negative second derivative) across wide ranges of exposure, as has been shown in the case of exposure to $PM_{2.5}$. It means that incremental pollution-abatement efforts may yield greater benefit in relatively clean areas compared to highly polluted areas. This motivates actions to be taken even in areas with relatively low levels of air pollutants.

5. Conclusions

The air pollution trends regarding NO_x, NO_2, and O_3 in Stockholm, Gothenburg, and Malmo exhibit significant (95 % CI) tendencies during the measured time periods. For PM_{10}, the trends in Stockholm and Gothenburg are significant, but the trend in Malmo is non-significant. The slopes of the trends are very different, where the NO_x and NO_2 concentrations in all cities exhibit decreasing trends, while O_3 in all cities exhibits increasing trends. For PM_{10}, the trends are less clear with a decreasing trend in Stockholm, an increasing trend in Gothenburg, and no significant trend in Malmo. When the trends are divided into weekdays and weekends, it follows that the trends for NO_x and NO_2 in all cities exhibit large differences, where the trends associated with weekdays exhibit sharper declines compared to the weekends. This phenomenon is consistent with the fact that local emission reductions mostly explain those declines, since the traffic intensity is more prominent during weekdays. An anti-correlation between NO_x and O_3 can be seen in all cities, which can be attributed to an increased or decreased titration effect, where NO has the capability of scavenging free oxygen atoms wherein NO_2 is formed. When the trends in this article are compared to the trends in other studies, the trends associated with NO_x and NO_2 are in the same magnitude as those measured in the U.S. and Europe as a whole, but the trends associated with PM_{10} are completely different, mostly caused by the large contribution of mechanically generated particles in the Swedish cities.

The change in life expectancy, associated with the air pollution trends, is most obvious for NO_x and NO_2, where up to about 20 % of the total increase in life expectancy during the period 1990–2015 can be attributed to decreasing NO_x trends. Since NO_x and NO_2 are indicators of combustion-related air pollutants in general, an overall conclusion is that exposure to these air pollutants are particularly important in terms of health effects and health benefits. Our study indicates that policies aiming at

reducing local emissions from road traffic have been most important for the increased life expectancy associated with urban air pollution in these three Swedish cities.

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7. Competing interests

The authors declare that they have no conflict of interest.

References

Airviro, 2017: http://www.airviro.smhi.se (2017-11-22)

Bari, M. A. and Kindzierski, W. B.: Evaluation of air quality indicators in Alberta, Canada - An international perspective, Environ. Int., 92-93, 119-129, doi: 10.1016/j.envint.2016.03.021, 2016

Beelen, R., Raaschou-Nielsen, O., Stafoggia, M., Andersen, Z. J., Weinmayr, G., Hoffmann, B., Wolf, K., Samoli, E., Fischer, P., Nieuwenhuijsen, M., Vineis, P., Xun, W. W., Katsouyanni, K., Dimakopoulou, K., Oudin, A., Forsberg, B., Modig, L., Havulinna, A. S., Lanki, T., Turunen, A., Oftedal, B., Nystad, W., Nafstad, P., De Faire, U., Pedersen, N. L., Östenson, C. G., Fratiglioni, L., Penell, J., Korek, M., Pershagen, G., Eriksen, K. T., Overvad, K., Ellermann, T., Eeftens, M., Peeters, P. H., Meliefste, K., Wang, M., Bueno-de-Mesquita, B., Sugiri, D., Krämer, U., Heinrich, J., de Hoogh, K., Key, T., Peters, A., Hampel, R., Concin, H., Nagel, G., Ineichen, A., Schaffner, E., Probst-Hensch, N., Künzli, N., Schindler, C., Schikowski, T., Adam, M., Phuleria, H., Vilier, A., Clavel-Chapelon, F., Declercq, C., Grioni, S., Krogh, V., Tsai, M. Y., Ricceri, F., Sacerdote, C., Galassi, C., Migliore, E., Ranzi, A., Cesaroni, G., Badaloni, C., Forastiere, F., Tamayo, I., Amiano, P., Dorronsoro, M., Katsoulis, M., Trichopoulou, A., Brunekreef, B., and Hoek, G.: Effects of long-term exposure to air pollution on natural-cause mortality: an analysis of 22 European cohorts within the multicentre ESCAPE project, Lancet, 383, 785-95, doi: 10.1016/S0140-6736(13)62158-3, 2014

BilSweden, 2018. http://www.bilsweden.se/statistik/arkiv-nyregistreringar_1 (2018-08-29)

Brunekreef, B. and Forsberg, B.: Epidemiological evidence of effects of coarse airborne particles on health, Eur. Respir. J., 26, 309-318, doi: 10.1183/09031936.05.00001805, 2005

Carslaw, D. C.: Evidence of an increasing NO_2/NO_X emissions ratio from road traffic emissions, Atmos. Environ., 39, 4793-4802, doi:10.1016/j.atmosenv.2005.06.023, 2005

Carslaw, D. C. and Ropkins, K.: Openair — an R package for air quality data analysis. Environmental Modelling & Software, 27–28, 52–61, 2012

Cohen, A. J., Brauer, M., Burnett, R., Anderson, H. R., Frostad, J., Estep, K., Balakrishnan, K., Brunekreef, B., Dandona, L., Dandona, R., Feigin, V., Freedman, G., Hubbell, B., Jobling, A., Kan, H., Knibbs, L., Liu, Y., Martin, R., Morawska, L., Pope, C. A. 3th, Shin, H., Stralf, K., Shaddick, G., Thomas, M., van Dingenen, R., van Donkelaar, A., Vos, T., Murray, C. J. L., and Forouzanfar, H.: Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015, Lancet, 389, 1907-1918, doi: 10.1016/ S0140-6736(17)30505-6, 2017

Colette, A., Granier, C., Hodnebrog, Ø., Jakobs, H., Maurizi, A., Nyiri, A., Bessagnet, B., D'Angiola, A., D'Isidoro, M., Gauss, M., Meleux, F., Memmesheimer, M., Mieville, A., Rouil, L., Russo, F., Solberg, S., Stordal, F., and Tampieri, F.: Air quality trends in Europe over the past decade: a first multi-model assessment, Atmos. Chem. Phys., 11, 11657–11678, doi: 10.5194/acp-11-11657-2011, 2011

5   Correia, A. W., Pope, C. A. 3rd, Dockery, D. W., Wang, Y., Ezzati, M., and Dominici, F.: Effect of air pollution control on life expectancy in the United States: an analysis of 545 U.S. counties for the period from 2000 to 2007, Epidemiology, 24, 23-31, doi: 10.1097/EDE.0b013e3182770237, 2013

EEA, 2016. Air quality in Europe — 2016 report, No 28/2016. European Environment Agency. ISSN 1977-8449

EEA, 2017. Air quality in Europe — 2017 report, No 13/2017. European Environment Agency. ISSN 1725-9177

10   EU, 2008. EU Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on Ambient Air Quality and Cleaner Air for Europe, OJ L 152, 11.6.2008

Fann, N. and Risley, H. The public health context for $PM_{2.5}$ and ozone air quality trends. Air. Qual. Atmos. Health, 6, 1-11, doi: 10.1007/s11869-010-0125-0, 2013

Faustini, A., Rapp, R., and Forastiere, F.: Nitrogen dioxide and mortality: review and meta-analysis of long-term studies, 15   Eur. Respir. J., 44, 744-753. doi: 10.1183/09031936.00114713, 2014

Geddes, J. A., Martin, R. V., Boys, B. L., and van Donkelaar, A.: Long-term trends worldwide in ambient NO2 concentrations inferred from satellite observations. Environ. Health Perspect., 124, 281-289, doi: 10.1289/ehp.1409567, 2016

Gidhagen, L., Johansson, C., Langner, J., and Olivares, G.: Simulation of NOx and ultrafine particles in a street canyon in 20   Stockholm, Sweden. Atmos. Environ., 38, 2029–2044, doi: 10.1016/j.atmosenv.2004.02.014, 2004

Gothenburg City, 2015. http://goteborg.se/wps/portal/start/miljo/miljolaget-i-goteborg/luft/luftkvaliteten-i-goteborg/!ut/p/z1/hY7RCoIwGEafpRfYv81m83IGhhppYai7CY21BHXhpIFPnz1A9N0dzrn4QEIFcmzenW7mzoxNv3It_VtOkjMPicDZIYhwXKR5dEqP2f5CofwXyFXjHxMYEpBdOyB3HxBGHqE-CRjdcY9uGWZQhlCn12Ip3feJGFuPa5CTeqhJTehp7AyVcw5pY3SvkFXwGqoltmLzAZX73Zg!/dz/d5/L2dBISEvZ0FBIS9 25   nQSEh/ (2016-06-14)

Gothenburg Annual Report, 2015. Miljöförvaltningen, Göteborgs stad. http://goteborg.se/wps/wcm/connect/48ca3c67-ced5-415f-b538-1ddf4f246e70/N800_R_2016_5.pdf?MOD=AJPERES (2016-06-07)

Gramsch, E., Cereceda-Balic, F., Oyola, P., and von Baer, D.: Examination of pollution trends in Santiago de Chile with cluster analysis of PM_{10} and ozone data. Atmos. Environ., 40, 5464–5475. doi: 10.1016/j.atmosenv.2006.03.062, 2006

30   Grice, S., Stedman, J., Kent, A., Hobson, M., Norris, J., Abbott, J., and Cooke, S.: Recent trends and projections of primary NO_2 emissions in Europe. Atmos. Environ., 43, 2154–2167, doi: 10.1016/j.atmosenv.2009.01.019, 2009

Grundström, M., Hak, C., Chen, D., Hallquist, M., and Pleijel, H.: Variation and co-variation of PM_{10}, particle number concentration, NO_x and NO_2 in the urban air – Relationships with wind speed, vertical temperature gradient and weather type, Atmos. Environ., 120, 317-327, doi: 10.1016/j.atmosenv.2015.08.057, 2015

35   Guerreiro, C. B. B., Foltescu, V., and de Leeuw, F.: Air quality status and trends in Europe. Atmos. Environ., 98, 376–384, doi: 10.1016/j.atmosenv.2014.09.017, 2014

Henschel, S., Atkinson, R., Zeka, A., Le Tertre, A., Analitis, A., Katsouyanni, K., Chanel, O., Pascal, M., Forsberg, B., Medina, S., and Goodman, P. G.: Air pollution interventions and their impact on public health, Int. J. Public Health, 57, 757-768, doi: 10.1007/s00038-012-0369-6, 2012

Hirsch, R. M., Slack, J. R., and Smith, R. A.: Techniques of trend analysis for monthly water-quality data, Water Resour. Res., 18, 107–121, doi: 10.1029/WR018i001p00107, 1982

Hoek, G., Krishnan, R. M., Beelen, R., Peters, A., Ostro, B., Brunekreef, B., and Kaufman, J. D.: Long-term air pollution exposure and cardio- respiratory mortality: a review, Environ. Health, 12, 43, doi: 10.1186/1476-069X-12-43, 2013

5   IVL, 2016. Swedish National environmental monitoring (in Swedish with English summary). Report C 224. IVL Svenska Miljöinstitutet AB, Box 210 60, 100 31 Stockholm, Sweden ISBN 978-91-88319-33-3. https://www.naturvardsverket.se/upload/miljoarbete-i-samhallet/miljoarbete-i-sverige/miljoovervakning/Luft/ivl-rapport-c224-nationell-luftövervakning-sakrapport2015.pdf (2018-09-04)

Jerrett, M., Burnett, R.T., Pope, C.A. 3rd, Ito, K., Thurston, G., Krewski, D., Shi, Y., Calle, E., and Thun, M.: Long-term
10 ozone exposure and mortality, N. Engl. J. Med., 12, 1085-95, doi: 10.1056/NEJMoa0803894, 2009

Johansson, C., Andersson, C., Bergström, R., and Krecl, P., 2008. Exposure to particles due to local and non-local sources in Stockholm - Estimates based on modelling and measurements 1997 – 2006. Report 175, Department of Applied Environmental Science, Stockholm university, 106 91 Stockholm, Sweden. http://slb.nu/slb/rapporter/pdf8/itm2008_175.pdf (2018-09-04)

15 Johansson, C., Burman, L., and Forsberg, B.: The effects of congestions tax on air quality and health, Atmos. Environ., 43, 4843-4854, doi: 10.1016/j.atmosenv.2008.09.015, 2009

Johansson, C., Löverheim, B., Schantz, P., Wahlgren, L., Almström, P., Markstedt, A., Strömgren, M., Forsberg, B., and Nilsson Sommar, J.: Impacts on air pollution and health by changing commuting from car to bicycle, Sci. Total. Environ., 584-585, 55-63, doi: 10.1016/j.scitotenv.2017.01.145, 2017

20 Johansson, C., Norman, M., and Gidhagen, L.: Spatial & temporal variations of PM10 and particle number concentrations in urban air, Environ. Monit. Assess., 127, 477-487, doi: 10.1007/s10661-006-9296-4, 2007

Keuken, M., Roemer, M., and van den Elshout, S.: Trend analysis of urban NO_2 concentrations and the importance of direct NO_2 emissions versus ozone/NO_x equilibrium. Atmos Environ., 43, 4780-4783, doi: 10.1016/j.atmosenv.2008.07.043, 2009

Keuken, M., Zandveld, P., van den Elshout, S., Janssen, N. A. H., and Hoek, G.: Air quality and health impact of PM10 and
25 EC in the city of Rotterdam, the Netherlands in 1985-2008. Atmos. Environ., 45, 5294-5301, doi: 10.1016/j.atmosenv.2011.06.058, 2011

Künzli, N., Kaiser, R., Medina, S., Studnicka, M., Chanel, O., Filliger, P., Herry, M., Horak, F. Jr., Puybonnieux-Texier, V., Quénel, P., Schneider, J., Seethaler, R., Vergnaud, J. C., and Sommer, H.: Public-health impact of outdoor and traffic-related air pollution: a European assessment, Lancet 356, 795-801, doi: 10.1016/S0140-6736(00)02653-2, 2000

30 Künzli, N.: The public health relevance of air pollution abatement, Eur. Respir. J., 20, 198-209, doi: 10.1183/09031936.02.00401502, 2002

Molnar, P., Stockfelt, L., Barregard, L., and Sallsten, G.: Residential NO_x exposure in a 35-year cohort study. Changes of exposure, and comparison with back extrapolation for historical exposure assessment, Atmos. Environ., 115, 62-69, doi: 10.1016/j.atmosenv.2015.05.055, 2015

35 Munir, S., Chen, H., and Ropkins, K.: Quantifying temporal trends in ground level ozone concentration in the UK, Sci. Total Environ, 458–460, 217–227, doi: 10.1016/j.scitotenv.2013.04.045, 2013

Nafstad, P., Håheim, L. L., Wisløff, T., Gram, F., Oftedal, B., Holme, I., Hjermann, I., and Leren, P.: Urban air pollution and mortality in a cohort of Norwegian men, Environ. Health. Perspect., 112, 610-615, doi: 10.1289/ehp.6684, 2004

Olsson, D., Johansson, C., Engström Nylén, A., and Forsberg, B., Air pollution at home and pregnancy outcomes. Presented at the 28th Annual Conference of the International Society For Environmental Epidemiology. Rome, Italy, Aug 31 – Sep 4., 2016 (manuscript in preparation April 2018)

Orru, H., Lövenheim, B., Johansson, C., and Forsberg, B.: Potential health impacts of changes in air pollution exposure associated with moving traffic into a road tunnel, J. Expo. Sci. Environ. Epidemiol., 25, 524-531, doi: 10.1038/jes.2015.24, 2015

Pope, C. A. 3rd, Cropper, M., Coggins, J., and Cohen, A.: Health benefits of air pollution abatement policy: Role of the shape of the concentration-response function. J. Air Waste. Manag. Assoc., 65, 516-522, doi: 10.1080/10962247.2014.993004, 2015

SCAC, 2018, http://www.scac.se/ (2018-09-07)

SCB, 2017, Statistics Sweden: http://www.statistikdatabasen.scb.se/pxweb/sv/ssd/?rxid=824e5e7c-a718-462e-9dd9-a83c29e37685 (2017-09-29)

Segersson, D., Eneroth, K., Gidhagen, L., Johansson, C., Omstedt, G., Engström Nylen, A., and Forsberg B.: Health Impact of PM_{10}, $PM_{2.5}$ and black carbon exposure due to different source sectors in Stockholm, Gothenburg and Umea, Sweden, Int. J. Environ. Res. Public. Health., 14, 742, doi: 10.3390/ijerph14070742, 2017

Sen, P. K.: Estimates of regression coefficient based on Kendall's tau.: J. Am. Stat. Assoc., 63, 1379-1389, doi: 10.1080/01621459.1968.10480934, 1968

SEPA, 2017a, Swedish Environmental protection agency: http://www.naturvardsverket.se/Sa-mar-miljon/Statistik-A-O/Ozon---marknara-halter-i-luft-urban-och-regional-bakgrund-arsmedelvarden/?visuallyDisabledSeries=7bbc8e1c95d277e1 (2017-11-16)

SEPA, 2017b, Swedish Environmental protection agency: http://www.naturvardsverket.se/Sa-mar-miljon/Statistik-A-O/Partiklar-PM10-halter-i-luft-regional-bakgrund-25/?visuallyDisabledSeries=ca9f2dd590183278 (2017-12-09)

Sicard, P., Serra, R., and Rossello, P.: Spatiotemporal trends in ground-level ozone concentrations and metrics in France over the time period 1999–2012, Environ. Res., 149, 122–144, doi: 10.1016/j.envres.2016.05.014, 2016

SLB, 1995. Air Pollutants in the county of Stockholm (only in Swedish). Stockholm air quality association, LVF 3:95. http://slb.nu/slb/rapporter/pdf8/lvf1995_003.pdf (2018-09-04)

SLB, 2015. Air Quality in Stockholm – annual report 2015 (summary in English). SLB Analys. Stockholms stad. http://slb.nu/slb/rapporter/pdf8/slb2016_002.pdf (2018-09-07)

SLB, 2018. Air Pollutants in the region of the Eastern Sweden air quality association (only in Swedish). LVF 2018:22. http://slb.nu/slb/rapporter/pdf8/lvf2018_022.pdf (2018-09-04)

Socialstyrelsen, 2017, The Swedish National Board of Health and Welfare: http://www.socialstyrelsen.se/statistik/statistikdatabas/dodsorsaker (2017-09-07)

State of Global Air, 2017. Health Effects Institute. 2017. Special Report. Boston, MA: Health Effects Institute: https://www.stateofglobalair.org/sites/.../SOGA2017_report.pdf (2017-12-18)

Stockfelt, L., Andersson, E. M., Molnár, P., Rosengren, A., Wilhelmsen, L., Sallsten, G., and Barregard, L.: Long term effects of residential NO(x) exposure on total and cause-specific mortality and incidence of myocardial infarction in a Swedish cohort, Environ. Res., 142, 197-206, doi: 10.1016/j.envres.2015.06.045, 2015

Stockfelt, L., Andersson, E.M., Molnár, P., Gidhagen, L., Segersson, D., Rosengren, A., Barregard, L., and Sallsten, G.: Long-term effects of total and source-specific particulate air pollution on incident cardiovascular disease in Gothenburg, Sweden, Environ. Res., 158, 61-71, doi: 10.1016/j.envres.2017.05.036, 2017

5   Tang, D., Wang, C., Nie, J., Chen, R., Niu, Q., Kan, H., Chen, B., Perera, F., and CDC, T.: Health benefits of improving air quality in Taiyuan, China, Environ. Internat., 73, 235–242, doi: 10.1016/j.envint.2014.07.016, 2014

Theil, H.: A rank invariant method of linear and polynomial regression analysis, Advanced studies in theoretical and applied econometrics, 23, 345-381, doi: 10.1007/978-94-011-2546-8_20, 1992

Thurston, G. D., Kipen, H., Annesi-Maesano, I., Balmes, J., Brook, R. D., Cromar, K., De Matteis, S., Forastiere, F., Forsberg, B., Frampton, M. W., Grigg, J., Heederik, D., Kelly, F. J., Kuenzli, N., Laumbach, R., Peters, A., Rajagopalan, S.
10 T., Rich, D., Ritz, B., Samet, J. M., Sandstrom, T., Sigsgaard, T., Sunyer, J., and Brunekreef, B.: A joint ERS/ATS policy statement: what constitutes an adverse health effect of air pollution? An analytical framework. Eur. Respir. J., 49, 1600419, doi: 10.1183/13993003.00419-2016, 2017

Tong, D. Q., Lamsal, L., Pan, L., Ding, C., Kim, H., Lee, P., Chai, T., Pickering, K. E., and Stajner, I.: Long-term NO_x trends over large cities in the United States during the great recession: Comparison of satellite retrievals, ground
15 observations, and emission inventories, Atmos. Environ., 107, 70–84, doi: 10.1016/j.atmosenv.2015.01.035, 2015

Turner, M. C, Jerrett, M., Pope, C. A. 3rd, Krewski, D., Gapstur, S. M., Diver, W. R., Beckerman, B. S., Marshall, J. D., Su J., Crouse, D. L., and Burnett, R. T.: Long-Term Ozone Exposure and Mortality in a Large Prospective Study, Am. J. Respir. Crit. Care. Med., 193, 1134-1142, doi: 10.1164/rccm.201508-1633OC, 2016

WHO, 2016a. WHO's Urban Ambient Air Pollution database - Update 2016.
20 http://www.who.int/phe/health_topics/outdoorair/databases/cities/en/ (2016-08-11)

WHO, 2016b. AirQ+: software tool for health risk assessment of air pollution. http://www.euro.who.int/en/health-topics/environment-and-health/air-quality/activities/airq-software-tool-for-health-risk-assessment-of-air-pollution (2017-11-22)

Xing, J., Pleim, J., Mathur, R., Pouliot, G., Hogrefe, C., Gan, C.-M., and Wei, C.: Historical gaseous and primary aerosol
25 emissions in the United States from 1990 to 2010, Atmos. Chem. Phys., 13, 7531–7549, doi: 10.5194/acp-13-7531-2013, 2013

Appendix A

Table A1: General facts about the three cities regarding population structure, life expectancy at birth, and baseline mortality in terms of the number of deaths per 100 000 inhabitants. The number of deaths per 100 000 inhabitants are age standardized according to the average population for the year 2000.

	Stockholm		Gothenburg		Malmo	
	1990	2015	1990	2015	1990	2015
Population size	1 040 907	1 515 017	465 474	572 799	223 663	301 706
Density of population (inhabitants per km²)	3 601	4 935	965	1 224	1 522	2 060

Expected life expectancy at birth	76.8	82.6	77.2	82.1	77.6	82.1
Baseline mortality (all causes, age standardized), number per 100 000 inhabitants.	1 157*	833	1 187*	862	1 162*	849

* Extrapolated from a linear regression based on data from 1997–2015
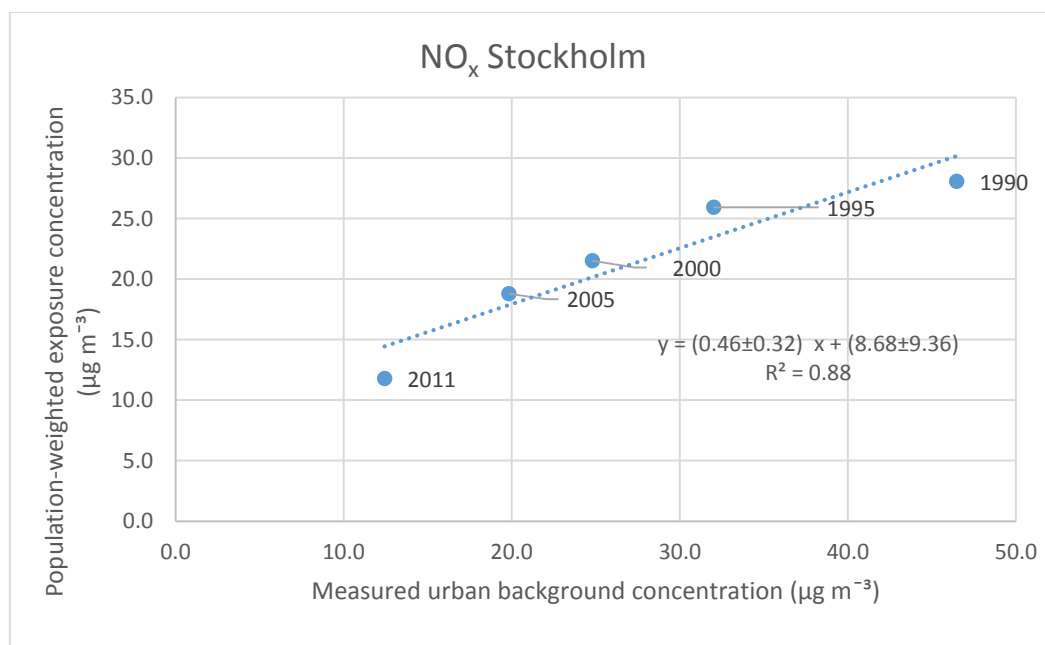
Figure A1. Relationship between measured urban-background concentrations and population-weighted exposure concentrations of NO_x in Stockholm. Slope and intercept are specified with 95 % confidence intervals.
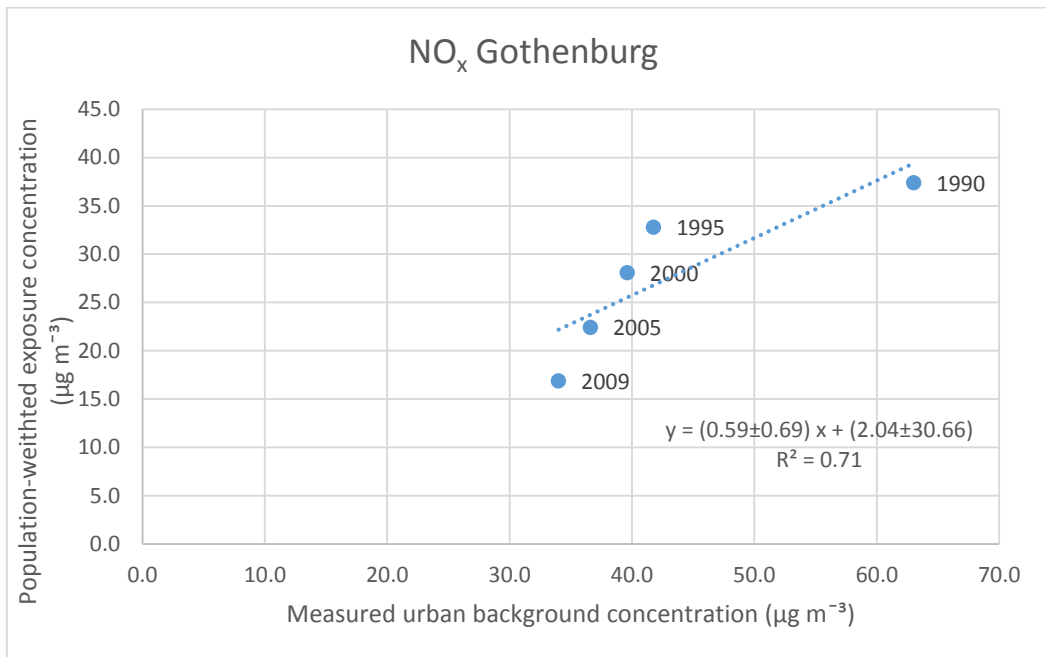
Figure A2. Relationship between measured urban-background concentrations and population-weighted exposure concentrations of NO_x in Gothenburg. Slope and intercept are specified with 95 % confidence intervals.
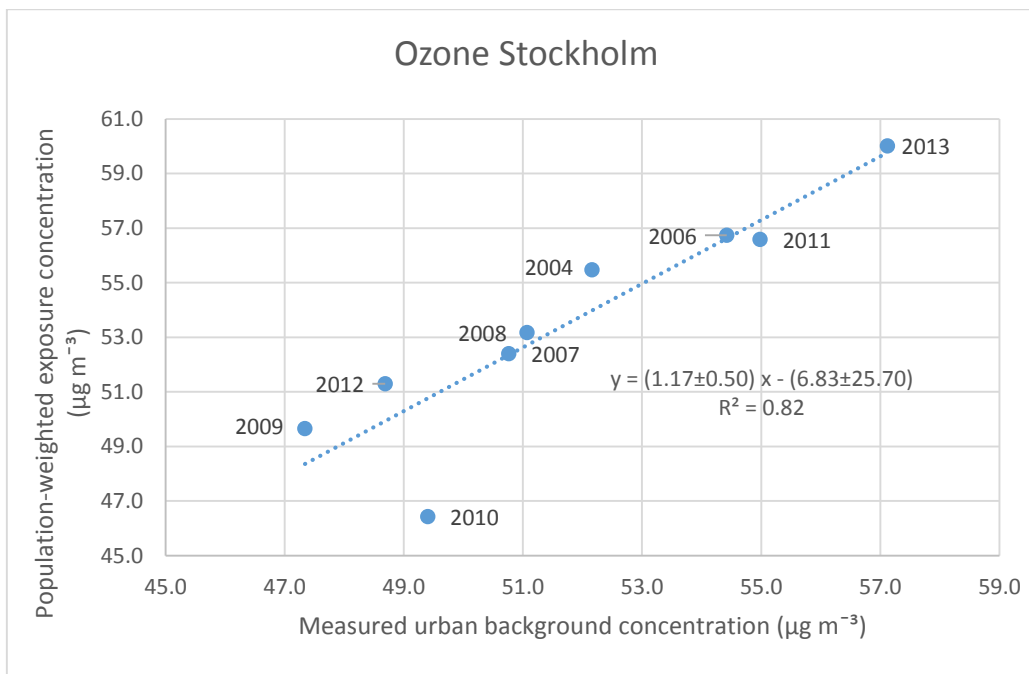
Figure A3. Relationship between measured urban-background concentrations and population-weighted exposure concentrations of O_3 in Stockholm. Slope and intercept are specified with 95 % confidence intervals.
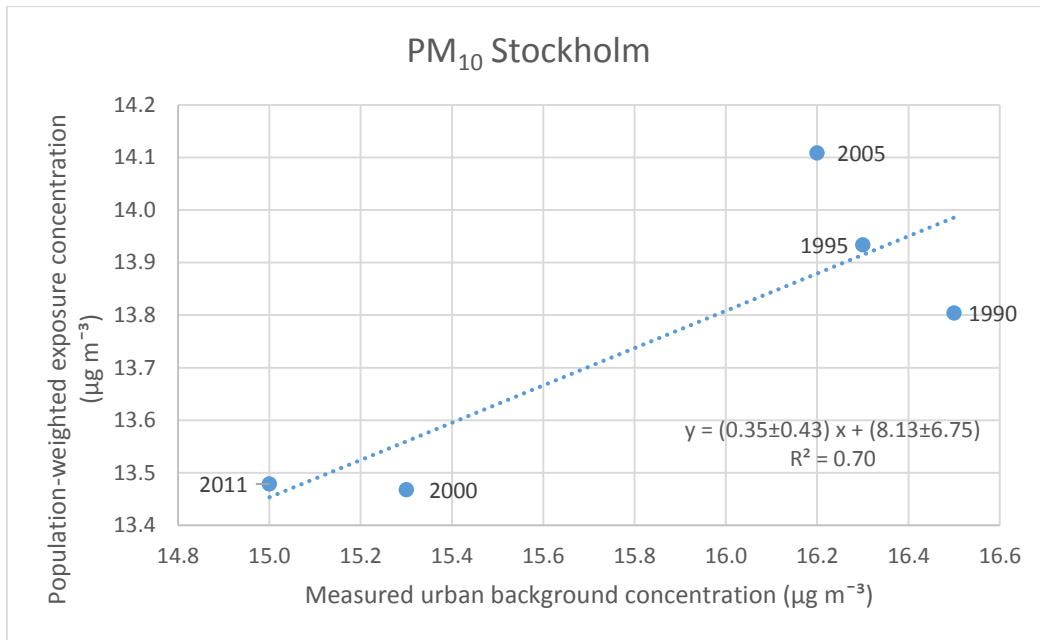
Figure A4. Relationship between measured urban-background concentrations and population-weighted exposure concentrations of PM_{10} in Stockholm. Slope and intercept are specified with 95 % confidence intervals.