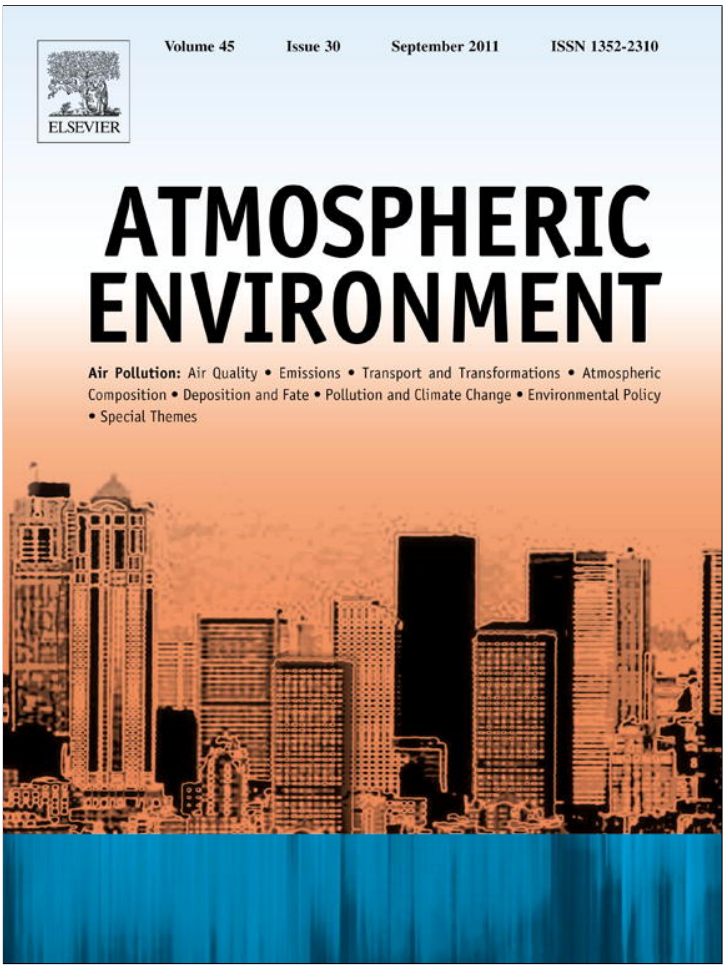

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# Air quality and health impact of $PM_{10}$ and EC in the city of Rotterdam, the Netherlands in 1985–2008

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ABSTRACT

A health impact assessment (HIA) of $PM_{10}$ and elemental carbon (EC) was performed for the period 1985–2008 in the city of Rotterdam. The spatial distribution of the concentrations was modeled by the URBIS model. The modeling results for 2008 were validated by $PM_{10}$ and EC measurements at various locations in Rotterdam. This paper describes the HIA related to improved air quality in the period 1985–2008: at urban background locations 18 $\mu g\ m^{-3}$ $PM_{10}$ and 2 $\mu g\ m^{-3}$ EC. The gain in life years saved due to long-term exposure to $PM_{10}$ and EC in this period was, respectively 13 and 12 month per person. The similar health impacts for $PM_{10}$ and EC suggests that reduction of combustion aerosol was important for the reduction in health impact of $PM_{10}$.



## 1. Introduction

Exposure to elevated levels of particulate matter (PM) has been associated with health effects in epidemiological surveys (Brunekreef and Holgate, 2002; Anderson, 2009). In urban areas the population is in particular exposed to PM emissions by road traffic related to exhaust emissions, tire/engine wear and re-suspension of road dust. Combustion aerosol in exhaust emissions mainly consists of submicron particles. These particles may penetrate deep into the respiratory system and have been associated with various health effects (Laden et al., 2000; Aalto et al., 2005; Sioutas et al., 2005; Fischer et al., 2009). Typically, combustion aerosol contains a large number of particles smaller than 100 nm ("ultrafine particles") but most of the mass is found in larger particles (Kittelson et al., 2001; Maricq, 2007). The latter particles originate from incomplete combustion and are mainly composed of elemental carbon (EC) and condensed organic compounds (OC).

In order to address the health impact of road traffic, a variety of international and local measures have been implemented over the years to reduce exhaust emissions. Examples are more stringent emission standards and environmental zoning in cities to prevent high emitters near residential areas in certain EU regions. Consequently, there is a need to assess the impact of these measures on air quality and health. Likely indicators are $PM_{10}$ and $PM_{2.5}$ as air quality standards for the mass of PM have been established. However, even near heavy traffic locations, $PM_{10}$ and $PM_{2.5}$ are dominated by regional background concentrations, especially in North-Western European countries (Sillanpää et al., 2006; Yin et al., 2010). The concentration of EC is considered a more appropriate indicator for dispersion of combustion emissions especially by diesel engines (Schauer, 2003).

In our study, we applied both $PM_{10}$ and EC as indicators to assess the trend in air quality and health over the period 1985–2008 in the city of Rotterdam (the Netherlands). The city of Rotterdam with a population of almost 600,000 inhabitants has the largest harbor of Europe. Consequently, road traffic is intensive with a relatively large contribution of heavy duty vehicles. In Sections 2 and 3, the approach of the study is detailed and in Section 4, the results are presented and discussed related to the trend in emission factors of $PM_{10}$ and EC by road traffic in the Netherlands, the spatial concentrations of $PM_{10}$ and EC in Rotterdam and the related HIA in the period 1985–2008. The conclusions of the study are presented in Section 5.

## 2. Study approach

The URBIS ("URBan environmental Information System") model has been applied to estimate the spatial distribution of annual

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average $PM_{10}$ and EC in the city of Rotterdam for the years 1985, 1995 and 2008. The URBIS model combines a street-canyon and line-source model to compute the contribution of emissions by urban traffic and motorways to air quality (Beelen et al., 2010). The spatial resolution of the URBIS model is a 10 × 10 $m^2$ grid up to 300 m or the housing façade along inner-urban road *and* up to 500 m near motorways.

The next step in the HIA (Ostro, 2004) is to combine the spatial distribution of annual average $PM_{10}$ and EC with GIS-based population distribution data. This provides information on the population exposure to long-term air pollution at house address. Though air pollution has been associated with both mortality and morbidity effects, quantitatively the effects of mortality have been shown to be most important in previous HIA's (Künzli et al., 2000). Mortality effects of long-term exposure are substantially larger than mortality effects related to short-term daily exposures (Künzli et al., 2000; WHO, 2006). Therefore, concentration-response function (Janssen et al., submitted for publication) for long-term health effects of $PM_{2.5}$ and EC have been applied to the established population exposure maps in Rotterdam. Finally, the trend in health effects in the period 1985–2008 is evaluated in relation to exposure to $PM_{10}$ and EC using life-table analyses (Miller and Hurley, 2003).

## 3. Modeling and measurements

### 3.1. Modeling

The required input for dispersion modeling of $PM_{10}$ and EC concerns:

- *regional and urban background*; The regional and urban background of $PM_{10}$ in the Netherlands, including the Rotterdam area is annually updated for the previous year based on measurements in the National Air Quality Monitoring Network in combination with dispersion modeling at a 1 × 1 $km^2$ grid (PBL, 2008). This network consists of eighteen regional background stations, eight urban background stations and sixteen street locations. $PM_{10}$ is measured by automatic (beta-attenuation) monitors for which equivalence with the reference method (CEN, 1998) has been demonstrated (Beijk et al., 2008). The trend in $PM_{10}$ concentrations as measured at regional, urban and traffic locations in the period 1992–2008 in the Netherlands is illustrated in Fig. 1.

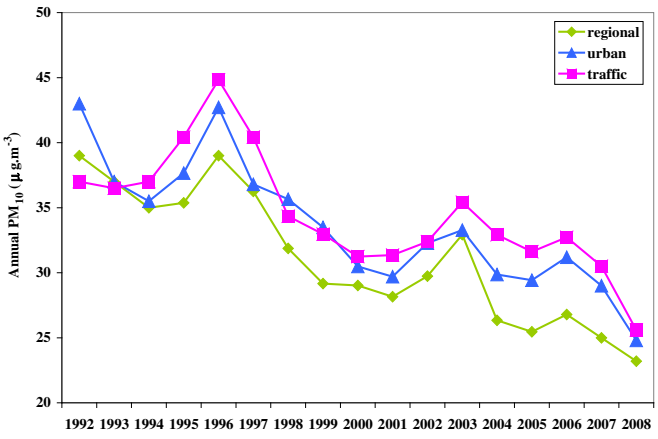


**Fig. 1.** Annual average $PM_{10}$ (µg $m^{-3}$) at regional, urban and traffic locations in the Netherlands 1992–2008 (Beijk et al., 2009).

Fig. 1 concerns average trends for the Netherlands and not specifically for Rotterdam. In view of the limited size of the Netherlands (~65,000 $km^2$) and relatively small differences between regional, urban and traffic stations in the Netherlands, these trends are considered representative for Rotterdam. For the Netherlands, an increasing trend of 0.7 µg $m^{-3}$ $PM_{10}$ per year for previous years has been estimated (Hoogerbrugge et al., 2010). Based on these trends, the regional and urban background in 1985 for $PM_{10}$ at 1 × 1 $km^2$ near and in Rotterdam was extrapolated.

The regional and urban background of EC was based on time series of Black Smoke measurements in and near Rotterdam in the period 1985–2008. A factor 10 was applied to convert Black Smoke to EC concentrations based on parallel measurements in 2006–2007 of Black Smoke and EC in the city of Rotterdam. This is elaborated in Section 3.2;

- *meteorological conditions*; The ten-year average meteorological conditions for the period 1995–2004 have been used for all three years 1985, 1995 and 2008 to eliminate the effect of meteorological variation on the HIA;
- *traffic data*; Actual traffic data on the motorways and main urban roads for the years 1985, 1995 and 2008 were available in Rotterdam. The data for 1985 and 1995 concerned aggregated traffic volume on motorways and main inner-urban roads. For 2008 more detailed information from the municipal traffic department was available which was used to further elaborate the 1985 and 1995 traffic data;
- *emission factors*; PM emission factors for road traffic (e.g. exhaust and friction emissions) are annually updated by the National Environmental Agency in the Netherlands. For the period 1990–2008, emission factors are available for the car fleet in the Netherlands (PBL, 2008). These emission factors were linearly extrapolated to obtain emission factors for 1985. For EC, emission factors were derived from an EU database with information on EC as a fraction of PM *exhaust* emissions (Ntziachristos and Samaras, 2009). This database was combined with PM exhaust emission factors in the Netherlands to compute EC emission factors for 1985, 1995 and 2008. The establishment of emission factors is further elaborated in Section 4.1.

### 3.2. Measurements

Black Smoke measurements in the period 1985–2008 have been used to estimate urban and regional background concentrations of EC. The Black Smoke method is based on optical measurement of the reflectance of PM sampled on a filter (Whatman 1), which is converted to mass units by the Black Smoke Index (ISO 9835, 1993). In Rotterdam Black Smoke measurements at the regional location and the urban location were performed by respectively, the National Institute for Health and Environment (RIVM) and the Regional Environmental Agency (DCMR) both following similar QA/QC accreditation procedures. This assured good quality over the whole period from 1985 to 2008. From 2003 onwards, the manual Black Smoke measurements were exchanged with automatic measurements by the SX200 (ETL, Hereford, UK). The manual and automatic have been shown to be equivalent (Green et al., 2007).

In order to compare the Black Smoke results with thermal EC measurements, two-weekly parallel samples were collected at regional, urban and traffic sites in Rotterdam in 2006–2007. The samples for thermal EC analysis were collected on pre-fired quartz filters (QMA, Pallflex) and analyzed following the EUSAARII protocol (Cavalli et al., 2010). The results are presented in Fig. 2.

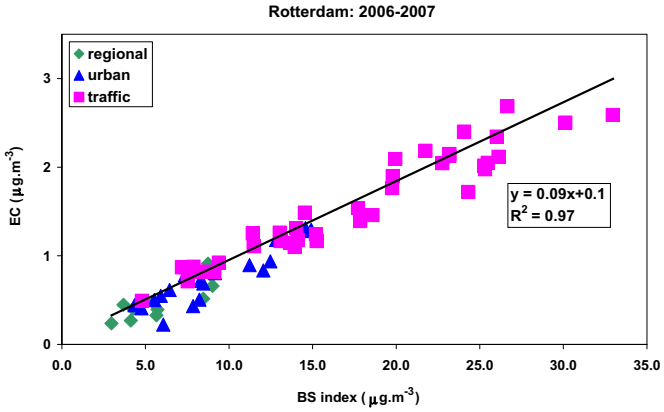


**Fig. 2.** Two-weekly average thermal EC (μg $m^{-3}$) and Black Smoke index at regional, urban and traffic locations near and in Rotterdam in 2006–2007.

Fig. 2 illustrates that EC is linear correlated with the Black Smoke index at various locations in and near Rotterdam. The linear regression has a slope 0.09, an intercept near zero and a correlation coefficient ($R^2$) 0.97. This is in agreement with other studies in the Netherlands (Schaap and Denier van der Gon, 2007) where a similar slope was found for urban and traffic locations. Different slopes have been found for background and traffic sites but it is difficult to compare results from different studies due to different methods used for the determination of EC and Black Smoke or Black Carbon (Cyrys et al., 2003). In our study, a conversion factor 10 for all locations has been used to derive EC concentrations from Black Smoke measurements. At two traffic stations in Rotterdam ("Floreslaan" and "Vasteland") and a regional station 15 km south of Rotterdam ("Westmaas"), annual average Black Smoke data are available for the period 1985–2008 (Beijk et al., 2009). The relation presented in Fig. 2, has been used to derive the EC trend in this period and shown in Fig. 3.

Fig. 3 shows a decreasing trend in EC concentrations both at the urban, traffic locations and the regional location. The *delta* between the average EC at the traffic locations and the regional background decreased in particular in the period 1995–2003. This reflects in particular the introduction of catalytic convertors in the nineties and emission reduction in combustion aerosol by road traffic in Europe. This measure had a relatively larger impact on EC concentrations at the traffic locations than the regional background. Since 2003, no further decreasing trend is detected neither at the regional nor at the traffic locations. This indicates that further reduction of exhaust emissions by "cleaner" vehicles do not longer compensate the growth in traffic volume in Rotterdam. From Fig. 3 it was estimated that the annual urban background for EC in Rotterdam was 2.6–3.2 μg $m^{-3}$ (1985), 1.1–2.6 μg $m^{-3}$ (1995) and 0.7–1.1 μg $m^{-3}$ (2008). These values have been used as input for dispersion modeling in Section 4.2.

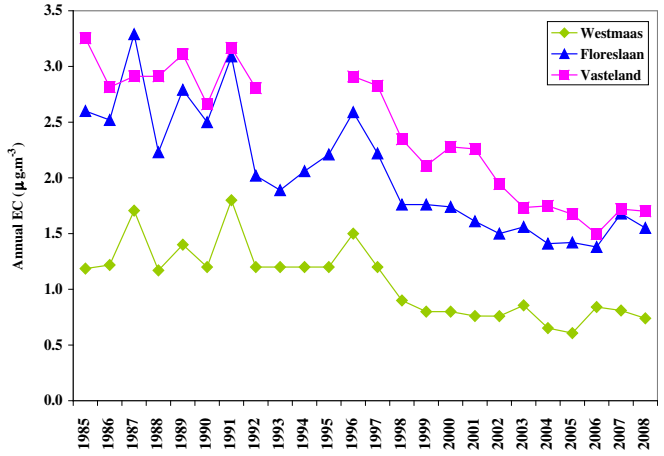


**Fig. 3.** Annual EC (μg $m^{-3}$) at a regional (Westmaas) near and two traffic locations (Floreslaan and Vasteland) in Rotterdam derived from Black Smoke index monitoring in 1985–2008.

## 4. Results and discussion

### 4.1. Emissions of $PM_{10}$ and EC by road traffic in 1985–2008

The mass of exhaust emissions has been regulated by EU emission limit values since 1992. The PM emission limits for vehicles have decreased for diesel-fueled light duty vehicles with a weight under 3.5 ton (LDV) from 140 to 25 mg $km^{-1}$ (Euro 1–5) and for heavy duty vehicles with a weight over 3.5 ton (HDV) from 360 to 20 mg $kWh^{-1}$ (Euro I–V). Emission factors of exhaust emissions are established by dynamometer testing. This concerns specific driving cycles to simulate typical driving conditions at different road types. Though, emission factors derived from driving cycles may not fully represent real-life driving conditions, this decreasing trend in emission factors reflects the actual trend in decreasing exhaust emissions per vehicle kilometer mainly by technical measures such as catalytic convertors. Emission factors may also take into account non-exhaust emissions of particulates as a result of friction processes by tires and brakes.

For EC there are no emission limits for road traffic and (thus) emission factors for EC have not been established. However, the fraction of EC in PM of exhaust emissions has been derived from dynamometer testing (Ntziachristos and Samaras, 2009). The EC fraction was relatively constant for vehicles meeting emission limits from 1992 till date: for petrol-LDV, diesel-LDV and HDV respectively 20%, 80% and 70%. For the period before 1992, the EC fraction was significantly lower respectively 2%, 55% and 50%. A study in the USA (EPA, 2008) concluded however that "*no model year or age dependency on the EC/PM ratio in period 1975–2005 was measured*". Therefore, in our study the pre-Euro standards are based on an average of the overall estimates by Ntziachristos and Samaras (2009). This resulted in EC fractions for petrol-LDV, diesel-LDV and HDV of 10%, 65% and 60%. Road tunnel and road side measurements have also been used to establish emission factors for EC. The range in EC fractions of $PM_{2.5}$ was from 20 to 70% depending on the amount of heavy duty vehicles (Gramotnev et al., 2003; Ning et al., 2008). These experimental data from actual road traffic agree with the results from dynamometer testing. Therefore, it is concluded that the dynamometer data from Ntziachristos and Samaras (2009) are a valid basis to estimate EC emission factors from PM emission factors for road traffic. The results for the Netherlands are presented in rounded figures in Table 1.

Table 1 shows that both PM and EC emission factors have decreased significantly for light and heavy duty vehicles in the period 1985–2008. The number of kilometers driven by road traffic in the Netherlands in the period 1985–2008 is presented in Fig. 4.

Fig. 4 shows that the total number of vehicle kilometers in the Netherlands almost doubled in the period 1985–2008: from 68 billion kilometers in 1985 to 124 billion kilometers in 2008. The percentage of diesel-fueled vehicles increased from 30% to 50% in this period. The total exhaust emissions of PM and EC by road traffic in the Netherlands are computed from the product of the emission factors (Table 1) and the total number of vehicle kilometers (Fig. 4). The results for the period 1985–2008 are presented separately for urban and non-urban roads (e.g. motorways and regional roads) in

**Table 1**
Emission factors of PM and EC (mg km$^{-1}$) in exhaust emissions of road traffic on urban roads and motorways in the Netherlands in the period 1985–2005.

| | PM (mg km$^{-1}$) | | EC/PM (%) | EC (mg km$^{-1}$) | |
|---|---|---|---|---|---|
| | Urban | Motorway | | Urban | Motorway |
| 1985 | | | | | |
| LDV (20% diesel)[a] | 250 | 50 | 25 | 65 | 15 |
| HDV | 1450 | 650 | 60 | 870 | 390 |
| 1995 | | | | | |
| LDV (30% diesel)[a] | 110 | 40 | 40 | 45 | 15 |
| HDV | 985 | 460 | 70 | 690 | 320 |
| 2008 | | | | | |
| LDV (50% diesel)[a] | 60 | 30 | 47 | 30 | 15 |
| HDV | 270 | 120 | 70 | 190 | 85 |

[a] % diesel from total number of transport kilometers by LDV in the Netherlands.

the Netherlands in Fig. 5. The fractions of vehicle kilometers on urban and non-urban roads are based on data for 2005, which are for personal cars/(HDV plus vans) respectively 25/75% (urban) and 10/90% (non-urban).

Fig. 5 illustrates that exhaust emissions of PM and EC on urban roads have been reduced by 50% while at non-urban roads (e.g. mainly motorways) these emissions did not decrease in the period 1985–2008. These trends in exhaust emissions are in agreement with the trend in EC concentrations at urban and regional locations, as presented in Fig. 3. The reduction in exhaust emissions are mainly attributed to technical measures such as catalytic convertors and improved engines and fuels. The differences in trends for urban and non-urban roads are related to differences in the growth of vehicle kilometers in the Netherlands at urban and non-urban roads of +1.5% and +3% per year, respectively (www.cbs.nl). Thus, on urban roads "cleaner" vehicles resulted in decreasing exhaust emissions by road traffic, while at non-urban roads exhaust emissions remained constant as the increase of traffic volume was balanced by lower emissions per vehicle. The percentage EC in the mass of exhaust emissions for all road traffic in the Netherlands increased from 47% in 1985 to 68% in 2008. This increase in EC content reflects especially the growth of diesel vehicles in the car fleet of the Netherlands as illustrated in Fig. 4. More than 80% of population in the Netherlands lives in urban areas with relatively high exposure to road traffic emissions. Therefore, the effects of these emissions were investigated on urban air quality and associated health effects in the city of Rotterdam.

### 4.2. $PM_{10}$ and EC in Rotterdam in 1985, 1995 and 2008

Based on the emission factors and urban background of $PM_{10}$ and EC, the spatial distribution for $PM_{10}$ and EC has been modeled with

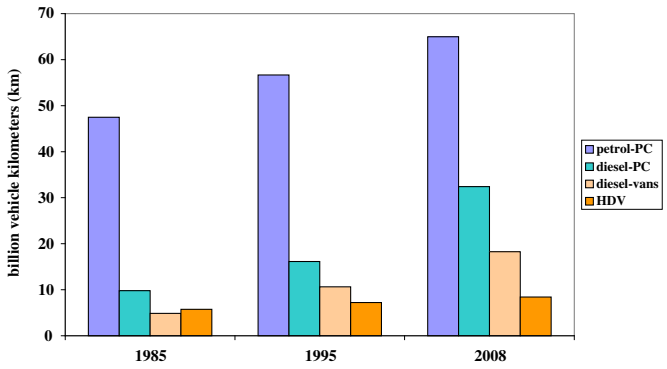


**Fig. 4.** The number of kilometers by petrol-personal cars (PC), diesel-PC, diesel-vans and heavy duty truck (HDV) in the Netherlands in the period 1985–2008 (source: Central Office of Statistics: www.cbs.nl).

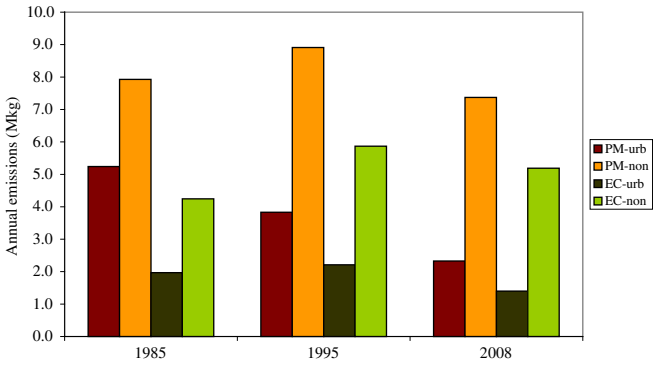


**Fig. 5.** Annual exhaust emissions (million kg) of PM and EC by road traffic on urban ("urb") and non-urban ("non") roads in the Netherlands in the period 1985–2008.

the URBIS model for the years 1985, 1995 and 2008 in Rotterdam. The urban and regional background of $PM_{10}$ – based on measurements and modeling for the Rotterdam area as discussed in Section 3.1 – takes into account all sources other than road traffic (e.g. industries, shipping, refineries and natural sources) and secondary PM. The emission factors for $PM_{10}$ were based on the PM in exhaust emissions in Table 1, but augmented with non-exhaust emissions by friction processes (e.g. brakes and tires). This increased the emission factors with 10 mg km$^{-1}$ for light duty vehicles, 50 mg km$^{-1}$ for middle duty vehicles and 70 mg km$^{-1}$ for heavy duty vehicles (PBL, 2008). In Figs. 6A–C and 7A–C the results are presented in 1985, 1995 and 2008 for respectively $PM_{10}$ and EC. In these figures, the motorways in Rotterdam are presented around the city center and extensions to the north, south, east and west. Also, the river "Oude Maas" is presented with harbor areas in the west.

Figs. 6 and 7 illustrate that the air quality for $PM_{10}$ and EC has improved significantly in the period 1985–2008 in the city of Rotterdam. The results for all years 1985, 1995 and 2008 are summarized in Table 2. The concentrations at the regional and urban background are based on a combination of modeling and measurements (see Section 3.1), while the concentrations at traffic locations are modeled by the URBIS model.

The results in Table 2 show that the urban background for $PM_{10}$ decreased from average 43 μg m$^{-3}$ in 1985 to average 25 μg m$^{-3}$ in 2008. This is caused for 70% by large-scale emission reduction by industry, energy production and road traffic of *precursors* (sulfur dioxide, ammonia and nitrogen oxides) for secondary particles (Hoogerbrugge et al., 2010). Only 10% reduction of $PM_{10}$ is related to primary combustion emissions and the remaining 20% by secondary organic aerosols and less water adsorbed to $PM_{10}$ particles.

Table 2 demonstrates that similar to $PM_{10}$ the air quality for EC also improved significantly in the period 1985–2008 and in particular from 1995 onwards. This is attributed to lower regional background concentrations as a result of reduced emissions of soot particles by combustion processes in general (e.g. industry, energy production, shipping and road traffic) and at the urban scale especially of exhaust emissions by road traffic after 1995. Consequently, the urban background of EC decreased from average 3 μg m$^{-3}$ in 1985 to average 1 μg m$^{-3}$ in 2008.

### 4.3. Validation of modeled air quality in Rotterdam in 2008

For the year 2008, the spatial distribution of $PM_{10}$ and EC in the city of Rotterdam modeled by the URBIS model were validated by measurements at an urban background and traffic locations in the monitoring network of the environmental protection agency (DCMR) in Rotterdam. The results are presented in Table 3.

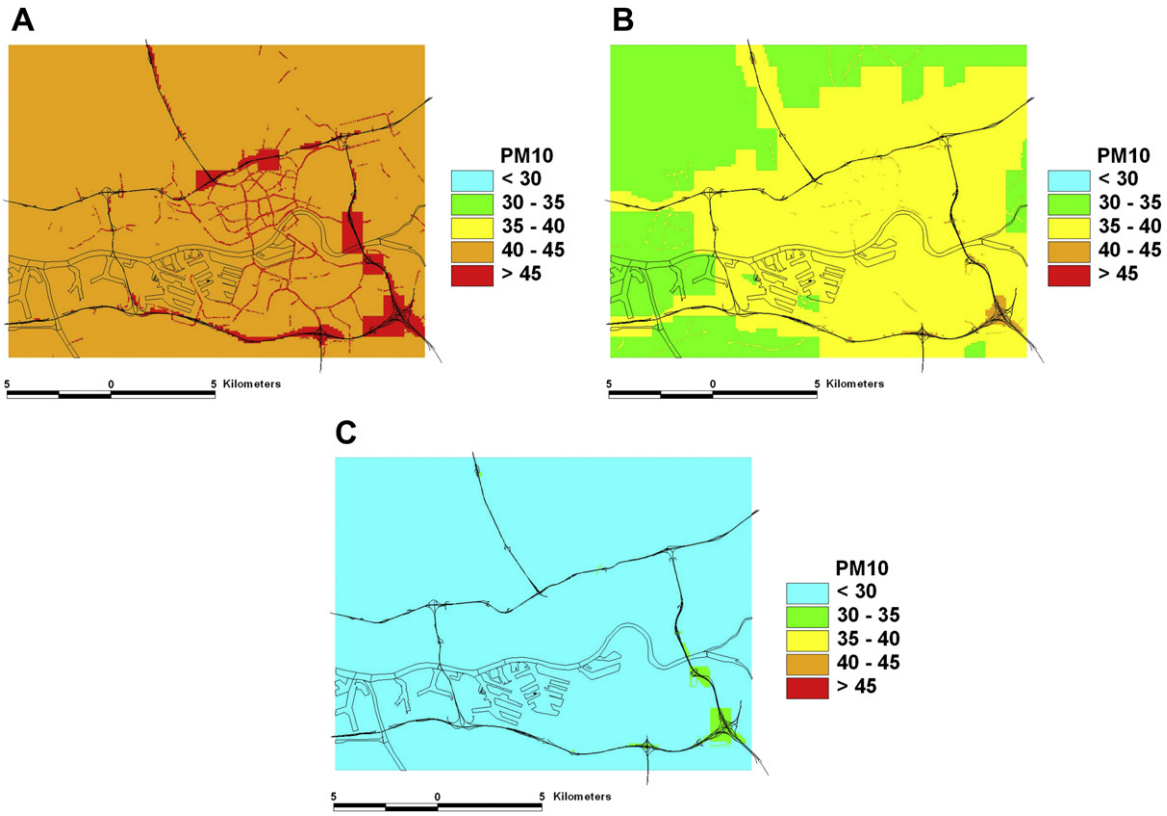


**Fig. 6.** A. Annual average concentrations of $PM_{10}$ (μg $m^{-3}$) in Rotterdam (1985). 6B: Similar as Fig. 6A for the year 1995. 6C: Similar as 6A for the year 2008.

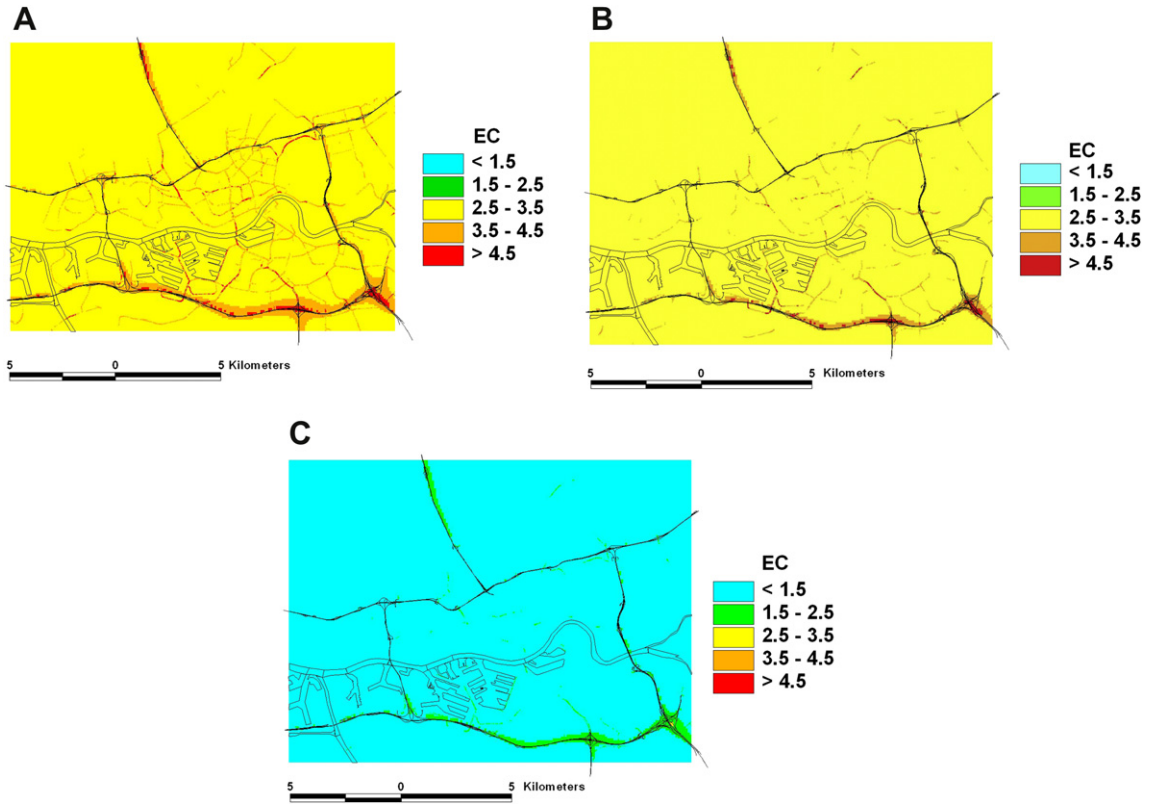


**Fig. 7.** A. Annual average concentrations of EC (μg $m^{-3}$) in Rotterdam (1985). 7B: Similar as Fig. 7A for the year 1995. 7C: Similar as Fig. 7A for the year 2008.

**Table 2**
Annual average $PM_{10}$ and EC concentrations ($\mu g\ m^{-3}$) at regional, urban and traffic locations in Rotterdam in 1985, 1995 and 2008.

| | $PM_{10}$ | | | EC | | |
|---|---|---|---|---|---|---|
| | Regional | Urban | Traffic | Regional | Urban | Traffic |
| 1985 | 40–45 | 40–45 | 40–55 | 2.5–3.5 | 2.5–3.5 | 3.5–6 |
| 1995 | 30–35 | 35–40 | 35–45 | 2.5–3.5 | 2.5–3.5 | 3.5–5 |
| 2008 | 20–23 | 23–27 | 27–35 | 0.5–0.8 | 0.8–1.2 | 1.2–3 |

The uncertainty in monitoring annual concentrations is in the order of 15%, while for modeling, the uncertainty is in the range of 25–40% for an urban background and road side location, respectively (Denby et al., 2010). Considering these uncertainties, the results in Table 3 show good agreement between modeled and monitoring annual averages for $PM_{10}$. For EC, the differences between modeling and monitoring results are relatively larger than for $PM_{10}$. This may be explained by a.) underestimation in the model of the urban background by a too large conversion factor 10, of Black Smoke to EC concentrations (see: Section 3.2) and/or b.) overestimation of the measured background by the MAAP instrument due to the *default* calibration by thermal EC analysis following the German "VDI" protocol. This protocol does not correct *artifact* formation of EC during thermal analysis by optical transmittance as developed by Birch and Cary (1996). Our study underlines that more experimental data on EC is required to further improve modeling of dispersion EC in urban areas.

### 4.4. Health impact assessment of $PM_{10}$ and EC in 1985, 1995 and 2008

#### 4.4.1. Population density in Rotterdam in 1985–2008

To investigate the health impact assessment in 1985–2008, we have maintained a constant figure for the distribution of the population and the number of inhabitants of Rotterdam at 570,000 in 2008. This number of inhabitants per *X*, *Y*-coordinate in the URBIS modeling domain have been combined with the air quality for $PM_{10}$ and EC in the period 1985–2008. The exposure of the population in Rotterdam has been classified to various levels of $PM_{10}$ and EC and presented in Table 4.

Table 4 shows that the population in Rotterdam has been exposed to significant lower air pollution in the period 1985–2008 following the improved air quality.

#### 4.4.2. Health effects of $PM_{10}$ and EC in Rotterdam in 1985–2008

In general, a health impact assessment (HIA) of outdoor air pollution by $PM_{10}$ and EC is based on four components (Ostro, 2004):

1. an assessment of the ambient air concentrations of $PM_{10}$ and EC by monitoring or model-based estimation;
2. a determination of the size of the population exposed to specific concentrations of $PM_{10}$ and EC;
3. a determination of the health effect of prime interest, including the baseline rate of the health effect being estimated (e.g. the underlying mortality rate in the population in deaths per thousand people);
4. a derivation and application of concentration-response functions from the epidemiological literature that relate ambient concentrations of $PM_{10}$ and EC to selected health effects.

**Table 3**
Measurements and modeling results of the annual average $PM_{10}$ and EC at locations in Rotterdam (2008).

| | $PM_{10}$ ($\mu g\ m^{-3}$) | | EC ($\mu g\ m^{-3}$) | |
|---|---|---|---|---|
| | Model | Monitoring | Model | Monitoring |
| Urban background "Schiedam" | 27.9 | 25.7[a] | 1.0[c] | 2.0[c] |
| Traffic location "Floreslaan" | 25.6 | 27.2[a] | 1.2 | 1.6[d] |
| Traffic location "Vasteland" | 28.9 | n.a. | 1.4 | 1.7[d] |
| Motorway location "Ridderkerk" | 30.7 | 28.3[b] | 2.2 | n.a. |

n.a.: not available.
[a] Gravimetric analysis.
[b] Tapered element oscillating monitor – TEOM corrected with 1.3.
[c] Multi angle absorption photometer – MAAP.
[d] Conversion from Black Smoke index.

**Table 4**
The number of inhabitants in Rotterdam exposed to various levels of annual average $PM_{10}$ and EC in the period 1985–2008.

| | Number of inhabitants (#) | | |
|---|---|---|---|
| | 1985 | 1995 | 2008 |
| $PM_{10}$ ($\mu g.m^{-3}$) | | | |
| 20–30 | – | – | 565,000 |
| 30–35 | – | 15,000 | 5000 |
| 35–40 | – | 550,000 | – |
| 40–45 | 515,000 | 5000 | – |
| 45–55 | 55,000 | – | – |
| EC ($\mu g\ m^{-3}$) | | | |
| 0.5–1.5 | – | – | 565,000 |
| 1.5–2.5 | – | – | 5000 |
| 2.5–3.5 | 535,000 | 560,000 | – |
| 3.5–4.5 | 30,000 | 10,000 | – |
| 4.5–6 | 5000 | – | – |

Population exposure distributions (steps 1 and 2) were taken from Table 4. Health impact calculations were performed for the midpoints of the exposure categories and a rounded value just below the lowest category and above the highest category. We focus on quantification of mortality effects of long-term exposure by exposure response functions based upon long-term exposure studies. These functions were selected from a recent review of the evidence for $PM_{2.5}$ and EC (Janssen et al., submitted for publication). For $PM_{2.5}$, we used a relative risk (RR) 1.007 (95% confidence interval 1.002–1.011) expressed per 1 $\mu g\ m^{-3}$. For EC we used RR 1.06 (95% confidence interval 1.02–1.10) expressed per 1 $\mu g\ m^{-3}$. Note that the RR of EC exceeds the RR of $PM_{2.5}$ substantially. The RR for $PM_{2.5}$ was used as the pooled effect estimates for long-term exposure on mortality were only established for $PM_{2.5}$. We assumed that we could apply the $PM_{2.5}$ exposure response function to the Rotterdam case, even though exposure was characterized as $PM_{10}$. This may be problematic for the calculation of the health impact for a particular year, but even more appropriate for the calculation of differences between years as most decrease in $PM_{10}$ in the Netherlands is due to a decrease of the fine fraction of $PM_{10}$ (Hoogerbrugge et al., 2010).

We have expressed mortality impacts in life years gained or lost estimated with life-table calculations (Miller and Hurley, 2003). For the calculation we used a population of 500,000 people aged 18 to 64, distributed in age categories comparable to the 2008 Dutch population. We have estimated *the effects on this population for a lifetime*, as follows: For 1985, 1995 and 2008 we first calculated the life years lost related to the exposure distribution in that year. We then subtracted the life years lost in 1995 and 2008 from the life years lost in 1985 to calculate the gain in life years related to a decrease in concentration. The outcome of the health impact assessment is that the decrease in $PM_{10}$ concentration from 1985 to 2008 results in a gain in life of on average 13 months per person with a range of 7–20 months. The range was obtained by applying the lower and upper confidence limit for the summary relative risk. For EC, a gain of 12 months per person is calculated with a range of 4–20 months. These gains in life years may be evaluated against a loss of life years in 1985 due to $PM_{10}$ of 31 months and for EC of 18 months.

The health impact is similar for $PM_{10}$ and EC. This is explained as follows. The population weighted $PM_{10}$ concentration dropped from 43 μg $m^{-3}$ in 1985 to 25 μg $m^{-3}$ in 2008 which is equivalent to 18 μg $m^{-3}$ $PM_{10}$. EC "only" decreased from 3 to 1 μg $m^{-3}$ over the same time period. As aforementioned in Section 4.2, the decrease in $PM_{10}$ in the last twenty years in the Netherlands is for 70% due to secondary inorganic aerosol and only for 10% due to EC (Hoogerbrugge et al., 2010). The latter means 10% of the 18 μg $m^{-3}$ drop in $PM_{10}$ which is equivalent with 1.8 μg $m^{-3}$ drop in EC. This is in good agreement with the drop of 2 μg $m^{-3}$ EC in the current study. Expressed per μg $m^{-3}$, the excess risk is 10 times larger for EC than for $PM_{10}$. This explains why a ten time higher decrease in $PM_{10}$ has similar health impact as EC. EC has been identified as one of the more health relevant components of PM (Janssen et al., submitted for publication). Because of the uncertainty in the summary relative risk estimates, the similarity in calculated health impacts in our study should not be interpreted as implying that reduction of other components such as secondary inorganic aerosol has not resulted in any benefit. More in general, it is concluded that EC is a more sensitive indicator (compared to $PM_{10}$ or $PM_{2.5}$) to monitor the health effects of traffic or other primary combustion aerosol measures. It is noted, that EC is likely *not* causing the health effects but acts as a proxy for the mass of combustion aerosol, including (toxic) metals and organic compounds.

## 5. Conclusions

In the period 1985–2008, road traffic in the Netherlands almost doubled in vehicle kilometers. Despite this growth in traffic volume, exhaust emissions of PM and EC on urban roads have been reduced by 50%, while at regional roads (e.g. motorways) these emissions did not decrease in the period 1985–2008. It is concluded that in urban areas "cleaner" vehicles resulted in less emissions by road traffic, while at regional roads lower emissions per vehicle were balanced by the increase in traffic volume. The percentage EC in the mass of exhaust emissions for all road traffic in the Netherlands increased from 47% in 1985 to 68% in 2008 which is attributed to the growth of diesel-fueled vehicle kilometers from 30% to 50% in this period.

A health impact assessment was performed in the city of Rotterdam to assess the effects of traffic emissions on long-term health effects in the period 1985–2008. The required inputs such as $PM_{10}$ background concentrations and emission factors could be derived from extrapolation of 2008 values back to 1985. Similar data for EC could be derived from Black Smoke time series dating back to the 1980s. A conversion factor of a factor ten was established between Black Smoke measurements and EC concentrations. The reliability of this conversion is hampered by the lack of an agreed analytical protocol for thermal analysis of EC in Europe. The latter also makes it difficult to compare modeled and measured EC concentrations in ambient air as automatic instruments (e.g. multi angle absorption photometer) are compared with thermal analysis of EC.

Our study shows that in Rotterdam in the period 1985–2008, the air quality of $PM_{10}$ and EC improved significantly both at urban background and near heavy traffic locations. For $PM_{10}$, the average urban background decreased from 43 μg $m^{-3}$ to 25 μg $m^{-3}$ in this period. This is attributed for 70% by large-scale emission reduction by industry, energy production and road traffic of *precursors* (sulfur dioxide, ammonia and nitrogen oxides) of secondary particles. Only 10% reduction of $PM_{10}$ is related to primary combustion aerosol. For EC, the average urban background decreased from 3 μg $m^{-3}$ to 1 μg $m^{-3}$ in this period. The main reason is lower regional background concentrations as a result of reduced emissions of soot particles by combustion processes in general (e.g. industry, energy production, shipping and road traffic) and at the urban scale of (diesel) emissions by road traffic in particular.

The improved air quality resulted in a gain of life on average of 13 months ($PM_{10}$) or 12 months (EC) per person in Rotterdam. The range in uncertainty is 6 months which is mainly related to the uncertainty in the estimate of the relative risk of long-term health effects per mass unit concentration. $PM_{10}$ and EC have similar health impacts despite the ten times larger decrease in $PM_{10}$ concentrations as compared to EC. This is explained by the 10 times higher excess risk per μg $m^{-3}$ for EC as compared to $PM_{10}$. This demonstrates that EC is a more sensitive indicator (compared to $PM_{10}$) to monitor the health effects of traffic measures. It is noted, that EC is likely *not* causing the health effects but acts as a proxy for the mass of combustion aerosol.

Further experimental research is recommended to improve modeling of EC in urban areas (i.e. establish emission factors of EC for free-flowing and congested road traffic) and to validate effects of traffic measures on air quality of EC (i.e. low emission zones and speed limitation).

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