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Effects of European emission reductions on air quality in the Netherlands and the associated health effects

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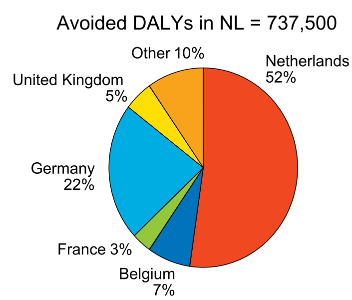
HIGHLIGHTS

- Air quality in the Netherlands has improved since 1980 due to European policies.
- A World Avoided scenario is defined with no air quality policies from 1980 to 2015.
- In 2015, avoided $PM_{2.5}$ comes from more than half from foreign emission reductions.
- Industry is the main contributing sector, followed by agriculture and transport.
- A 6 year increase in life expectancy is attributed to the avoided air pollution.

GRAPHICAL ABSTRACT



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ABSTRACT

Policies implemented in Europe since the 1970s to improve the air quality have resulted in decreases in emissions in many countries with corresponding reductions in concentrations of sulphur dioxide (SO_2), nitrogen dioxide (NO_2) and particulate matter (PM). We report here how much the air quality and associated health effects in the Netherlands have improved since 1980 and which countries, sectors and policies are responsible for this. To quantify the effects of emission reduction policies since 1980 we calculated the ambient concentrations of air pollutants in the Netherlands from 1980 to 2015, using two scenarios. A Baseline scenario with reported emissions in Europe and a World Avoided scenario which assumed that no air quality policies were adopted from 1980 onwards which would result in the growth in emissions of air pollutants. In the World Avoided scenario, the annual average $PM_{2.5}$ concentration in the Netherlands increases from 59 $\mu g\, m^{-3}$ in 1980 to 102 $\mu g\, m^{-3}$ in 2015, while in reality (Baseline scenario) concentrations decreased to about 12 $\mu g\, m^{-3}$. The avoided $PM_{2.5}$ concentration in 2015 accounts for more than half (56%) of reductions in emissions in sectors outside the Netherlands. Foreign (38%) and domestic (16%) industry is the main contributing sector, followed by agriculture (23%) and transport (15%). In 2015, the avoided concentrations of air pollutants correspond to about 700,000 avoided years of life lost in the Netherlands per year, with an associated number of avoided attributable deaths of about 66,000 per year, and an increase in average life expectancy of about 6 years. The corresponding avoided monetary health damage amounts to between € 35 and € 77 billion per year in 2015.

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1. Introduction

The adverse effects of air pollution on human health have been known since the beginning of the 20th century with, e.g., the Meuse valley fog in Belgium in 1930, the Donora smog in Pennsylvania in 1948, and the great smog of London in 1952 (Jacobs et al., 2018). In 1979 the relationship between soil acidification and forest damage was established in Germany (Ulrich et al., 1979). Acid rain was found to be caused by the emissions of sulphur and nitrogen oxides from the burning of fossil fuels throughout Europe, as well as by ammonia from agriculture. To combat acid deposition the Convention on Long-Range Transboundary Air Pollution (LRTAP) was drawn up under the United Nations Economic Commission for Europe (UNECE) in 1979. Several protocols were agreed under the convention to reduce the emissions of sulphur in 1985, nitrogen oxides (NO_x) in 1988, and volatile organic compounds (VOCs) in 1991. These emission reduction targets were combined and further strengthened in the Gothenburg protocol on acidification, eutrophication and ground level ozone of 1999 (UNECE, 1999), which was revised in May 2012.

Concerns over the effects of air pollution on human health resulted in the first policy measures adopted by the European Community in 1970 to reduce the emissions of carbon monoxide (CO) and hydrocarbons (HC) from motor vehicles (EC, 1970). This was followed by legislation which limited the emissions of sulphur dioxide (SO_2) from large combustion plants in 1988, the emissions of NO_x and particulate matter (PM) from motor vehicles from 1991onwards, and the emissions of SO_2 from liquid fuels in 1993. In 2001 national emission ceilings were agreed in Europe for SO_2, NO_x, ammonia (NH_3) and VOCs for 2010 onwards (EU, 2001), which were revised in December 2016 and extended with emission reductions for later years.

In tandem with policy measures to reduce emissions, limit values for the concentrations of air pollutants were agreed in Europe starting in 1980 for concentrations of SO_2 and suspended particles. The limit values were gradually lowered and extended and now include many compounds relevant for public health (EU, 2008).

The policies under the LRTAP convention and from the European Union (EU) have resulted in significant reductions in emissions of air pollutants in European countries in the last few decades (EEA, 2017). As a direct result, concentrations of air pollutants have also been decreasing in most countries in Europe since the 1980s or 1990s (Guerreiro et al., 2014; Maas and Grennfelt, 2016). However, air pollutants are transported over large distances and across country borders. Concentrations of air pollutants in a given country are therefore determined by the contributions from emissions originating from many different countries. This is especially true for smaller countries such as the Netherlands. Although the emissions of air pollutants have decreased and the EU limit values for concentrations are within reach in most places in Europe (EEA, 2018), the concentrations are still significantly above the air quality guidelines of the World Health Organization (WHO, 2005).

Several studies have reported on the benefits of emission reduction measures on the air quality and the health impacts in Europe (Carnell et al., 2019; Crippa et al., 2016; Maas and Grennfelt, 2016; Turnock et al., 2016). Maas and Grennfelt (2016) concluded, based on earlier work from Rafaj et al. (2013), that if air pollution trends had followed the same trend as economic output (i.e. no decoupling), the average $PM_{2.5}$ levels in Europe would have been similar to current levels at European hotspots, such as major cities in Eastern Europe and in the Po valley. Air quality related health impacts in Europe would be three times more than today and the average life expectancy would be 12 months less than today (Maas and Grennfelt, 2016). Turnock et al. (2016) estimated that emission reductions in Europe decreased European annual mean concentrations of $PM_{2.5}$ by 35% which has prevented 37,000–116,000 premature deaths annually across the EU, resulting in a perceived financial benefit to society of US$ 232 billion annually. Crippa et al. (2016) illustrated the substantial impacts on human health of cleaner EU technologies and emission standards both inside and outside Europe. Carnell et al. (2019) studied the decrease in air pollution in the UK and concluded that the attributable mortality in the UK due to exposure to $PM_{2.5}$ and NO_2 have declined by 56% and 44%, respectively, since 1970, primarily driven by policy interventions. A study from the WHO (2015) also demonstrated the large health effects and associated monetary damage attributable to air pollution for Europe. Other studies have focused on the effects of policy interventions on the SO_2 concentrations in the Netherlands (Velders et al., 2011b) and Sweden (Åström et al., 2017).

In this study we investigated how much the air quality and associated health effects have improved in the Netherlands since 1980 and how much of this can be attributed to emissions reductions in the Netherlands itself and how much to emissions reductions in other countries. We also investigated which sectors, domestic and foreign, and which policy measures contributed most to the improvements in air quality. To quantify this we performed model calculations with high spatial resolution for concentrations of NO_2, PM_{10}, $PM_{2.5}$, and elemental carbon (EC) in the Netherlands from 1980 to 2015 using two scenarios. One scenario, called the Baseline, follows the reported emissions of all relevant air pollutants in all European countries. A second scenario, called World Avoided, is defined as a scenario in which no emission reduction measures were taken and assumes that emissions would have continued to grow from 1980 onwards based on growth in economic activities and demographic changes. Changes in emissions which result from other effects, such as changes in the economic structure or improvements in energy efficiency are taken into account through the activity data that drives the emissions in the World Avoided scenario.

We quantify the benefits of the emission reductions in terms of reductions in the number of air pollutant limit value exceedances in the Netherlands and in the reductions in exposure of the Dutch population to high concentrations of air pollutants and reductions in the related health effects. These reductions in concentrations, number of exceedances, exposure and health effects are discussed in relation to the policy measures that have been taken over the years in Europe.

2. Methods

The concentration levels of NO_2, PM_{10}, $PM_{2.5}$ and EC at road-side locations result from the combination of large-scale concentrations and local traffic contributions. A detailed description of the methods used to calculate the large-scale concentration is given in Velders and Diederen (2009) and Velders et al. (2017) and of the local traffic contributions in Rutledge-Jonker et al. (2017). The methods used here are the same as those used by the Dutch government in the National Air Quality Cooperation Programme (https://www.nsl-monitoring.nl/) to monitor whether the Netherlands complies with the European air quality limit values (EU, 2008).

2.1. Large-scale concentrations

Large-scale concentrations of NO_2, PM_{10}, $PM_{2.5}$ and EC contain contributions from sources in the Netherlands and other European countries. These concentrations are representative of averaged spatial rural and urban background concentrations with a resolution of about 1×1 km^2. Maps of these spatial distributions in the Netherlands were calculated using the Operational Priority Substances (OPS) dispersion model (Sauter et al., 2018; Van Jaarsveld, 2004; Van Jaarsveld and De Leeuw, 1993) which calculates the annual average concentrations based on emissions and their dispersion, transport, chemical conversion and deposition. The model uses a Gaussian plume for dispersion on a local scale and a Lagrangian trajectory for the long-distance transport of compounds. The contributions of the emissions from each sector to the total concentrations can therefore be calculated separately. Nonlinear chemical effects are taken into account indirectly by using pre-determined background concentrations of all compounds. In the OPS model, substances whose behaviour can be described by first-order chemical reactions are modelled. For this study, the model was used to calculate the annual average concentrations of NO_x, SO_2, NH_3, primary PM_{10} and $PM_{2.5}$, EC, and ammonium, nitrate and sulphate aerosols.

As NO_2 is chemically active in the atmosphere, it cannot be modelled directly by the OPS model. Instead, NO_2 is calculated from the modelled NO_x concentration and an empirical relationship between annual average measured NO_x and NO_2 concentrations (Van de Kassteele and Velders, 2006).

Total PM_{10} and $PM_{2.5}$ concentrations are the sum of the contributions from primary particulate matter emissions, secondary aerosols, and sea salt, which are each calculated separately with the OPS model. The formation of the secondary aerosols is a nonlinear process which depends on the concentrations of several pollutants and is therefore different for, e.g. 1980 than 2015. This is especially relevant for the ammonium aerosol. The effect of the nonlinearity is taken into account in background concentrations used in the scenario calculations. The secondary aerosols, nitrate, ammonium, and sulphate are calibrated by comparison with observations for the years 2011–2015.

EC concentrations are calculated using the emission ratio between EC and $PM_{2.5}$ for each separate emission category (PRTR, 2018). These ratios could have changed over time as a result of, e.g., changes in car engine technology, but are assumed to be constant over time in the calculations, due to the lack of historical information on these ratios.

All calculations were performed with long-term average meteorology to only quantify the effects of changes in emissions on the concentrations.

The modelled large-scale concentration maps are made annually and compared with observations from Air Quality Monitoring Networks in the Netherlands (DCMR, 2018; GGD, 2018; LML, 2018) at 35–45 rural and urban background locations in the Netherlands (depending on the pollutant) and found in general to be in good agreement for NO_x (and NO_2). The situation is somewhat different for PM_{10} and $PM_{2.5}$. The calculated concentrations, taking into account the quantified direct anthropogenic emissions, the secondary inorganic aerosols, and the contribution from sea salt, account for about 70% of the observed PM_{10} concentrations and about 90% of the observed $PM_{2.5}$ concentrations (Velders et al., 2017). The remainder comes from windblown dust, hemispheric transport, secondary organic compounds, and metal oxides (Buijsman et al., 2005). The PM_{10} and $PM_{2.5}$ concentrations calculated with the OPS model have therefore been compared with concentrations measured at regional and city background locations. PM_{10} measurements are available from 1992 to 2017 and $PM_{2.5}$ measurements from 2009 to 2017 (DCMR, 2018; GGD, 2018; LML, 2018; Velders et al., 2017). The measured concentrations were averaged over five year periods to minimise meteorological influences and enable comparison with the calculated concentrations. This resulted in a calculated difference between observed and modelled concentrations of about 7 $\mu g\,m^{-3}$ for PM_{10} and 1.6 $\mu g\,m^{-3}$ for $PM_{2.5}$. These differences were added to the calculated concentrations as a bias correction, to account for the contributions that are not explicitly calculated with the OPS model.

2.2. Local traffic contributions

In addition to the large-scale concentrations, local contributions from road traffic are calculated by the TREDM software implementation of the Dutch Standard Models for Air Quality (Wesseling et al., 2011), consisting of the CAR model for street canyons and a line-source model for open field situations. With this combination of models the contributions from road traffic to the NO_2, PM_{10}, $PM_{2.5}$, and EC concentrations are calculated, with a resolution of $10 \times 10\,m^2$, and added to the large-scale concentrations. The model calculates the contributions from road traffic in city streets and from rural and motorway traffic. The TREDM model uses data on all roads and streets with a certain volume in the Netherlands for which concentrations are relevant in air quality assessments, amounting to a total length of about 22,000 km, of which about 3000 km are motorways. Concentrations, relevant for exposure, are estimated at points 1.5 m above road level and normally some 2 m from the centre of roads in cities and at about 10 m from the centre of motorways, depending on the situation. A small fraction of the NO_x emission is emitted directly as NO_2. The majority consists of NO and is partially photochemically converted into NO_2 depending on the available ozone background concentration and photochemical parameters. In the calculations the total NO_2 traffic contribution is the sum of the direct and indirect (converted) contributions. At distances of a few kilometres from the road, the fraction of directly emitted NO_2 hardly affects the NO_2 concentrations, as a chemical equilibrium is established within minutes (Jacob, 1999).

The TREDM model suite consists of a street canyon sub model, an updated version of the CAR model (Calculation of Air pollution from Road traffic (Eerens et al., 1993; Van Velze and Wesseling, 2014; Wesseling and Sauter, 2007)) for calculating the contribution of road traffic in city streets, and a line-source model that calculates the contributions from roads outside cities, mostly along motorways.

The CAR model is a generic model for determining air quality near roads in urban areas. The traffic contribution is calculated by multiplying the traffic emissions by a dispersion factor. The traffic emissions depend on the traffic proportions (composition, intensity and driving conditions), and the dispersion factor on street characteristics (buildings, trees and distance from the centre of the road).

The line-source model (Wesseling and Van Velze, 2014) is used for estimating the contributions of road traffic emissions to concentrations in open field situations, such as along motorways. It is based on an earlier model of TNO (Wesseling and Zandveld, 2006) and calculates dispersion according to a Gaussian plume model. Corrections are applied to local concentrations for emissions that are included in the calculation of the large-scale concentrations. The total concentrations have been validated extensively (Wesseling et al., 2013, 2016; Wesseling and Sauter, 2007; Wesseling and Zandveld, 2006).

Similar to the OPS model, the TREDM output consists of NO_x and directly emitted NO_2 concentrations, but on a local scale. It uses an empirical relationship to estimate NO_2 concentrations from O_3 and NO_x concentration contributions.

According to the last validation exercise (Wesseling et al., 2016), the modelled total concentrations (large-scale + traffic contribution) overestimate the measured NO_2, PM_{10} and $PM_{2.5}$ concentration on average by roughly 1 $\mu g\,m^{-3}$, with a standard deviation (1 sigma) of almost 5 $\mu g\,m^{-3}$ for NO_2 and 2 $\mu g\,m^{-3}$ for PM_{10} and $PM_{2.5}$.

2.3. Emissions baseline scenario

The Baseline scenario is based on the reported emissions in the EU and as such takes into account Dutch and EU air quality policy measures that have been implemented over the years.

The historical anthropogenic emissions in the Netherlands for 1990 to 2015 used in the OPS calculation are the official national emissions collected by the Pollutant Release & Transfer Register (PRTR, 2018). They are also used for reporting emissions to, for example, the European Commission, the Convention on Long-Range Transboundary Air Pollution, and the United Nations Framework Convention on Climate Change. For the year 1980, for which no emissions were available in the PRTR (2018), data from the Centre on Emission Inventories and Projections (CEIP, 2018) was used, see Table 1.

The emissions are subdivided into some 120 source categories, including nine categories for industry, seven for agriculture, and 30 for road transport (including exhaust emissions and emissions from tyre, brake and road wear) (Velders et al., 2017). The emissions in each of the categories are spatially distributed according to various statistical and geographical datasets. They are available as point sources or area sources at a resolution of $1 \times 1\,km^2$. For each source category the spatial distributions in emissions corresponding to the year 2015 have been used for all years and for both the Baseline and World avoided scenario, because historical spatial distributions of emissions are not available.

Table 1
Sources of the anthropogenic emissions for the Netherlands.

	1980	1990–2015
NO_x	CEIP (2018) emissions (SNAP format) on top of 1990 sectoral split	PRTR (2018)
SO_2	CEIP (2018) emissions (SNAP format) on top of 1990 sectoral split	PRTR (2018)
NH_3	Emission as 1990 used. Data from CEIP (2018) for 1980 seem unreliable: SNAP10 (agriculture) emissions are much lower than those of 1990, which is not seen for other countries.	PRTR (2018)
PM_{10}, $PM_{2.5}$	Emission of 1990 used. Data from CEIP (2018) are not available for the Netherlands for 1980.	PRTR (2018)

For most source categories this is a valid approach since the land use in the Netherlands has not changed significantly in the past decades. In a few places new sections of motorways have been built though since 1980, resulting in new locations with emissions. The fact that the spatial distributions for each sector are held constant for all years will not affect the result very much, because the focus here in on the year 2015 and for the Netherlands as a whole and not for specific regions.

For the historical emissions of other European countries for 1980 to 2015, the emissions ('as used in models') of CEIP (2018) are used, see Table 2.

For the calculation of the local traffic contributions to the total concentrations (Section 2.2) specific emissions factors (in g km^{-1}) are used for various typical vehicle fleet-traffic situation combinations, see Appendix A (Ligterink, 2018; Spreen et al., 2016b). For the Baseline scenario the emission factors are based on tailpipe measurements in the laboratory, from 1988 onwards, and on the road, from 2009 onwards, of various types of cars and light and heavy duty trucks, in combination with information on the composition of the car and truck fleet in specific years. As such, emission factors decrease significantly from 1980 to 2015 in the Baseline scenario.

2.4. Emissions World Avoided scenario

The World Avoided (WA) scenario describes a situation without explicit policies to reduce the emissions of air pollutants. In this scenario emissions continue to grow from 1980 to 2015 based on growth in economic activities and demographics. The emissions of NO_x, SO_2, NH_3, and primary PM_{10} and $PM_{2.5}$ (and EC) for the Netherlands and other European countries change from 1980 to 2015 proportional to changes in underlying activity data. Different activity data is used for the various sectors as shown in Table 3. The trend in activity data for the Netherlands and neighbouring countries is shown in Fig. 1 and Fig. 2. Most activity data is from the OECD (2018). For the Netherlands more detailed activity data is used for road transport and inland shipping and fishing from PRTR (2018). This is especially relevant for the emissions of road transport in the Netherlands, since these yield relatively high spatial gradients and occur close to places where people live. The OECD (2018) data only consist of the total number of passenger kilometres driven on all roads, while the PRTR (2018) has detailed data on the number of vehicle kilometres driven for passenger cars, light duty trucks, heavy duty trucks, buses, and motorcycles, individually, for urban roads, motorways, and other (rural) roads.

Fig. 1 shows that road transport on motorways in the Netherlands has increased strongly since 1980 for both passenger cars and heavy duty transport. On urban roads passenger car transport has remained

Table 2
Source of the anthropogenic emissions for European countries, apart from the Netherlands.

	1980–2015
NO_x	CEIP (2018) emissions in SNAP format[a]
SO_2	CEIP (2018) emissions in SNAP format[a]
NH_3	CEIP (2018) emissions in SNAP format[a]
PM_{10}, $PM_{2.5}$	CEIP (2018) emissions in SNAP format;[a] NRF09/NRF14 format for 1990.[b] 1990 emissions are also used for 1980.[c]

[a] Selected Nomenclature for sources of Air Pollution (SNAP), see Table 3.
[b] The data in NRF format cannot always consistently be converted to SNAP format. Scaling factors for 1990 vs 2000 emissions are therefore calculated and applied to the SNAP data for the year 2000. If NFR data for 1990 was not available for a country an average scaling factor (averaged over all countries) was applied.
[c] CEIP (2018) has very limited data for 1980, which is therefore not used here.

Table 3
Setup of the World Avoided scenario assuming no air quality policies. The emissions of NO_x, SO_2, NH_3, and primary PM_{10} and $PM_{2.5}$ in the Netherlands and other European countries change from 1980 to 2015 and are assumed to be proportional to the changes in underlying activity data.

	SNAP codes[a]	Activity data used to drive the changes in emissions
Electricity production	1, 3	Electricity generation from OECD[b]
Households	2	Population from OECD[c]
Industry	4, 5, 6, 9	Industrial production index from OECD[d]
Passenger cars	7	Passenger transport on roads (in passenger km) from OECD[e,f,g]
Heavy duty transport	7	Freight transport on roads (in tonne km) from OECD[h,f,g]
Non-road transport	8	Gross domestic product from OECD[i,f]
Agriculture	10	Livestock units calculated from NEC/IIR data[j]
Sea shipping		NO_x emissions in the baseline scenario used as scaling factors for all species[k,f]

[a] SNAP codes: 1) Combustion in energy and transformation industry, 2) Non industrial combustion, 3) Combustion in manufacturing industry, 4) Production processes, 5) Extraction and distribution of fossil fuels, 6) Solvents, 7) Road transport, 8) Other mobile sources, 9) Waste, 10) Agriculture.
[b] Electricity generation (gigawatts hour), all types excluding nuclear energy; OECD (2018), Electricity generation (indicator). https://doi.org/10.1787/c6e6caa2-en (Accessed on 23 November 2017).
[c] Population; OECD (2018), Population (indicator). https://doi.org/10.1787/d434f82b-en (Accessed on 22 March 2017).
[d] Industrial production index; OECD (2018), Industrial production (indicator). https://doi.org/10.1787/39121c55-en (Accessed on 22 November 2017).
[e] Passenger transport on roads (million passenger km); OECD (2018), Passenger transport (indicator). https://doi.org/10.1787/463da4d1-en (Accessed on 22 March 2017).
[f] For the Netherlands more detailed data from (PRTR, 2018) is used for road transport (passenger km; for light and heavy duty transport on different types of roads), inland shipping (fuel used) and fishing (fuel used) which is consistent with the NEC reported data.
[g] Road transport for countries other than the Netherlands is described by one category (SNAP7). To take into account the different trends in activities for passenger cars and heavy duty transport, the SNAP7 emissions are split according to the ratio between the emissions of passenger cars and light/heavy duty vehicles in the Netherlands in 1990: NO_x: 60% passenger cars, 40% light/heavy duty vehicles, PM_{10} and $PM_{2.5}$: 50-50%, SO_2 40–60%, and NH_3 100-0%.
[h] Freight transport on roads (million tonne km); OECD (2018), Freight transport (indicator). https://doi.org/10.1787/708eda32-en (Accessed on 22 March 2017).
[i] GDP; OECD (2018), Gross domestic product (GDP) (indicator). https://doi.org/10.1787/dc2f7aec-en (Accessed on 22 March 2017).
[j] Livestock units (number of animals) calculated from the National Emissions Inventories under the NEC directive (http://cdr.eionet.europa.eu) and the livestock units coefficients for various species and age on the basis of the nutritional or feed requirement of each type of animal.
[k] For sea shipping the number of ships or amount of cargo are not a good measure for the activity data in the World Avoided scenario because of the increases in ship sizes. Since there have been no international emission controls on the NO_x emissions of sea ships, the NO_x emissions in the baseline scenario (Velders et al., 2017; VITO, 2013) are used to scale the PM_{10}, $PM_{2.5}$, SO_2 and NH_3 emissions of sea shipping.

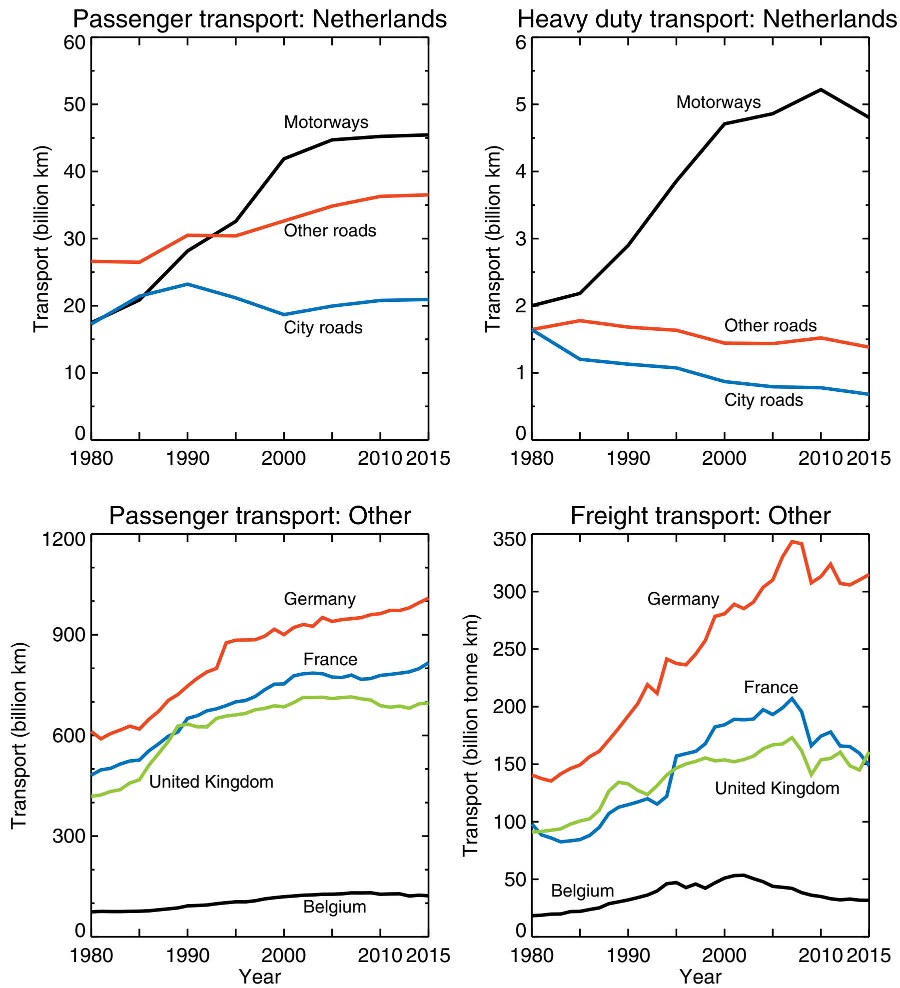

Fig. 1. Activity data used as drivers for the emissions of light and heavy duty road transport used for the World Avoided scenario. The road transport volumes for the Netherlands (in billion vehicle km) are based on detailed information from the Pollutant Release and Transfer Register (PRTR, 2018). For other European countries the data is from passenger transport on roads (in passenger km) and freight transport on roads (in billion tonne km) as reported by OECD (2018), see Table 3.

more or less constant since 1980, while heavy duty transport has reduced in intensity. For other countries the total road transport volumes have increased since 1980 for both passenger cars and heavy duty transport (expressed here as freight transport on roads). The trends in other activity data for the Netherlands and neighbouring countries are shown in Fig. 2.

In a few instances the emissions in the World Avoided scenario for a specific sector, compound, and year, were lower than in the Baseline scenario. When this occurred the emissions of the World Avoided scenario were replaced with those of the Baseline scenario to account for the potentially higher emissions.

In the World Avoided scenario the emissions change in proportion to the change in activity data. In reality, there will not be a direct linear relationship. For example, the emissions of heavy duty transport will depend on the number of vehicles used to transport goods and on the mix of vehicles and their capacities. Such effects are not taken into account in the World Avoided scenario as discussed here.

In the World Avoided scenario some policy measures are taken into account implicitly through the activity data that is used. Energy savings are included by using energy use data as basis for estimating (unabated) emissions. Likewise reductions in the size of the livestock as a result of manure policies are also implicitly taken into account, as the number of animals is the basis for estimating (unabated) ammonia emissions.

In the World avoided scenario it is assumed that there are no Euro standards for passenger cars and light and heavy duty trucks. The emissions factors used in the calculation of the local traffic contributions to the total concentrations (Section 2.2) for the year 1980 are therefore also used in the calculations for the 1990–2015 period. As such, changes in traffic volumes (Fig. 1) are taken into account.

2.5. Policies related to air quality

Limit values for ambient concentrations of air pollutants have been in place in Europe since 1980, starting with SO_2 and particulates (Table 4). The 2008 EU air quality directive (EU, 2008) now has limit values for several pollutants, including NO_2, PM_{10}, and $PM_{2.5}$ which are the most relevant ones for health effects.

To improve the air quality in Europe by emission reduction, controls for air pollutants have been agreed at various levels, such as through the International Maritime Organization (IMO), the LRTAP convention, the EU, and nationally. An overview of the relevant policy measures is given in Table 5. The controls are related to emissions reductions for all relevant pollutants and set specific standards for industry, power plants, motor vehicles and shipping as well as emission ceilings for countries. For local air quality concentrations the EU directives (Euro standards) for motor vehicles (passenger cars and light and heavy duty vehicles) are especially relevant.

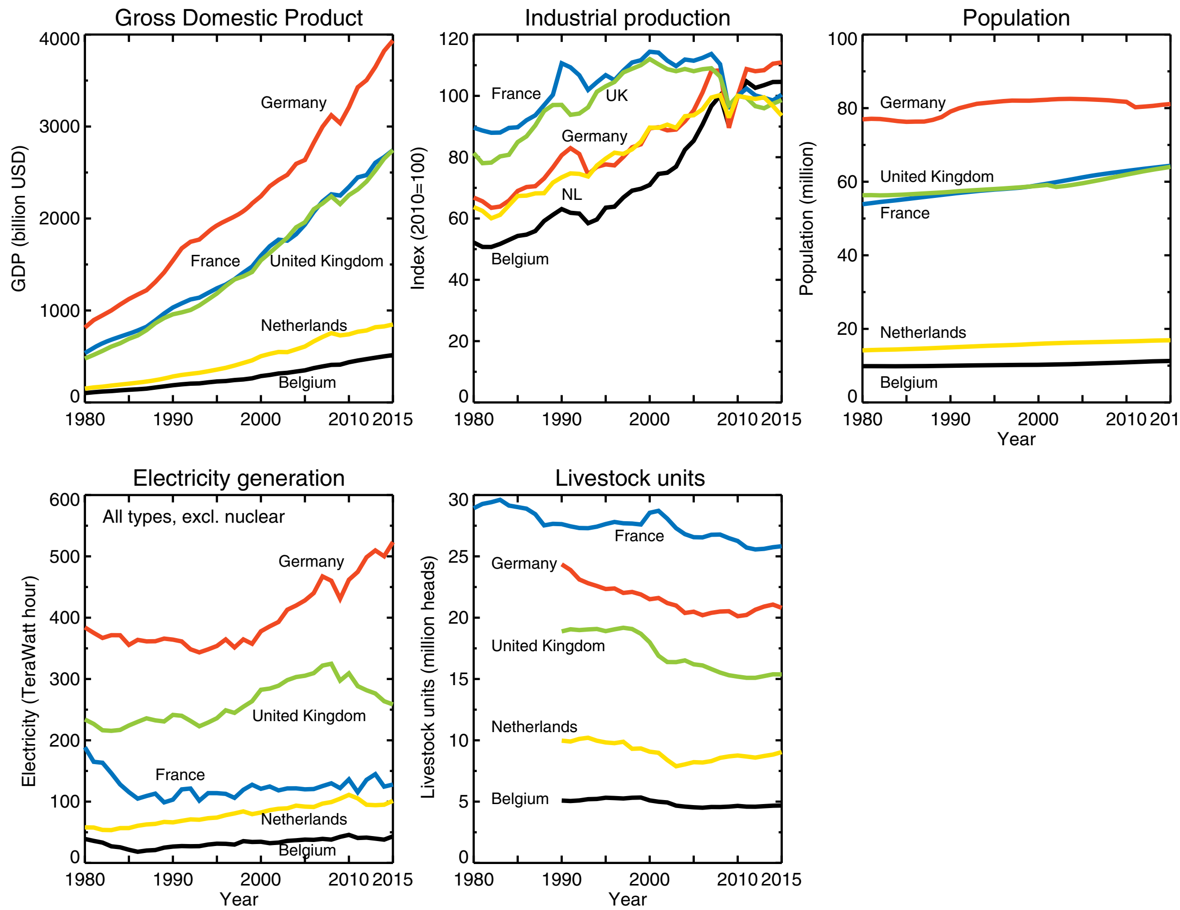

Fig. 2. Activity data used as drivers in the World Avoided scenario to derive the emissions of the various sectors, i.e., industry, electricity production, households, non-road transport, and agriculture. See Table 3 for the details of how the activity data is used.

Table 4
European Union policies related to air quality limit values.

Year	Compounds	Policy
1980	SO_2, suspended particles	Directive on air quality limit values and guide values for sulphur dioxide and suspended particulates (80/779/EEC)
1985	NO_2	Directive on air quality standards for nitrogen dioxide (85/203/EEC)
1996	SO_2, NO_2, PM, TSP, lead, ozone, etc.	Directive on ambient air quality assessment and management (96/62/EC). Framework directive
1999–2002	SO_2, NO_x, NO_2, PM_{10}, etc.	Several directives relating to limit values for sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead in ambient air, directives (1999/30/EG) (2000/69/EG) (2002/3/EG)
2008	SO_2, NO_x, NO_2, PM_{10}, $PM_{2.5}$, etc.	Directive on ambient air quality and cleaner air for Europe (2008/50/EC)

2.6. Source attribution particulate matter

The OPS model calculates the contributions of the emissions from each of the sectors to the concentrations separately. The attribution of concentrations to sectors can be estimated in various ways and depends on its purpose. This is especially true for the attribution of the secondary inorganic aerosols (SIAs), as part of the PM concentration, to sectors. This attribution can be done on the basis of the mass of the different aerosols, which is a valid attribution when considering the contributions of the sectors to the total mass of the aerosol and the total particulate matter concentration. However, it does not give a good estimate of the effect of emission reducing measures in a sector on the particulate matter concentration. This is because more than one ion is needed to form ammonium nitrate or ammonium sulphate and the molecular weights of the ions are rather different, with ammonium about 3–5 times lighter than sulphate and nitrate. For example, there is much less mass (about 70% less) ammonium than nitrate needed to yield a given number of ammonium nitrate particles. This can be taken into account using a source attribution based on the number of aerosol molecules (moles). The sector contributions for the SIAs are then calculated by adding the number of moles of aerosol per sector, instead of the mass of the aerosol.

The SIA concentration on a mass basis, C_{SIA}, of sector i is defined as,

$$C_{SIA}(i) = C_{NO3}(i) + C_{NH4}(i) + C_{SO4}(i).$$

The number of moles of SIAs, Mol_{SIA}, of sector i is defined as,

$$Mol_{SIA}(i) = \frac{C_{NO3}(i)}{M_{NO3}(i)} + \frac{C_{NH4}(i)}{M_{NH4}(i)} + \frac{C_{SO4}(i)}{M_{SO4}(i)},$$

in which, M is the molecular weight of the aerosol. The relative number of moles of SIAs per sector can then be used as scaling factors to estimate the SIA concentration on a mole basis, C^M_{SIA}, per sector by,

$$C^M_{SIA}(i) = \frac{Mol_{SIA}(i)}{\sum_j Mol_{SIA}(j)} \cdot \sum_j C_{SIA}(j).$$

The primary PM contributions can then be added to the SIA contributions to obtain the total source attribution for PM_{10} and $PM_{2.5}$ concentrations. The fact that two ammonium ions are needed to form an ammonium sulphate molecule is also taken into account.

Table 5
Overview of policy measures directly related to anthropogenic emissions of air pollutants.

Year	Compounds	Policy
Convention on Long-Range Transboundary Air Pollution (UNECE/CLRTAP)		
1988	NO_x	Sofia protocol on nitrogen oxide emissions (UNECE, 1988)
1999, 2012	SO_2, NO_x, $PM_{2.5}$, VOC, NH_3	Gothenburg Protocol to abate acidification, eutrophication and ground-level ozone with emissions ceilings for European countries for 2010-on and 2020-on (UNECE, 1999)
International Maritime Organization (IMO)		
2008	SO_x, NO_x	Emission controls for sea shipping limiting the sulphur contents of fuels and setting standards for NO_x emissions, including stricter controls for the North sea (IMO, 2008)
European Union (EU; between brackets the directive numbers)		
1970	CO, hydrocarbons	Directive on measures to be taken against air pollution by gases from positive-ignition engines of motor vehicles (70/220/EEC)
1988	SO_2	Directives on the limitation of emissions of certain pollutants into the air from large combustion plants (88/609/EC, 94/66/EC, 2001/80/EC)
1991-on	NO_x, PM, CO, hydrocarbons	Directives on measures to be taken against air pollution by emissions of motor vehicles[a]
1993	SO_2	Directives relating to a reduction in the sulphur content of certain liquid fuels (93/12/EEC, 1999/32/EC, 2012/33/EU)
2001	SO_2, NO_x, $PM_{2.5}$, NH_3, VOC	Directive on National Emission Ceilings (NEC; 2001/81/EC) with emission ceilings for the year 2010-on (EU, 2001)
2009	Various	Directive on settings for eco-design for energy-related products (2009/125/EC)
2010	SO_2, NO_x, PM, CO, VOC	Directive on industrial emissions setting emission limits from 2014-on (2010/75/EU)
2015	SO2, NO_x, PM	Directive on medium combustion plants setting emissions limits from 2025-on (2015/2193/EU)
2016	SO_2, NO_x, $PM_{2.5}$, NH_3, VOC	Directive on the reduction of national emissions of certain atmospheric pollutants (revised NEC directive; 2016/2284/EU) with emission reductions for the year 2020–2029 and 2030-on relative to the year 2005
Netherlands		
2005	PM	Particulate filters encouraged in NL for passenger cars starting in 2005
1990s-on	PM, NH_3	Various national measures to reduce emissions in agriculture
2009	NO_x, PM	National air quality collaboration program (NSL), a Dutch programme for complying with the EU Air quality directive.
2015	NO_x, NH_3	Programme approach towards nitrogen (PAS), a Dutch programme to reduce nitrogen deposition.

[a] Euro standards.

- Euro 1 applies from 1993-on for passenger cars and light duty commercial vehicles (91/441/EEC, 93/59/EEC)
- Euro 2 applies from 1996-on for passenger cars and motorcycles (94/12/EC, 96/69/EC, 2002/51/EC, 2006/120/EC)
- Euro 3 applies from 2000-on for any vehicle (98/69/EC, 2002/51/EC, 2006/120/EC)
- Euro 4 applies from 2005-on for any vehicle (98/69/EC, 2002/80/EC)
- Euro 5 applies from 2009-on for light duty and commercial vehicles (715/2007/EC)
- Euro 6 applies from 2014-on for light duty and commercial vehicles (459/2012/EC)

The main effect of the source attribution calculated on a mole basis is that the relative contribution of emissions from agriculture, the main source of ammonia, to the PM concentrations is larger (approximately twice as large) than when calculated on a mass basis.

2.7. Exposure and health effects

For calculating health impacts from air pollution, local contributions (Section 2.2) are added to the large-scale contributions (Section 2.1) and total concentrations are calculated for all 8.8 million address locations of buildings in the Netherlands (BAG, 2018) to obtain the exposure (population weighted concentrations) to NO_2 and $PM_{2.5}$. Health impacts are assessed using the average population weighted concentrations for each year and the relative risk factors obtained from the Health Risks of Air Pollution in Europe (HRAPIE) project (WHO, 2013) in combination with the results of the Dutch Environmental Longitudinal Study (DUELS) (Fischer et al., 2015). In order to be able to easily assess the impact of changed in emissions and air pollution on health alone, the population, age structure and average mortality rates of 2015 are used for each year (Van den Brenk, 2018). Using a fixed population means that health impacts for the past are overestimated (by around 20% in 1980). But this effect is compensated for by the fact that mortality rates for people younger than 65 were higher in the past.

Different indicators are calculated to assess the health impact for the long-term exposure to NO_2 and $PM_{2.5}$:

1. The annual number of deaths attributable to air pollution.
2. The loss in average life expectancy (LLE).
3. The number of years of life lost (YLL).
4. The disability adjusted life years (DALYs), i.e., the sum of YLL and years lost due to disability (YLD).
5. The monetary health damage using a value per life year lost (YLL). Here a value of a life year is taken at € 50,000 to € 110,000 (2015 prices) (De Bruyn et al., 2017).

The number of deaths attributable to air pollution is calculated for the total Dutch population using the population weighted average exposure for each year. For each 5-years age class the attributable number of deaths are calculated using a relative risk of 1.06 associated with each 10 $\mu g\, m^{-3}$ exposure to $PM_{2.5}$. For NO_2 a relative risk factor of 1.02 is used. The loss in life expectancy is calculated by applying the survival rates with and without exposure to air pollution to each age class. The number of life years lost is the sum of the attributable deaths multiplied by the remaining life expectancy in each age class in the situation without air pollution (Van den Brenk, 2018).

The WHO-HRAPIE study (WHO, 2013) suggest using risk factors of 1.06 for $PM_{2.5}$ and of 1.055 for NO_2 in the health impact assessment. But as these risk factors are based on single pollutant studies there will be an overlap. The DUELS-study (Fischer et al., 2015) includes a two-pollutant analysis and finds a relative risk factor for the direct effects of NO_2 of 1.02. Here we calculate $PM_{2.5}$ impacts for concentrations above 2.5 $\mu g\, m^{-3}$ and NO_2 effects above 5 $\mu g\, m^{-3}$.

The HRAPIE study also includes relative risk factors for several morbidity impacts. The number of years lost because of disability is the weighted sum of these impacts. Weights reflect the severity of the diseases.

The largest health effects of air pollution are associated with exposure to $PM_{2.5}$ (about 85%), with a smaller contribution from expose to NO_2 (about 15%). Health effects that are directly associated with high ambient SO_2 concentrations, as calculated for the World Avoided scenario, are not taken into account here.

3. Results

3.1. Calculated and observed concentrations

The calculated large-scale concentrations were compared with measured concentrations (Fig. 3). The average concentrations for the Netherlands calculated for the Baseline scenario agree well with the average of observed concentrations at rural background locations. The agreement is good for the whole period from 1980 to 2015 for NO_x, NO_2 and SO_2, both in absolute terms and in the trend. Good agreement is also seen for PM_{10} and $PM_{2.5}$ concentrations, but this is the result of the calibration procedure to account for contributions from sources not calculated explicitly by the OPS model (see Section 2.1). The agreement for NH_3 is somewhat less; the absolute concentrations in 2010–2015 in

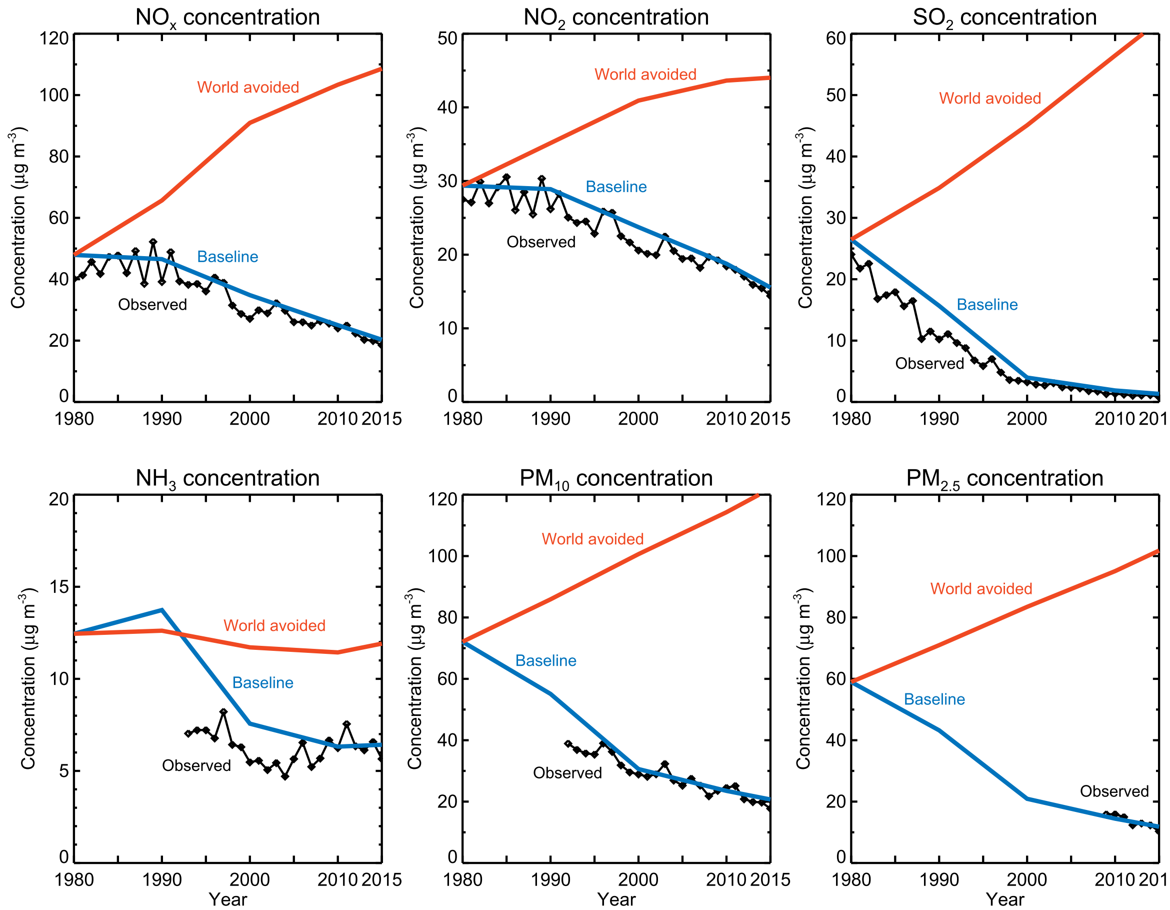

Fig. 3. Modelled and observed concentrations (in $\mu g\ m^{-3}$) of NO_x, NO_2, SO_2, NH_3, PM_{10}, and $PM_{2.5}$ averaged for the Netherlands. The observed concentrations are the averages of measurements at rural background locations (DCMR, 2018; GGD, 2018; LML, 2018). The modelled concentrations are from the Baseline and World Avoided scenarios. The modelled NO_2 concentrations are derived from the modelled NO_x concentrations using an empirical relationship based on observations in the Netherlands.

the Baseline scenario agree with the observations, but the calculated trend from 1993 to 2010 is larger than observed. Ammonia shows large spatial gradients in concentrations and there are only six to eight locations with long-term measurements in the Netherlands, which may explain the difference between modelled and measured concentrations (Wichink Kruit et al., 2018). Overall the modelled concentrations slightly overestimate the observations, but the focus is this study is not on absolute values, but on the difference between the World Avoided and Baseline scenario.

While the Baseline concentrations all show significant decreases from 1980 to 2015, the concentrations in the World Avoided scenario mostly increase. The average NO_2 concentration increases from about 30 $\mu g\ m^{-3}$ to 45 $\mu g\ m^{-3}$ which is significantly higher than the concentration in the Baseline scenario of about 15 $\mu g\ m^{-3}$. The concentrations of PM_{10} and $PM_{2.5}$ also show large increases in the World Avoided scenario, reaching about 120 $\mu g\ m^{-3}$ and about 100 $\mu g\ m^{-3}$, respectively, in 2015. This is much larger than the 20 $\mu g\ m^{-3}$ and 10 $\mu g\ m^{-3}$ for PM_{10} and $PM_{2.5}$, respectively, in the Baseline scenario.

The concentrations in the World Avoided scenario in 2015 are much higher that currently observed in the Netherlands and other European countries, but smaller than concentrations currently observed in other parts of the world. In large cities in China and India annual average PM_{10} concentrations over 200 $\mu g\ m^{-3}$ and annual average $PM_{2.5}$ concentrations over 120 $\mu g\ m^{-3}$ are observed in the period 2010–2018 (WHO, 2018). High concentrations are also observed in large cities in other countries in Asia and in the Middle East and Africa.

The EC concentration (not shown) in the World Avoided scenario increases from about 6 $\mu g\ m^{-3}$ to 15 $\mu g\ m^{-3}$, while it decreases to about 2.5 $\mu g\ m^{-3}$ in the Baseline scenario. EC is only measured routinely in the Netherlands at one location. A related quantity, black carbon, has only been routinely measured at 13 locations in the Netherlands since 2015 and is therefore not useful for comparison with the trend in calculated concentrations of EC.

3.2. Sector contributions to concentrations

The contributions of the different sectors to the large-scale concentrations of NO_2, $PM_{2.5}$ and EC are shown in Figs. 4–6 and of PM_{10} in Figure B1 (Appendix B). The concentrations are shown for the average of the Netherlands and for the average of the four largest cities in the Randstad (an urban conglomerate comprising of the cities Amsterdam, Rotterdam, The Hague, and Utrecht) and for the Baseline (1980, 2015) and the World Avoided (2015) scenarios. The relative contributions of the sectors are shown in Fig. 8.

3.2.1. NO_2 concentrations

The emissions of road transport are the dominant source of the NO_2 concentration in the Netherlands, with a contribution of about 57% in 1980 and about 40% in 2015 (Baseline scenario) (Fig. 8). Almost half of this is from domestic emissions of light duty transport (passenger cars and vans). In the World Avoided scenario, without policy measures to reduce the emissions of air pollutants, the emissions of road transport contribute about 52% to the NO_2 concentration in the Netherlands in 2015. Other sectors with major contributions are industry, non-road transport; minor contributors are agriculture, households, office buildings, services, etc. (labelled “Other”). The emissions of sea shipping are

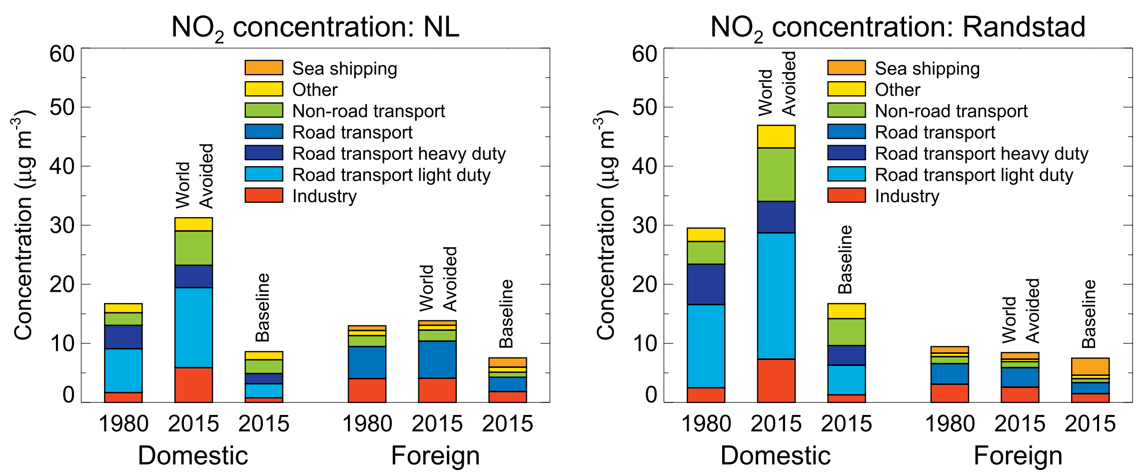

Fig. 4. Contribution of the different sectors to the average NO_2 concentration for the Netherlands (left panel) and for the average of the four largest cities in the Randstad (right panel). Shown are the contributions of emissions from the domestic sectors industry (including electricity production, refineries, and waste), light and heavy duty road transport, non-road transport, sea shipping and others (agriculture, households, etc.) and the contributions of emissions from sectors in other European countries (Foreign) for 1980 and 2015 for the World Avoided and Baseline scenarios.

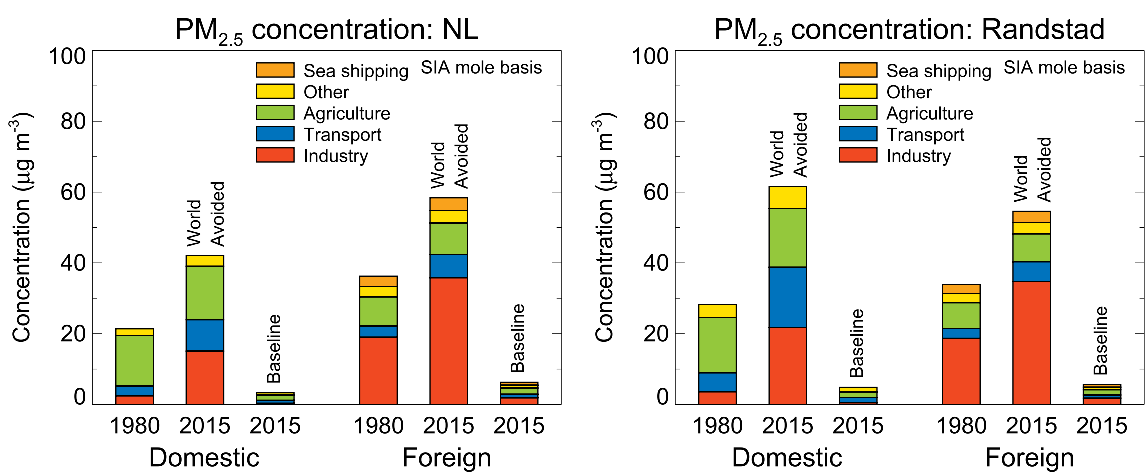

Fig. 5. Contribution of the different sectors to the average $PM_{2.5}$ concentration for the Netherlands (left panel) and for the average of the four largest cities in the Randstad (right panel). Shown are the contributions of emissions from the domestic sectors industry (including electricity production, refineries, and waste), transport (on roads and off road), agriculture, sea shipping and others (households, etc.) and the contributions of emissions from sectors in other European countries (Foreign) for 1980 and 2015 for the World Avoided and Baseline scenarios. Other contributions, such as those from secondary organic carbon, windblown dust, and sea salt (estimated to be about 2.5 $\mu g\,m^{-3}$) are not shown.

only a major contributor in the Baseline scenario in 2015, although the absolute contribution is about the same as in the World Avoided scenario.

In absolute terms (Fig. 4), the largest reductions in NO_2 concentrations in the Netherlands can be attributed to the reductions in emissions of domestic sectors, especially light duty vehicles (i.e., passenger cars and light commercial vehicles). The reductions in emissions of domestic sectors contribute about 8 $\mu g\,m^{-3}$ and those of foreign sectors about 6 $\mu g\,m^{-3}$ to the reduction from 1980 to 2015 in the Baseline scenario. The contribution from international sea shipping increase by about 2 $\mu g\,m^{-3}$ from 1980 to 2015 in the Baseline scenario. For the average of the four largest cities in the Randstad, the effect of reductions in emissions of domestic sectors (about 13 $\mu g\,m^{-3}$) is much larger than those of foreign sectors (about 4 $\mu g\,m^{-3}$, excluding sea shipping) in 2015. Changes in the emissions from road transport give the largest reduction in NO_2 concentration in the Netherlands, with 5 $\mu g\,m^{-3}$ and 2 $\mu g\,m^{-3}$ from domestic light and heavy duty vehicles, respectively, and 3 $\mu g\,m^{-3}$ from foreign road transport in 2015.

Without measures, the contribution from road transport would have increased from about 17 $\mu g\,m^{-3}$ in 1980 to about 24 $\mu g\,m^{-3}$ in the Netherlands in 2015, while it decreased to about 7 $\mu g\,m^{-3}$ in the Baseline scenario in 2015. Domestic light duty transport is responsible for 11.1 $\mu g\,m^{-3}$ of the avoided contributions in 2015 (World Avoided scenario; Table 6). Domestic industry (5.1 $\mu g\,m^{-3}$), foreign road transport (3.9 $\mu g\,m^{-3}$), domestic non-road transport (3.5 $\mu g\,m^{-3}$), and domestic heavy duty transport (2.0 $\mu g\,m^{-3}$) are also major contributors to avoided NO_2 concentrations in 2015. In terms of foreign countries, the largest contribution in avoided NO_2 concentration comes from the United Kingdom (3.8 $\mu g\,m^{-3}$) and Belgium (1.6 $\mu g\,m^{-3}$). Similar results are seen for the average concentrations in the Randstad, but with larger domestic contributions.

3.2.2. $PM_{2.5}$ and PM_{10} concentrations

The situation is rather different for PM concentrations compared with NO_2. The dominant anthropogenic contributions to the total concentration come from foreign sources (Figs. 5 and 8). The domestic emissions contribute about 40% to the anthropogenic $PM_{2.5}$ concentration in the Netherlands, emissions from Germany about 27%, and from Belgium, France and the United Kingdom combined about 20% for both scenarios in 1980 and 2015. The dominant sectors for $PM_{2.5}$

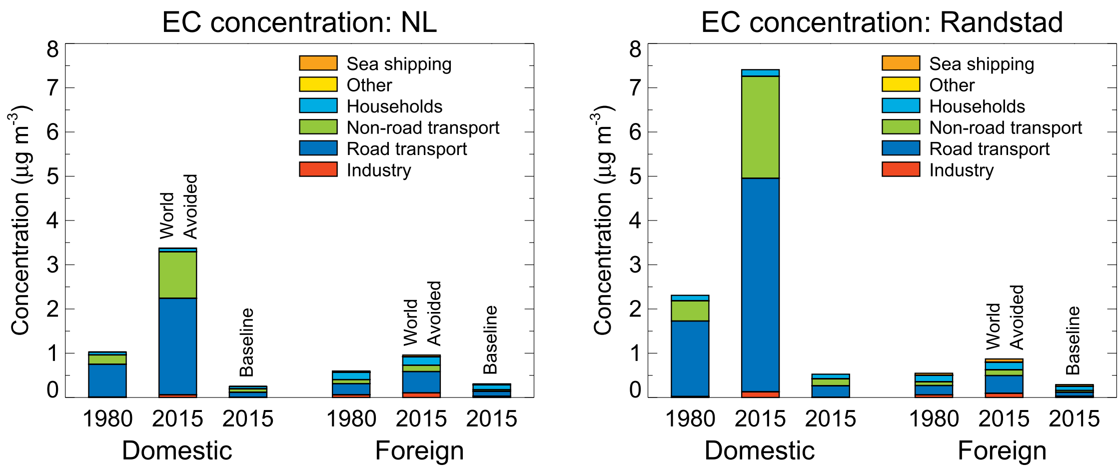

Fig. 6. Contribution of the different sectors to the average EC concentration for the Netherlands (left panel) and for the average of the four largest cities in the Randstad (right panel). Shown are the contributions of emissions from the domestic sectors industry (including electricity production, refineries, and waste), road transport (light and heavy duty), non-road transport, households, sea shipping and others (agriculture etc.) and the contributions of emissions from sectors in other European countries (Foreign) for 1980 and 2015 for the World Avoided and Baseline scenarios.

Table 6
Benefits of air quality emission reductions on the concentrations of air pollutants for the Netherlands[a].

				Secondary inorganic aerosols[b]					
	NO_2	primary PM_{10}	primary $PM_{2.5}$	NO_3	SO_4	NH_4	total PM_{10}	total $PM_{2.5}$	EC
Concentrations ($\mu g\, m^{-3}$)									
1980	29.7	6.8	5.7	9.3	32.5	14.3	72.1	59.0	1.63
2015 World Avoided	45.1	18.5	15.0	19.2	60.3	14.8	122.0	101.8	4.33
2015 Baseline	16.1	3.3	2.3	4.4	2.0	1.9	20.7	11.8	0.56
Avoided contributions ($\mu g\, m^{-3}$) in 2015: World Avoided minus Baseline									
Netherlands	22.7	12.5	10.3	6.4	11.7	8.4	44.7	39.9	3.12
Belgium	1.6	0.7	0.6	1.1	6.4	0.8	8.3	7.4	0.16
France	0.4	0.4	0.4	0.6	3.0	0.3	3.9	3.5	0.10
Germany	0.7	0.9	0.8	1.0	22.2	2.4	27.3	24.1	0.23
United Kingdom	3.8	0.1	0.1	4.1	4.3	0.2	4.8	4.2	0.04
Other countries	0.6	0.5	0.4	1.6	9.9	0.8	11.5	10.2	0.11
National sectors									
Industry[c]	5.1	6.5	4.7	1.9	10.8	0.6	17.9	14.7	0.06
Light duty road transport	11.1	2.6	2.5	2.6	0.2	0.1	4.8	4.3	1.71
Heavy duty road transport	2.0	0.7	0.7	0.6	0.1	0.0	1.1	1.0	0.36
Non-road transport	3.5	2.0	1.9	1.0	0.3	0.0	2.9	2.7	0.98
Households	0.1	0.2	0.2	0.1	0.0	0.7	1.8	1.7	0.02
Agriculture	−0.2	0.0	0.0	0.0	0.0	6.3	14.0	13.6	0.00
Other	1.0	0.6	0.3	0.3	0.3	0.6	2.3	1.8	0.00
Sectors other countries									
Industry[c]	2.2	1.3	0.9	2.8	41.2	0.8	39.0	33.9	0.08
Road transport	3.9	0.7	0.7	4.3	1.1	0.3	4.9	4.2	0.37
Non-road transport	1.0	0.2	0.2	1.1	0.7	0.0	1.5	1.3	0.11
Households	0.1	0.4	0.5	0.2	2.7	0.1	3.1	2.8	0.08
Agriculture	−0.2	0.0	0.0	0.0	0.0	3.3	7.3	7.2	0.00
Sea shipping	−0.8	0.1	0.1	0.0	0.8	0.0	0.8	0.7	0.02
Total	29.0	15.3	12.7	14.8	58.4	12.9	101.3	90.0	3.78

[a] The concentrations are area weighted.
[b] PM_{10} and $PM_{2.5}$ includes, apart from the SIAs, contributions from secondary organic carbon, windblown dust, and sea salt, estimated to be about 10 $\mu g\, m^{-3}$ for PM_{10} and 2.5 $\mu g\, m^{-3}$ for $PM_{2.5}$. For the contributions of the SIAs to the total $PM_{2.5}$ concentration, a fraction of 0.9 is used for SO_4, 0.8 for NO_3 and 1.0 for NH_4. The contributions from the secondary inorganic aerosols cannot directly be added to total PM_{10} and $PM_{2.5}$ contributions because molar ratios are used for the source attribution to the sectors (see Section 2.6).
[c] Industry includes also electricity production, refineries and waste management.

concentration in the Netherlands are industry and agriculture with both about 40% in 1980 and about 23% (industry) and 34% (agriculture) in 2015 (Baseline scenario). Transport (mostly on roads) contributes about 10% to the concentrations in 1980 and about 20% in 2015. Other emissions, mainly from households (domestic and foreign), contribute about 8%–15% to the $PM_{2.5}$ concentrations. These percentages are not very different when considering only the large cities in the Randstad.

The largest reductions in $PM_{2.5}$ concentrations in the Netherlands from 1980 to 2015 can be attributed to the reductions in foreign emissions (about 60%), of which about half is from German sectors. Total $PM_{2.5}$ concentration decreases from about 59 $\mu g\, m^{-3}$ in 1980 to about 12 $\mu g\, m^{-3}$ in 2015 (Baseline scenario). The sectors contributing most to this reduction are foreign industry (about 17 $\mu g\, m^{-3}$ or 36%) and domestic agriculture (about 13 $\mu g\, m^{-3}$ or 27%), followed by foreign agriculture (about 6.5 $\mu g\,m^{-3}$ or 14%). Transport (road and non-road) contributes less, with about 2 $\mu g\,m^{-3}$ (or 4%) for both domestic and foreign transport. Because of the long-range character of $PM_{2.5}$ concentrations, there are no significant increases in concentrations in cities (as in the Randstad) compared with the average for the Netherlands.

Without measures, the $PM_{2.5}$ concentration in the Netherlands would have increased from about 59 $\mu g\,m^{-3}$ in 1980 to about 102 $\mu g\,m^{-3}$ in 2015, while it decreased to about 12 $\mu g\,m^{-3}$ in 2015 in the Baseline scenario. Domestic and foreign industry are responsible for about 15 $\mu g\,m^{-3}$ (16%) and about 34 $\mu g\,m^{-3}$ (38%), respectively, of the avoided contributions in 2015 (World Avoided scenario; Table 6). Agriculture (domestic 13.6 $\mu g\,m^{-3}$ or 15%, foreign 7.2 $\mu g\,m^{-3}$ or 8%) and transport (domestic 8.0 $\mu g\,m^{-3}$ or 9%, foreign 5.5 $\mu g\,m^{-3}$ or 6%) are also major contributors to avoided $PM_{2.5}$ concentrations in 2015. The large reductions in the contributions from industry are the result of reductions in the emissions from SO_2 as can be seen from the contribution of sulphate aerosols in Table 6, while the reductions in the contributions from agriculture come from emissions of NH_3 which produce the ammonium aerosol. The contributions from transport come from both primary $PM_{2.5}$ emissions and nitrate aerosols (through NO_x emissions). The contributions from the primary and secondary aerosols is also shown in Fig. 7 for both the Baseline and World Avoided scenarios.

Very similar contributions and reductions in concentrations are found for PM_{10} as for $PM_{2.5}$ (see Figure B1 Appendix B).

3.2.3. EC concentrations

The largest anthropogenic contributions to the total EC concentration come from domestic emissions (Figs. 6 and 8). The domestic emissions contribute about 63% to the EC concentration in the Netherlands in 1980, while emissions from Germany, Belgium, and France contribute between 7% and 12% each. In 2015, in the Baseline scenario, the contribution of the domestic sectors decreases to about 45%. The dominant sector in 1980 is road transport with 61%, followed by other transport (18%), and households (14%). These sectors are also the dominant sectors in 2015 in the Baseline scenario, but the relative contribution of road transport is reduced, while that of households is increased.

The largest reductions in EC concentrations in the Netherlands from 1980 to 2015 can be attributed to the reductions in domestic emissions (about 73%). Total EC concentration decreases from 1.6 $\mu g\,m^{-3}$ in 1980 to 0.6 $\mu g\,m^{-3}$ in 2015 (Baseline scenario). This reduction comes mainly from domestic and foreign road transport (0.8 $\mu g\,m^{-3}$) and non-road transport (0.2 $\mu g\,m^{-3}$). In contrast to PM_{10} and $PM_{2.5}$, EC has significant spatial gradients in concentration, with high concentrations close to cities and busy roads and low concentrations in rural areas. The average EC concentration in the four largest cities in the Randstad is, therefore, about twice as high as the average for the Netherlands and dominated by domestic emissions. In the Randstad the EC concentration decreases from 2.9 $\mu g\,m^{-3}$ in 1980 to 0.8 $\mu g\,m^{-3}$ in 2015. The domestic emissions are responsible for 81% of the concentration.

Without measures, the EC concentration in the Netherlands would have increased from 1.6 $\mu g\,m^{-3}$ in 1980 to 4.3 $\mu g\,m^{-3}$ in 2015, while it decreased to 0.6 $\mu g\,m^{-3}$ in 2015 in the Baseline scenario. The emissions of domestic sectors are responsible for the majority (about 3.1 $\mu g\,m^{-3}$) of the avoided contributions in 2015 (Table 6), with light duty transport (1.7 $\mu g\,m^{-3}$), and non-road transport (1.0 $\mu g\,m^{-3}$) the major sectors.

3.3. Avoided limit value exceedances

The attribution of the concentrations to different sectors as discussed in Section 3.2 is based on the large-scale concentrations (Section 2.1) of air pollutants. When comparing concentrations with limit values or calculating exposure of populations to air pollutants, local concentrations are needed. This is especially relevant for locations close to busy roads where large spatial gradients in concentrations are found. Local traffic contributions are therefore added to the large-scale concentrations (see Section 2.2).

The air quality limit values addressed here are for NO_2 the annual average concentration of 40 $\mu g\,m^{-3}$, for PM_{10} the daily average concentration of 50 $\mu g\,m^{-3}$ which may not be exceeded more than 35 times a year, and for $PM_{2.5}$ the annual average concentration of 25 $\mu g\,m^{-3}$. The PM_{10} daily limit value is found to be equivalent to an annual average concentration of about 32 $\mu g\,m^{-3}$ (Matthijsen and Visser, 2006; Velders and Matthijsen, 2009). The modelled concentrations are therefore compared with this lower concentration.

In 1980, 1990 the limit values for PM_{10} and $PM_{2.5}$ concentrations are exceeded at all address locations (i.e. all 17 million people) in the Netherlands in both the Baseline and World Avoided scenarios (Fig. 9). The number of exceedances drops rapidly after 1990 to very low values since 2010. The limit value for the annual average NO_2 concentration is exceeded at about 25% of the address locations in 1980. This increases to about 93% in 2015 in the World Avoided scenario, while it drops to less than 1% in the Baseline scenario in 2010.

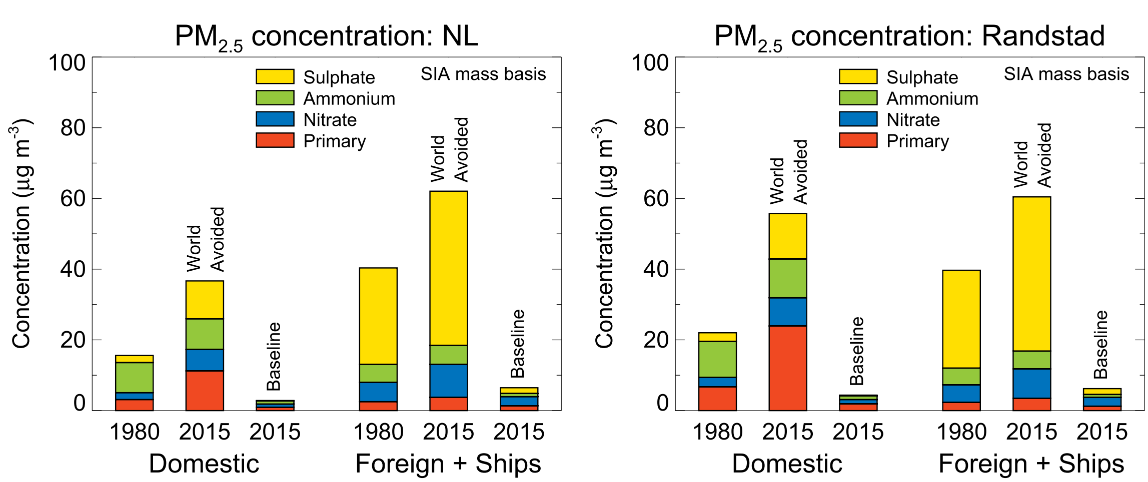

Fig. 7. Contribution of the different types of aerosols (primary aerosol and secondary inorganic aerosols: nitrate, ammonium and sulphate) to the average $PM_{2.5}$ concentration for the Netherlands (left panel) and for the average of the four largest cities in the Randstad (right panel). Shown are the contributions for 1980 and 2015 for the World Avoided and Baseline scenarios. The contributions from sea shipping are included in the foreign contributions. Other contributions, such as those from secondary organic carbon, windblown dust, and sea salt (estimated to be about 2.5 $\mu g\,m^{-3}$) are not shown.

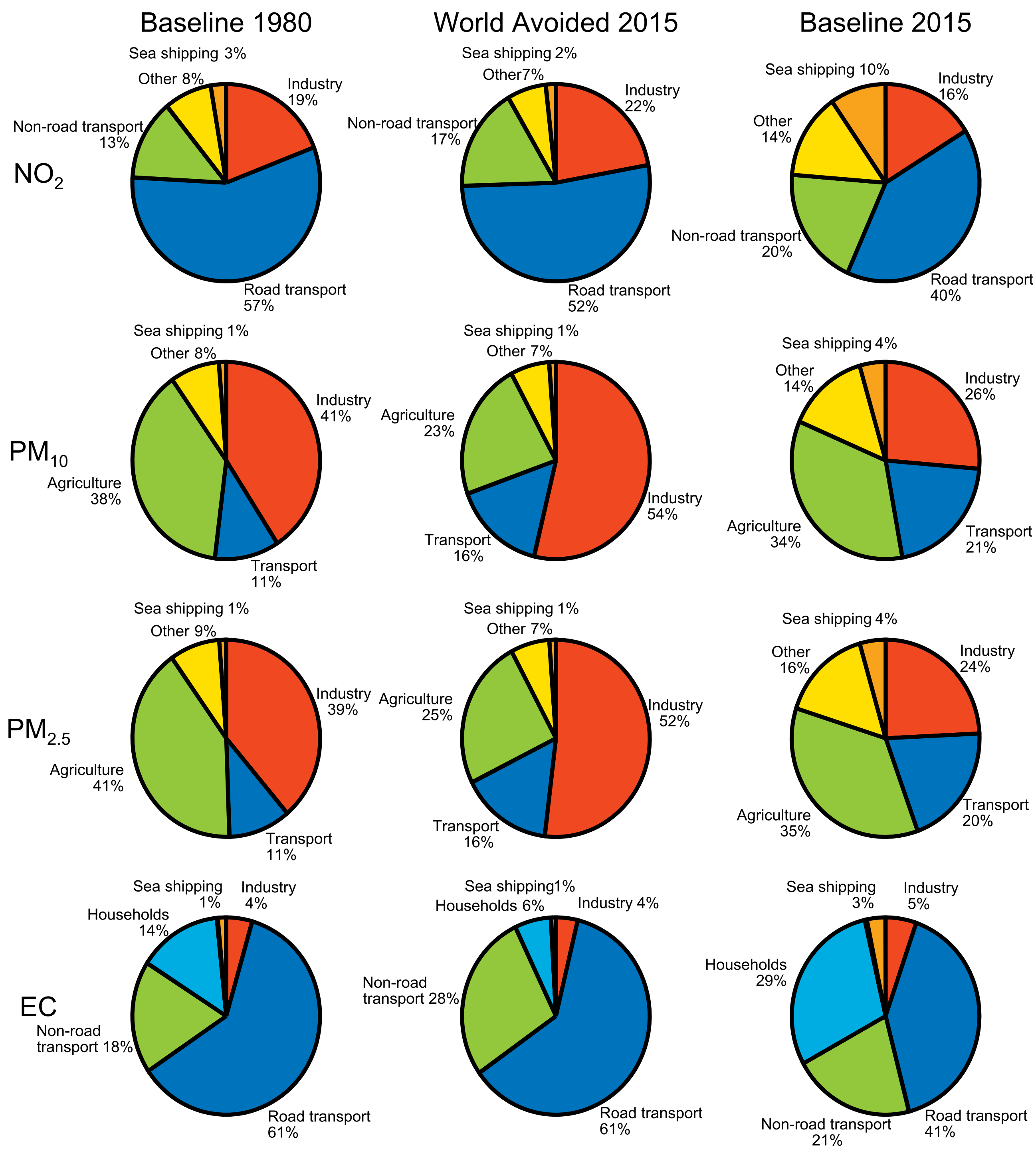

Fig. 8. Relative contribution of the different sectors to the average NO_2, PM_{10}, $PM_{2.5}$ and EC concentrations for the average of the Netherlands. Shown are the combined contributions of domestic and foreign emissions from industry (including electricity production, refineries, and waste), road transport, non-road transport, households, sea shipping, agriculture, and others for 1980 and 2015 for the World Avoided and Baseline scenarios. Similar percentages are found for the average of the four largest cities in the Randstad.

3.4. Avoided health and monetary damage

The DALYs is an indicator used in many studies (GBD, 2016; Holnicki et al., 2017; Murray and Lopez, 1996; WHO, 2016) and therefore also used in Figs. 10 and 11 and Table 7 to show the impacts of exposure to air pollution (NO_2 plus $PM_{2.5}$) on health. In the Baseline scenario the DALYs decrease from about 560,000 in 1980 to 135,000 in 2015, while in the World Avoided scenario it increases to about 875,000 in 2015. The avoided DALYs of about 740,000 relate for 52% to the emission reductions in the Netherlands and for 22% to Germany (Fig. 11 and Table 7). The avoided DALYs attributable to sectors in the Netherlands relate to 34% from industry, 33% from transport (road and non-road), and 24% from agriculture. The avoided DALYs attributable to sectors in other countries relate mainly to industry (67%), with smaller contributions from agriculture (14%) and transport (14%). So, the avoided emissions from industry in Europe are responsible for half (50%) of the avoided DALYs, while agriculture (19%) and all transport (17%) contribute about one fifth each.

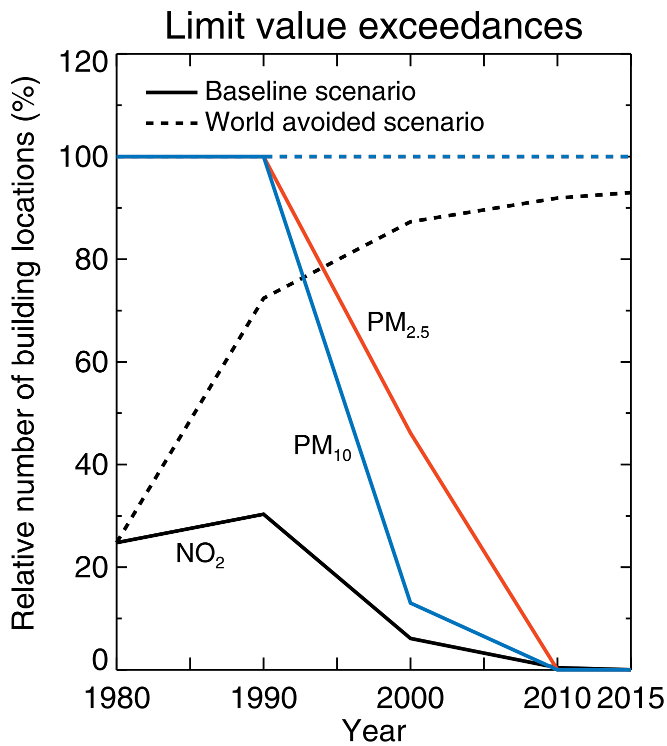

Fig. 9. Relative reduction in the number of address locations were the limit values for NO_2, PM_{10} and $PM_{2.5}$ concentrations are exceeded in the Baseline and World Avoided scenarios. The limit values applied here are: for NO_2 the annual average concentration of 40 μg m^{-3}, for PM_{10} the daily average concentration of 50 μg m^{-3} allowed to be exceeded not more than 35 days, which is equivalent to an annual average concentration of about 32 μg m^{-3}, and for $PM_{2.5}$ the annual average concentration of 25 μg m^{-3}. The total number of address locations of buildings in the Netherlands is about 8.8 million.

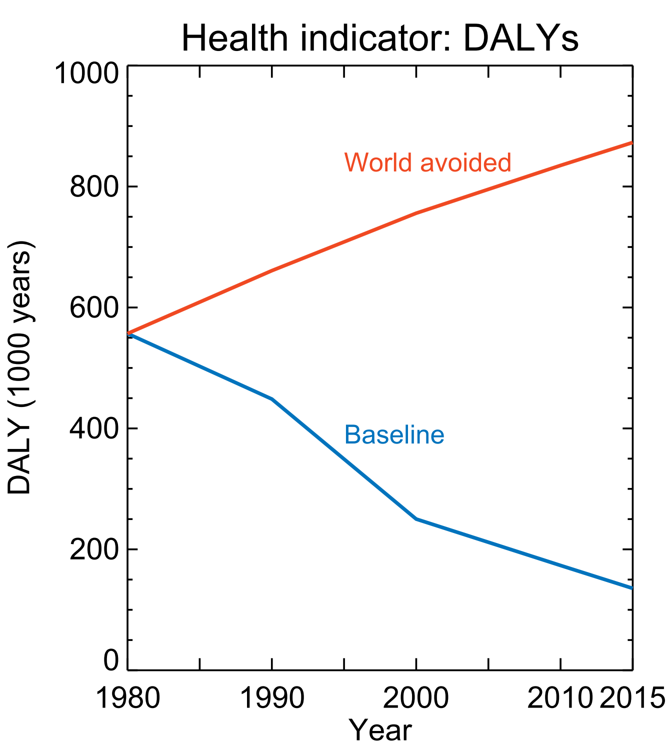

Fig. 10. Number of Disability Adjusted Life Years (DALYs) in the Netherlands in the Baseline and World Avoided scenarios.

The number of years of life lost (YLL) attributable to air pollution in the Netherlands is about 530,000 per year in 1980 (Table 7). In the World Avoided scenario this increases to about 830,000 per year in 2015, while it decreased to about 130,000 in the Baseline scenario in 2015. So, the avoided emissions resulted in a reduction in YLL of about 700,000 per year in 2015. The associated avoided number of attributable (or premature) deaths in the Netherlands is about 66,000 per year in 2015.

According to our estimates, in 2015, around 9% of the total mortality in the Netherlands can be attributed to air pollution. In the World Avoided scenario this percentage would have increased to 50%. The loss of life expectancy in 1980 caused by exposure to air pollution is about 4 years. In the World avoided scenario this increases to about 7 years in 2015, while it decreased to about 1 year in the Baseline scenario in 2015. So, without reductions in emissions the average life expectancy would have been 6 years shorter than actually is the case in 2015.

The current contributions (i.e., Baseline scenario in 2015) of the different countries and sectors to the DALYs are shown in Fig. 12. The largest avoided DALYs can be attributed to the sectors in the Netherlands. The Dutch sectors are also, with 49%, the largest contributors to the DALYs in 2015, but contributions from German sectors in particular are significant. Four European sectors make up large contributions to the DALYs, with 35% from all transport, 22% from agriculture, 19% from industry, and 13% from households. So, for further reductions in the health effects of air pollution, currently estimated at 135,000 DALYs or a loss of life expectancy of about 12 months, emission reductions are needed in all sectors in both the Netherlands and neighbouring countries. The 135,000 DALYs calculated for 2015 are slightly higher than the 109,000 DALYs reported for the Netherlands in 2010 by the WHO (2015).

Monetary health damage can be estimated from health effects. As total health impact is dominated by the mortality attributable to PM and NO_2 exposure, we estimated the monetary damage from air pollution at € 26 to € 58 billion per year in 1980 and € 6 to € 14 billion per year in 2015 (Baseline scenario). Here we have used a value of a life year of € 50,000 to € 110,000 (Amann et al., 2017; De Bruyn et al., 2017). In the World avoided scenario the monetary damage is estimated at € 42 to € 91 billion per year in 2015. The avoided damage between 1980 and 2015 amounts, therefore, to € 35 to € 77 billion per year or on average € 2100 to € 4500 per capita. The WHO (2015) estimated the monetary damage associated with premature deaths alone at about € 21 billion per year (about $ 26 billion for 2010), based on a significantly higher value of a life year. For an overall costs-benefit analyses, the costs associated with the implementation of the various air quality regulations also need to be considered, but this is outside the scope of this work.

4. Discussion and conclusions

The avoided concentrations of air pollutants and associated health impacts, as discussed in the previous sections, originate from reductions in emissions in various sectors and are driven by autonomous technological developments and/or policy measures (Table 5). National emission reductions are obliged under the Gothenburg protocol (UNECE, 1999) and EU National Emission Ceilings directive (EU, 2001) in which ceilings are set for the emissions of SO_2, NO_x, $PM_{2.5}$, NH_3 and VOCs in 2010 and 2020.

The largest contribution (50% of the DALYs) to the avoided health impacts can be attributed to industry (including electricity production and refineries) and associated with avoided SO_2 emissions, since the reductions in sulphate aerosols are by far the largest contributor to the reductions in PM_{10} and $PM_{2.5}$ concentrations (Fig. 7). The contribution of the SO_2 emission reductions from industry in foreign countries is about four times larger than from industry in the Netherlands. The SO_2 emission reductions are the result of the EU directives for limiting SO_2 emissions, such as the directives on the emissions of large combustion plants and other industrial emissions, as well as on the sulphur content

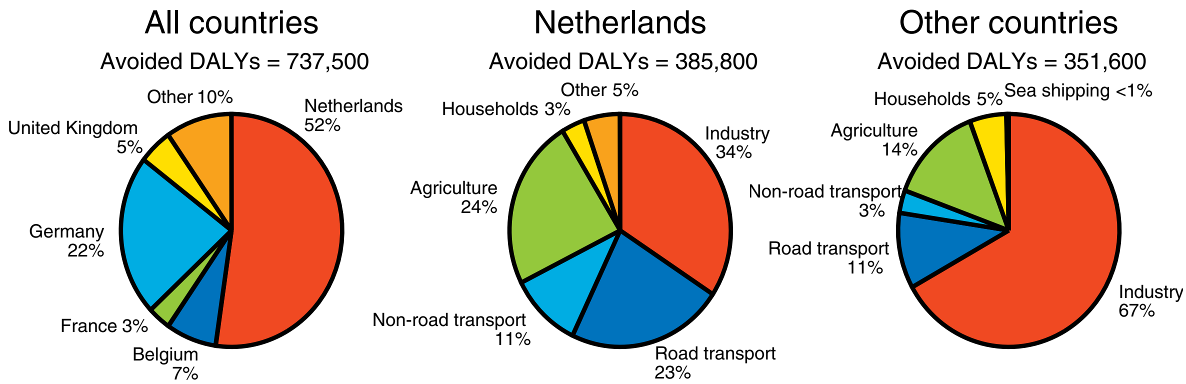

Fig. 11. Contributions of the different countries and sectors to the avoided number of Disability Adjusted Life Years (DALYs) in the Netherlands in 2015. The avoided DALYs are calculated as the difference between the World Avoided scenario and the Baseline scenario in 2015. Shown are the avoided DALYs for all countries (left panel), for the different sectors in the Netherlands (middle panel), and for the different sectors in other countries (right panel).

Table 7
Benefits of air quality emission reductions on the exposure to NO_2 and $PM_{2.5}$ and associated health effects for the Netherlands[a].

	NO_2 ($\mu g\,m^{-3}$)	$PM_{2.5}$ ($\mu g\,m^{-3}$)	Attributable deaths	YLL: Years of life lost	LLE: Loss of life expectancy (months)	DALYs (years)	Monetary health damage (mln Euro[c])
Absolute values							
1980	35.8	61.4	49,900	528,500	48	556,900	26,400–58,100
2015 World Avoided	51.3	108.3	78,400	830,900	85	872,900	41,500–91,400
2015 Baseline	21.0	12.5	12,100	128,300	12	135,400	6400–14,100
Avoided contributions in 2015: World Avoided minus Baseline							
Netherlands	26.2	45.7	34,700	367,600	37	385,800	18,400–40,400
Belgium	1.2	7.5	4900	51,800	6	54,400	2600–5700
France	0.3	3.5	2200	23,600	3	24,800	1200–2600
Germany	0.5	24.1	14,800	157,000	17	164,700	7800–17,300
United Kingdom	2.7	4.2	3300	34,500	3	36,200	1700–3800
Other countries	0.4	10.2	6300	66,900	7	70,300	3300–7400
National sectors							
Industry[b]	5.6	17.1	11,900	125,800	13	132,100	6300–13,800
Light duty road transport	12.5	5.4	6500	69,300	6	72,800	3500–7600
Heavy duty road transport	1.9	1.4	1400	14,700	1	15,400	700 - 1600
Non-road transport	4.6	4.0	3600	38,700	4	40,600	1900–4300
Households	0.0	1.9	1200	12,600	1	13,200	600 - 1400
Agriculture	−0.3	13.6	8200	87,100	10	91,400	4400–9600
Other	1.9	2.2	1800	19,400	2	20,400	1000–2100
Sectors other countries							
Industry [b]	1.6	33.9	21,100	224,100	24	235,300	11,200–24,700
Road transport	2.9	4.2	3300	35,300	4	37,000	1800–3900
Non-road transport	0.8	1.3	1000	10,600	1	11,100	500 - 1200
Households	0.0	2.8	1700	17,900	2	18,800	900 - 2000
Agriculture	−0.2	7.2	4300	45,900	5	48,200	2300–5100
Sea shipping	−1.1	0.7	100	1200	0	1200	0–100
Total	30.2	95.8	66,300	702,600	73	737,500	35,100–77,300

a) The concentrations are population weighted (i.e., weighted by the address locations of all buildings in the Netherlands). Health impacts are calculated for $PM_{2.5}$ concentrations above 2.5 $\mu g\,m^{-3}$ and NO_2 concentrations above 5 $\mu g\,m^{-3}$. For the health effects a total population of 17 million is used.
b) Industry includes also electricity production, refineries and waste management.
c) In 2015 prices.

of liquid fuels (Table 5).

The second largest contribution (24% of the DALYs) to the avoided health impacts can be attributed to road transport (17%) and non-road transport (7%), with the contributions from the Netherlands being about three times that from the other countries combined. Reductions in emissions of NO_x, primary $PM_{2.5}$, and SO_x contribute in decreasing order to these health benefits. The NO_x and primary $PM_{2.5}$ emission reductions are mainly the result of the EU directives on motor vehicles (Euro emission standards for passenger cars and light and heavy duty vehicles), notwithstanding the fact that the real-world emissions are higher than was expected based on the standards (Jonson et al., 2017; Velders et al., 2011a). For primary $PM_{2.5}$ emissions the particulate filters have proved to be very effective in reducing emissions (Spreen et al., 2016a). The reductions in SO_x emissions from road transport can be directly attributed to the EU directive on the sulphur contents of liquid fuels (CEIP, 2018).

The third largest contribution (19% of the DALYs) to the avoided health impacts can be attributed to agriculture in the Netherlands and

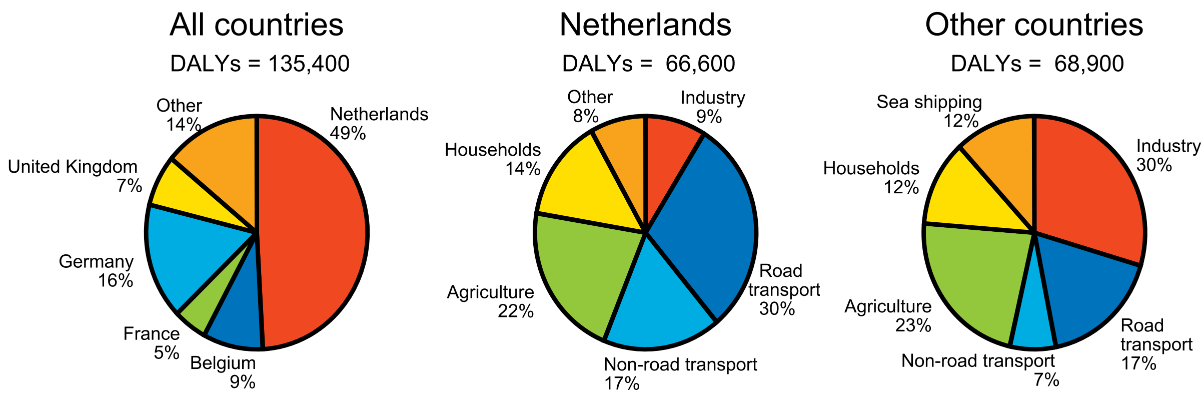

Fig. 12. Contributions of the different countries and sectors to the Disability Adjusted Life Years (DALYs) in the Netherlands in the Baseline scenario in 2015. Shown are the DALYs for all countries (left panel), for the different sectors in the Netherlands (middle panel), and for the different sectors in other countries (right panel). DALYs are only calculated for the contribution of the anthropogenic emissions.

other countries, with the former being about twice as large as the latter. These benefits, which are associated with reductions in NH_3 emissions, with more or less constant livestock numbers, are likely driven by national regulations to reduce emissions from applying manure to agricultural land and from stables. These emission reductions are driven by the national emission ceilings for NH_3.

The final significant contribution (4% of the DALYs) to the avoided health impacts can be attributed to households and specifically the reductions in SO_x emissions in foreign countries associated with the EU directive on the sulphur contents of liquid fuels and shifts in the use of coal for residential heating.

The Euro standards for passenger cars and light and heavy duty trucks are not taken into account in the World Avoided scenario. Changes in traffic volumes (Fig. 1) are taken into account, but changes in the fleet composition are not. For example, the passenger car fleet in 1980 in the Netherlands consisted for the most part of gasoline cars. Since 1990, the number of diesel cars has increased rapidly (this is even more the case in many other EU countries, where the share of diesel cars in the fleet has increased to over 50 percent in some cases). The impact of these changes in the fuel mix is not taken into account in this analysis. As such, the resulting reduction in $PM_{2.5}$ emissions might be underestimated, as diesel cars have higher $PM_{2,5}$ emissions per vehicle kilometre than gasoline cars.

Apart from specific policies to limit emissions, autonomous developments, such as improvements in energy efficiency and changes in the mix of fuels used, also effect emissions. Autonomous developments are in part taken into account in the World Avoided scenario through the activity data that drives the emissions, but the effects of autonomous developments and of policies, are hard to disentangle. In this study we define the benefits of policies in terms of concentration reductions and health effects as the difference between the World Avoided and the Baseline scenario in 2015. The benefits could also be ascribed to the difference between the Baseline scenario in 1980 and 2015 but, considering the large increases in, e.g., gross domestic product, industrial output, and traffic volumes, it is likely that emissions would have increased, relative to 1980 levels, without policy measures. The health benefits from the decrease in emissions from 1980 to 2015 only (Baseline scenario) are still significant; about 50–60% of those described above.

As discussed above, the obtained health benefits are the results of emission reductions in industry, agriculture and road transport. In 2015 in the Baseline scenario, the largest contributions to the health effects of air pollution can be attributed to emissions from these same sectors, including from households, with agriculture the largest contributor of the four sectors (Fig. 12). The current contribution from sectors in the Netherlands is about 49%, while sectors in other counties and sea shipping make up the remaining 51% of the health impacts.

In this study the impact of changes in air pollution alone on health from 1980 to 2015 is assessed. The relationships between ambient air pollution and health effects may not be completely linear, especially under high concentrations (see e.g. Pinault et al. (2017)), which is taken into account in this study. Also, the health impacts of air pollution not only depend on the air pollution concentrations, but also on changes in, e.g., the age structure of the population, mortality rates, and medical treatments. For example, the mortality rate is higher for older people, and as the population is aging, this means that relatively more deaths that are attributable to air pollution can be expected among elderly people. On the other hand, improvements in medical treatment reduce the mortality rates and also the number of attributable deaths, as deaths that are attributable to air pollution are a fixed percentage of total mortality rates.

In conclusion, large increases in concentrations of many air pollutants have been avoided in the Netherlands through reductions in emissions of SO_2, NH_3, NO_x and primary PM in the Netherlands itself and in other countries in Europe. The largest health effects of air pollution are attributed to $PM_{2.5}$ with smaller contributions from NO_2. The avoided health effects can be attributed to reductions in emissions in the sectors in the Netherlands (52%), Germany (22%) and other countries, with the largest contributions from industry (50%), transport (24%), and agriculture (19%).

It is clear that public health in the Netherlands has profited from international cooperation to abate transboundary air pollution. Similarly health in surrounding countries will have profited from the measures taken in the Netherlands as the country was, and still is, a net-exporter of NO_2 and PM.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2019.117109.

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