Effects of traffic policies on air pollution and health – an intervention study

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Effects of traffic policies on air pollution and health – an intervention study

Effecten van verkeersmaatregelen op lucht-verontreiniging en gezondheid – een interventiestudie

(met een samenvatting in het Nederlands)

Proefschrift

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Contents

Chapter 1

General introduction

Health effects of air pollution

Associations between air pollution and numerous adverse health effects are now well established[1-4]. Exposure to air pollution has been linked to a number of different adverse health effects, ranging from the more modest transient changes in the respiratory tract and impaired pulmonary function, to restricted activity/reduced performance, emergency room visits, hospital admissions and mortality[3]. There is now increasing evidence for adverse health effects of air pollution not only on the respiratory system, but also on the cardiovascular system. This evidence stems from studies on both acute (short-term) and chronic (long-term) exposure to air pollution. An analysis by the World Health Organization (WHO) indicated that (fine particulate) air pollution levels resulted in an average loss of about 9 months of life expectancy in Europe[5]. This analysis was partly based upon large cohort studies performed in both the USA as well as in Europe where the health status of many people was followed for a long time and long-term exposure to air pollution was assessed. Air pollution effects are not only restricted to more susceptible persons like children, asthmatics or the elderly[6]. Overall global burden of diseases due to air pollution is about 3% of mortality from cardiopulmonary disease, about 5% of mortality from cancer of the trachea, bronchus and lung, and about 1% of mortality from acute respiratory infections in children under 5 yr, worldwide[7]. Burden of disease due to air pollution will be even larger when morbidity is included as well[8].
Health effects of traffic-related air pollution have been studied extensively in the past decade. Many studies documented adverse health effects of living close to major roads[9].

Efforts to reduce air pollution

Air pollution has substantially decreased in the past decades due to effective emission control of both stationary and mobile air pollution sources[1]. Nowadays, traffic can be considered as the major source of air pollution in residential areas in the Netherlands, as well as in other Western countries[10]. Increases in urban populations, number of cars and vehicle km travelled are a few trends that suggest that exposure to traffic-related air pollution is on the rise[9]. More stringent emissions controls and cleaner fuels have resulted in lower emission factors per vehicle, may (partly) counteracting these trends.
To protect public health, the European Union has set air quality standards of which the standards for particulate matter (particles smaller than 10 μm (PM_{10}) and smaller than 2.5 μm ($PM_{2.5}$)) and nitrogen dioxide (NO_2) are the most important. Air quality standards will become stringent in 2015. However, the current values for PM are above recommendations by the WHO as well as above air quality standards in the USA and other developed countries[11]. In addition, until now epidemiological studies have not identified a threshold

below which no adverse health effect occurred. This means that although compliance is met to EU standards for certain pollutants, adverse health effects may still be substantial[12].
European-wide policies to reduce air pollution emissions from traffic are substantial including emission standards for motor vehicles, definition of national emission ceilings and regulations for fuel. Air quality standards are still exceeded especially near busy roads. Local policies have been implemented to reduce air pollution concentrations at busy urban roads. Examples of local traffic policy measures include congestion charging directed to all passenger cars entering the inner-city, and low emissions zones (LEZ) designed to limit traffic of old vehicles in city centres.
Policy makers and the public want to know whether the efforts to reduce air pollution have paid off[13]. The costs associated with compliance to increasingly tight air quality standards and, importantly, the need to ensure that those policies are effective and achieve the intended public health benefits have been important driving forces for a series of empirical studies of the health benefits of policy interventions[13]. Intervention studies may contribute substantially in the causality debate, and would support further efforts to stringent air pollution standards.

Intervention studies

One of the most quoted examples is the study of Clancy et al.[14] on a coal ban in Dublin, Ireland. After the coal ban in 1990 air pollution concentrations (soot) dropped almost immediately with about 70%. Standardized total mortality rates declined as well with about 5.7%, whereas respiratory mortality rates decreased with about 15.5%. Another study published at the same time originated from Hong Kong, China where the government restricted the use of sulphur in gasoline[15]. Air pollution (sulphur dioxide) concentrations dropped significantly (45%) and up to a 3.9% decline in respiratory mortality was found.
After these striking results, other initiatives to evaluate policies have started to emerge. The US-based Health Effects Institute (HEI) has promulgated a research programme Accountability since 2003 to investigate interventions[13]. To evaluate policies in the Accountability research programme of HEI, a full chain approach has been used where policies are assessed at different stages including drivers, emissions, concentrations, exposures, doses and health effects (Figure 1). Studies typically compare air pollution and/or population health before and after implementation of a policy intervention.
Opportunities like a city-wide coal ban or a sulphur-restricted fuel policy with such a dramatic impact, implemented within a very short time period, and causing long-term changes are rare. Most studies investigated policies with more subtle changes in air pollution. Some studies investigated policies implemented more gradually, like the woodstove exchange program in Montana, USA[16], and the LEZ in London, UK[17]. The LEZ targeted different type

of vehicles at different time points between 2008 and 2012, which makes a proper evaluation extremely challenging. Some studies investigated policies which are only temporarily in place, like the traffic policies taken at the Olympics in for example Atlanta, USA[18] and Beijing, China[19,20]. Especially during the Olympics in Beijing radical policies were implemented including tightening of fuel standards, banning old vehicles and incorporating odd and even number plate days for cars. Substantial reduction in air pollution concentrations up to 70% (Beijing) was found during the Olympics compared to periods before[19].

Some studies showed that policy interventions resulted in reductions in mortality[14,15,21,22], respiratory-related hospital admissions and emergency room visits[23-25], and reduced airway inflammation[19]. Other studies have found associations between long-term decrements in air pollution concentrations and mortality[26,27] and improvements in lung function[28,29].

Intervention studies can be constrained by limited power (due to small reductions in air pollution, small changes in health outcomes, a limited number of days with reduced air pollution, or a combination of these) and inability to control for potential confounders such as temporal trends, changes in behaviour and in health care utilization, and widespread epidemics[18]. Methodological issues of intervention studies will be described in more detail in the discussion chapter (Chapter 7).

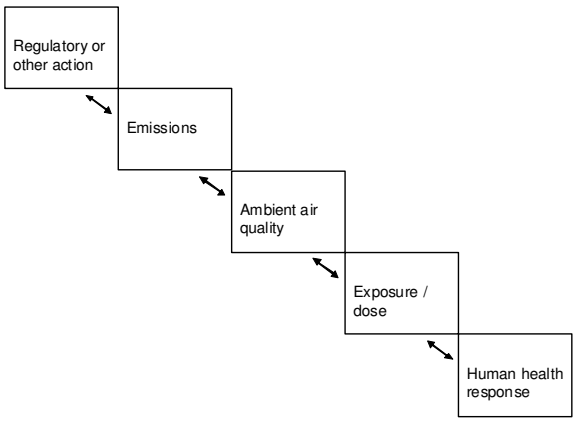

Figure 1 Chain of accountability, adapted from van Erp et al.[13]

Traffic-related air pollution

Traffic-related air pollution originates from combustion and wear of tyres, brakes and road surface and is a complex mixture of many particulate and gaseous pollutants. Highest traffic-related air pollution concentrations occur at or close to major roads up to a few hundred meters depending on the pollutant, spatial characteristics and meteorological conditions. At greater distances traffic-related air pollution concentrations is more difficult to trace as it is well mixed with other air pollution sources[9].

Important traffic-related air pollutants include PM_{10}, $PM_{2.5}$, transition metals like copper (Cu) and iron (Fe), and NO_2. PM air pollution consists of both solid and liquid particles suspended in the air that vary in number, size, shape, surface area, chemical composition, solubility and origin[2]. For practical and regulatory purposes, a size distinction based on particle mass is made between PM_{10}, $PM_{2.5}$ and ultrafine particles (UFP, particles with a diameter of 0.1 µm). UFP is not regulated, and contributes little to particle mass but is most abundant in terms of numbers and offer a very large surface area. Therefore, UFP is characterised by particle number concentration (PNC) rather than particle mass. Contrasts of PM_{10} and $PM_{2.5}$ close to major roads are small compared to other more traffic-related components like soot and PNC[30]. Apart from components directly emitted from the engine, non-exhaust emissions of traffic like from brake- and tyre wear, attracts significant attention recently[31]. Transition metals like Cu and Fe are examples of mainly non-exhaust PM components which can be highly elevated near major roads as well.

NO_2 is a gaseous component and is regulated as well. Until now it is still not clear whether NO_2 can cause adverse health effects by itself or that it serves as a marker of traffic-related air pollution[3]. NO_2 can be directly emitted from the engine (primary NO_2) or formed in the atmosphere by nitrogen oxide (NO) and ozone (O_3) (secondary NO_2). The fraction of primary NO_2 was typically about 5% of the NO_x emission of motor vehicles, but is rising due to an increase in diesel cars and their particulate filters in most European countries[32,33].

Oxidative potential of PM is a novel metric, and may provide a more health-based exposure measure by integrating various biologically relevant properties of traffic-related PM into a single predictor of biological activity[34]. Oxidative stress has been suggested as an important underlying mechanism of action by which exposure to PM may lead to adverse health effects[35].

Omission of a more traffic-related component in a traffic policy evaluation may lead to an underestimation of the health impacts, because reductions of these components are likely more substantial in response to these policies than reductions in for example the regulated components.

Aim of the thesis

The aim of this thesis is to assess the effect of local traffic policy measures on 1) air quality; 2) oxidative potential of PM and 3) respiratory health of residents.

The study was performed in collaboration with the National institute for Public Health and the Environment (RIVM), Energy Research Centre of the Netherlands (ECN), Municipal Health Service Amsterdam (GGD Amsterdam), and the Municipal Health Service Brabant/Zeeland. The study was funded by the Dutch Ministry of Infrastructure and the Environment with additional funding from the Province of Noord-Brabant.

Study design

We started a study to evaluate the air quality and health benefits of local traffic policy measures including low emission zones (LEZ) directed at heavy duty vehicles in several Dutch cities. The study includes comprehensive measurements of air quality and population health status before and two years after the implementation of the policies. We included a control population to account for generic trends not related to the policy. Exposure to air pollution was measured in the same street where the residents lived. Measurements of air pollution were performed for a large range of air pollutants: PM_{10}, $PM_{2.5}$, PNC, soot, NO_2, NO_x and elemental composition of PM_{10} and $PM_{2.5}$. In addition oxidative potential of PM was assessed. Respiratory function of residents was assessed twice by spirometry and interrupter airway resistance. NO in exhaled air was measured as a marker for airway inflammation.

Local traffic policy measures

To date, LEZ have been implemented in 152 cities in nine EU countries, and are being considered for other cities worldwide[36]. From July 2007 to October 2008 the LEZ was gradually implemented in several Dutch cities. The aim of the policy was to comply with EU air quality standards for PM_{10} and NO_2. The major component of the policy was directed at forbidding old trucks to enter the LEZ in the inner-city. Initially, only EURO-0 en EURO-I trucks were forbidden, whereas EURO-II and EURO-III trucks were only allowed if they where retrofitted –if filters were available. Since 2010 all EURO-II trucks are forbidden as well, and EURO-III trucks are only allowed if retrofitted with particulate filters and if not older than 8 years. The emission standards progressively decrease NO_x and PM emissions with about a factor 2 (NO_x) to 8

(PM) comparing EURO-IV with EURO-II trucks, due to the increasingly stringent EURO standards. Enforcement was tightened with cameras and special staff since 2010, with high fines for drivers entering illegally.
Apart from the LEZ other traffic policies measures were introduced in the same period as well. In the inner-city of The Hague a traffic circulation plan was implemented in 2009 aimed at reducing traffic flows at hotspots including the street we studied.

Outline of the thesis

In chapter 2 and 3 we compare the difference in concentration between major urban roads and background locations for a large range of regulated and non-regulated air pollutants, prior to the introduction of the policies.
Chapter 4 describes the oxidative potential of particles at different major urban roads and background locations simultaneously measured.
In chapter 5 air pollution concentrations before and after the implementation of local traffic policies including the LEZ are evaluated.
Chapter 6 describes the relation between changes in air quality and changes in the health status of residents.
Finally, in chapter 7 we discuss the main results, strengths and limitations of this study and of other intervention studies in general.

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Chapter 2

Contrast in air pollution components between major streets and background locations: particulate matter mass, black carbon, elemental composition, nitrogen oxide and ultrafine particle number

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Atmospheric Environment 2011;45:650-8.

Abstract

Background
Policies to reduce outdoor air pollution concentrations are often assessed on the basis of the regulated pollutants. Whether these are the most appropriate components to assess the potential health benefits is questionable, as other health-relevant pollutants may be more strongly related to traffic. The aims of this study were to compare the contrast in concentration between major roads and (sub)urban background for a large range of pollutants and to analyse the magnitude of the measured difference in the street–background for major streets with different street configurations.

Methods
Measurements of PM_{10}, $PM_{2.5}$, PNC, soot, elemental composition of PM_{10} and $PM_{2.5}$ and NO_x were conducted simultaneously in eight major streets and ten (sub)urban background locations in the Netherlands. Measurements were done six times for a week during half a year in 2008.

Results
High contrasts between busy streets and background locations in the same city were found for chromium, copper and iron (factor 2-3). These elements were especially present in the coarse fraction of PM. In addition, high contrasts were found for soot and NO_x (factor 1.8), typically indicators of direct combustion emissions. The contrast for PNC was similar to soot. NO_2 contrast was lower (factor 1.5). The largest contrast was found for two street canyons and two streets with buildings at one side of the street only.

Conclusion
The contrast between busy streets and urban background in NO_2 was less than the contrast found for soot, PNC and elements indicative of non-exhaust emissions, adding evidence that NO_2 is not representing (current) traffic well. The study supports a substantial role for non-exhaust emissions including brake- and tyre wear and road dust, in addition to direct combustion emissions. Significant underestimation of disease burden may occur when relying too much on the regulated components.

Introduction

Numerous studies have documented the effects of air pollution on morbidity and mortality from respiratory and cardiovascular diseases[1,2]. Efforts to reduce air pollution emissions from stationary and mobile sources are substantial in Europe. The evaluation of the effectiveness and health benefits of traffic policies is usually modeled instead of measured, focusing on regulated components. It is now well-documented that proximity to traffic is much better reflected in contrasts in PNC or soot than in PM_{10} and $PM_{2.5}$ levels[3-6]. Omission of these components in an evaluation of a traffic policy measure may lead to an underestimation of the health impacts of traffic policies. Often the easy to measure and regulated pollutant NO_2 is used to characterise traffic–related air pollution e.g. in the evaluation of the London congestion charging zone[7].

The issue of concentration contrasts of various pollutants is also of importance in the interpretation of studies that have shown increased cardio-respiratory morbidity and mortality related to living near major roadways[8]. It has been argued that PNC may play a role in this association, partly based upon the large contrast in concentration[9]. However, the contrast of other components may be even higher.

With the reduction of tailpipe emissions, interest in non-tailpipe emissions is increasing. Johansson et al.[10] reported high elemental concentrations of Cu and Sb in a street canyon compared to an urban background location. Another study showed that in particular Fe, Cu, Ba and Sb were elevated at a roadside compared to background locations[11]. Currently few exposure studies have measured an extensive set of traffic-related air pollutants at various urban roads simultaneously. From the different elements, in particular Cu, Fe, and Zn are of toxicological interest since these transition metals may play a role in the pro-inflammatory effects of PM. There is also considerable evidence that ultrafine particles and soot are related to adverse health effects[12].

We started a study to evaluate the air quality and health benefits of low emission zones directed at heavy duty vehicles in several Dutch cities. The study includes comprehensive measurements of air quality and population health status prior to the implementation of the policy and two years after the implementation. In this paper we report on the 2008 baseline air quality measurements of traffic-related air pollution at moderately busy streets, urban and suburban backgrounds locations. The aims of this study were to compare the quantitative contrast in concentration between the major roads and background for a large range of pollutants: PM_{10}, $PM_{2.5}$, PNC, soot, NO_2, NO_x and elemental composition of PM_{10} and $PM_{2.5}$ and to analyse the magnitude of the measured difference in the street–background for major streets with different street configurations.

Methods

Study design

The evaluation of the air quality and health benefits of low emission zones in the Netherlands includes comprehensive measurements of air quality in the second half of 2008 and 2010. The major component of the policy was directed at forbidding old heavy duty vehicles to enter the low emission zone of the city. Inner-city streets were selected that were expected to be affected by the policy. Eight street locations were selected in five Dutch cities: Amsterdam (750 000 inhabitants), The Hague (480 000 inhabitants), Utrecht (280 000 inhabitants), Den Bosch (136 000 inhabitants) and Tilburg (220 000 inhabitants) (Figure 1). Most important local source of air pollution in those cities is traffic, since in none of the cities large industrial sources were present. Shipping can be another local source, especially in the channels of Amsterdam. The smallest distance from a sampling site in Amsterdam was 30 m away from a minor canal.

For each busy street location, an urban background and a suburban location in the neighbourhood of the city were chosen to disentangle temporal trends related to the local policy and more generic emission trends or weather patterns. In total eighteen monitoring sites were selected: eight busy streets, five urban background sites (one per city) and four suburban background sites. One suburban location was used for two nearby cities (Den Bosch, Tilburg). An additional monitoring site was selected as a reference site where continuous measurements were made. At each site, one-week measurements of PM_{10}, $PM_{2.5}$, soot, NO_2 and NO_x have been conducted six times during June 2008–January 2009. All PM_{10} and $PM_{2.5}$ filters were analysed for elemental composition using energy dispersive X-ray fluorescence spectrometry. PNC and the size distribution of particles were measured at two busy streets and the two nearby suburban backgrounds because of the health study involving the population in the street and in the suburban area.

Characteristics of the monitoring sites

The eight street locations had a traffic intensity varying between approximately 10 000 and 19 000 motorized vehicles passing every 24-hour (Table 1). The sampling site was located between 9 and 15 m from the road axis. Two street locations can be considered as canyon type of streets. Equipment at the street locations was placed outside a dwelling or at a balcony at the first floor. Inlets were placed 4-5 m above ground level.

The five urban background locations were located in the city centres but not in the direct vicinity of busy streets (>50 m). Distance to the street sites ranged between 300 m (Koningsweg) and 4 km (Haarlemmerweg). The 4 suburban locations were in villages (~30 000 inhabitants) in the neighbourhood of the selected cities (10–30 km) and are background locations as well (Figure 1). Nearest major road at the suburban background location was 100 m. Equipment of all but two of the background locations was placed in the

backyard of a dwelling and inlets of seven background locations were placed 3-5 m above the ground. At two background locations, inlets were placed 1 m above ground level. The reference background site was located at the Utrecht University Campus, in the centre of the country. None of the locations was near industrial areas, small industries, gas stations or large parking areas.

Ambient concentration measurements

Six one-week average measurements were conducted on eighteen locations. Due to equipment constraints locations were measured in two rounds, Amsterdam plus The Hague sites in one round and Den Bosch, Tilburg and Utrecht in another round. For a valid comparison of the spatial contrast between sites, an additional reference site was operated during all twelve sampling weeks to limit potential bias due to temporal variation. This sampling schedule with discontinuous measurements spread over a year has been used before to assess spatial variation in outdoor air pollution, often in the framework of providing the monitoring bases for land use regression studies[13]. Though efficient and valid estimates of the spatial contrast are obtained, the absolute concentrations can deviate from the annual average. In addition PNC and the size distribution of particles were measured at one street and suburban background location simultaneously.

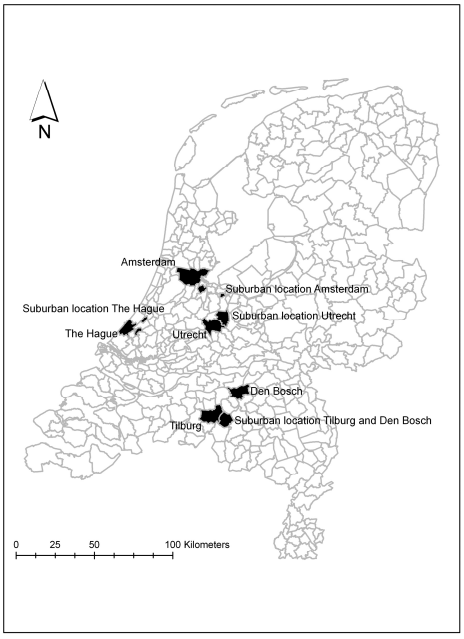

Figure 1 Geographical location of the measurement sites in the Netherlands

Table 1 Detailed characteristics of the different urban streets

Cities	Urban streets	Traffic intensity	Fraction[a]		Speed (km/h)		Road type	Distance road axis
		Per 24-h	Middle[b]	Heavy[c]	Per 24-h	In rush hours		
Amsterdam	Haarlemmerweg	15 253	0.03	0.02	41	38	Adjoining buildings on one side	11
Amsterdam	Hoofdweg	9774	0.01	0.06	41	41	Adjoining buildings on two sides	16
The Hague	Stille Veerkade	17 438	0.05	0.02	34	32	Canyon	11
Den Bosch	Brugstraat	17 896	0.05	0.05	32	28	Canyon	9
Den Bosch	Koningsweg	17 138	0.05	0.03	46	39	Adjoining buildings on two sides	14
Tilburg	HVB	18 812	0.03	0.07	51	49	Adjoining buildings on two sides	12
Utrecht	Vleutenseweg	13 553	0.06	0.05	39	36	Adjoining buildings on two sides	15
Utrecht	Weerdsingel Wz	14 831	0.06	0.03	35	31	Adjoining buildings on one side	9

[a]Fraction of total traffic intensity; [b]distance between wheel axes 3.5-7 m; [c]distance between wheel axes >7 m.

PM_{10}, $PM_{2.5}$ and soot

PM_{10} measurements were conducted with PM_{10} personal samplers (MSP Corp., Shoreview, MN, USA). $PM_{2.5}$ was measured with $PM_{2.5}$ GK2.05 cyclones (BGI Inc., Waltham, MA, USA). Flow-controlled battery operated pumps (BGI-400) were used at a flow rate of 4 l/min (PM_{10}) or 3.5 l/min ($PM_{2.5}$). The sampling flow was measured before and after each sampling period using calibrated rotameters. In order to prevent filter overload, timers were used to sample five minutes during every twenty minutes. Thus in one week, effectively a 42-hour representative sample was taken. Elapsed-time indicators were used to calculate average sample volumes. For all measurements the same type of filters were used (Anderson Teflon 37 mm).

Procedures for weighing and reflectance measurements have been described before[14,15]. Both before and after sampling, all filters were double weighted with a Mettler MT5 micro-balance (Mettler-Toledo, Greifensee, Switzerland). Filters were stored at 4°C and were weighted after equilibrating for 24-hour in a temperature (20-23°C) and humidity (30-40%) controlled room. Reflectance of the PM_{10} filters was measured at five standard spots and average reflectance was calculated. Next, the reflectance was transformed into absorbance according to the following formula[16]:

$$a=(A/2V)*Ln(R_0/R_s).$$

Where a=absorption coefficients (10^{-5}/m), A=loaded filter area (m^2), V=sampled volume (m^3), R_0=average reflectance of field blank filters, and R_s=reflectance of the sampled filter.

Absorbance has been shown to be a good indicator for elemental carbon (EC), black carbon (BC), and soot[17]. In this paper we will further use the term soot for the absorbance measurements. For quality assurance twelve field blanks and nineteen duplicate samples were collected at multiple sites on a rotating basis. Mean field blank weight changes were subtracted from all sample weights.

On two of our Amsterdam sites there were also fixed site monitors running of NO_x (chemiluminescence monitors, EcoPhysics, Switzerland, type CLD 700AL) and PM_{10} (gravimetrically, low volume samplers, Derenda, Germany), operated by the Municipal Health Service Amsterdam. Therefore a comparison was made to test accuracy.

NO_x

Average one-week NO and NO_2 concentrations were measured with OGAWA passive samplers (OGAWA & Company Inc., FL, USA) with pre-coated NO_2 and NO collection pads. They were placed at the same height near the PM equipment. Sampling and analysis was performed according to procedures reported before[18]. Twenty one duplicate samples were taken to test precision. From each batch of forty samples, four lab blanks were retained. In addition, in total twenty field blanks were taken spread over the sampling period. Batch average blank levels were subtracted from all samples.

Elemental composition
All filters were analysed for different elemental components with energy dispersive X-ray fluorescence spectrometry (ED-XRF) at the Cooper Environmental Services, Portland, USA. Quality assurance included testing of standard material from the National Institute of Standards and Technology (SRM 1228, SRM 987) and with a Micromatter Multi-elemental Quality Control Standard (Multi 13391). Measurements of certain elements on certified contents of standard material showed percentages of error up to 7% except for Cu. Cu was underestimated with about 13%. We have not corrected for this since there is no indication that this would be different between locations. To test the precision of the ED-XRF analysis eighteen (9%) of the samples have been re-measured. In this paper, sixteen elements are reported which were consistently above the limit of detection and had a good reproducibility. Mean field blank concentrations were subtracted from all samples. Details of the elemental analyses have been reported earlier[19].

Size specific particle number concentrations
Additional PNC have been performed at two street locations (Haarlemmerweg, Amsterdam and Vleutenseweg, Utrecht) and the two matching suburban location for four and three weeks respectively in the period September 2008–January 2009. Particle size distributions were measured using a Scanning Mobility Particle Sizer (SMPS) comprising a TSI Model 3071 Electrostatic Classifier and Model 3022 Condensation Particle Counter. Procedures have been reported before[20]. The SMPS distinguished particles with a diameter of 16 to 470 nm in 48 predefined classes. From the size distributions, PNC were calculated.

Traffic counts and meteorological data
Automated traffic counts were done at the street locations for one week in November 2008 by making use of a standard Minuteman EVR (Counters & Accessories Ltd, England), which is a two-tube counter. Apart from total traffic counts, speed and classification (light, middle and heavy) of the motorised vehicles has been registered as well.
Data on hourly standard meteorological parameters were obtained from the Royal Dutch Meteorological Institute, the Bilt located in the middle of the Netherlands (<5 km from Utrecht), to characterise the weather conditions during the sampling weeks.

Data management and analysis
To limit potential bias in the comparison between locations due to temporal variation, an adjustment was made using data from the continuous reference site[15]. Correlation of the standard air pollutants was high between all sites; therefore the critical assumption that temporal patterns are correctly reflected at the reference site seems justified. This adjustment was not made

for elemental concentrations, as correlation in time between sites was typically quite low (R<0.8) for most elements. Comparison of absolute elemental concentrations between streets can only be made for sites in the same sample round.
For NO_2 and NO all samples were available for analysis. Some PM measurements were lost for various technical reasons, leaving 209 (92%) filters available for analysis. Most missing samples were at the reference location (N=7). In the absence of available fixed site monitors from the National Air Quality Monitoring Network in the neighbourhood, we have estimated missing PM reference values with the use of the nearest background site. We have made a regression line per pollutant and calculated predicted concentrations. Correlation was high between those sites (R>0.96).
Eighteen and 16 paired measurement days were available for analysis, after checking the SMPS data. Missing data was due to power supply problems, site-related problems and initial instrumental problems (cracked electrical isolator in classifier of SMPS).
Descriptive statistics were calculated. Median ratios between individual simultaneously measured samples were calculated to compare streets to the matching urban and suburban sites. The median is less influenced by outlier values. The overall median ratio was tested on significance by the use of the non-parametric signed rank test.

Data quality

Limits of detections
Limit of detection (LOD), calculated as three times the SD of all field blanks divided by the nominal sampling volume, was 3.5 $\mu g/m^3$ for PM_{10}, 0.5 $\mu g/m^3$ for $PM_{2.5}$ concentrations and 0.2 $10^{-5}/m$ for soot. None of the filters were below LOD. The LODs were 3.6 and 10.1 $\mu g/m^3$ for NO_2 and NO_x respectively, after exclusion of one very high field blank (NO_2: 14.4 $\mu g/m^3$). All NO_2 measurements were above the LOD. All but one NO_x measurements were above LOD, after exclusion of one high field blank. Supplement Table 1 reported the LODs for the various elements. All reported PM_{10} elemental concentrations were above LOD apart from some samples of Al (85%>LOD), Ba (72%>LOD) and V (99%>LOD). For the $PM_{2.5}$ samples percentages detected were somewhat lower ranging from 67% for Cl to 100% for Fe, Pb and S. Values below the LOD were retained rather than replaced by e.g. 0.5*LOD.

Precision and comparison with fixed site monitors
Precision expressed as coefficients of variability (CV) from duplicate measurements was good: PM_{10} (CV=6%), $PM_{2.5}$ and soot (CV both 7%), NO_2 (4%) and NO_x (10%). We found somewhat higher PM_{10} (mean difference: 14%) and NO_2 (mean difference: 8%) urban background concentrations then measured by the fixed site monitor in Amsterdam, running on exactly the same location. Correlations were high (R>0.93). NO_2 concentrations were somewhat lower

(4%) on the street location, compared to another fixed site monitor in Amsterdam, measured at the same street but closer to the kerb.
Supplement Table 2 showed CVs between replicate measurements for different elements. For the laboratory replicates, CVs were under the 10% for Ca, Cu, Fe, S, Si, and Zn for both fractions. Pb concentrations of the $PM_{2.5}$ fraction are not reported due to the large CV. For the PM_{10} field replicate measurements, CVs were below the 12.5% for Al, Ba, Ca, Cu, Fe, Mn, S, Si, and Zn. Ni had one unexplained outlier field replicate measurement. The CVs for $PM_{2.5}$ replicates were mostly somewhat larger.

Results and discussion

Concentration differences between locations

PM mass, soot and NO_x

Averages of PM_{10}, $PM_{2.5}$, soot, NO_2 and NO_x concentrations for the different street locations are given in Table 2. Substantial contrast was found between the moderately busy streets and the respective urban background locations, especially for soot (factor 1.9) and NO_x (factor 1.8). A small contrast (factor 1.2) was found for PM_{10} and $PM_{2.5}$, which is in line with previous studies[3-6,21]. The contrast for NO_2 was in between PM and soot (factor 1.5). Table 2 further illustrates that the ratio between busy street and respective urban background varies substantially between streets, especially for soot. For soot ratios above 2.0 are found for the two canyon streets (Stille Veerkade and Brugstraat) and the two streets which are built on one side of the street only (Haarlemmerweg, Weerdsingel Wz).

Table 2 Averages of different traffic-related air pollution concentrations (μg/m^3) at the various streets and median ratio street vs. urban background location in the same city

Urban streets	PM_{10}	$PM_{2.5}$	Soot[a]	NO_2	NO_x
Haarlemmerweg	27.5	17.8	3.9	54.3	100.5
	1.2	1.2	2.1	1.5	2.0
Hoofdweg	22.4	15.1	2.6	47.5	75.6
	1.1	1.0	1.7	1.3	1.5
Stille Veerkade	32.2	19.4	4.3	54.0	109.9
	1.3	1.2	2.7	1.7	2.1
Brugstraat	32.9	18.0	3.7	53.8	99.3
	1.3	1.3	2.1	2.0	2.8
Koningsweg	30.2	17.4	2.8	44.2	78.3
	1.1	1.2	1.9	1.6	2.1
HVB	29.4	17.2	2.4	36.6	60.6
	1.1	1.1	1.5	1.3	1.4
Vleutenseweg	25.9	15.4	2.1	41.7	70.3
	1.0	1.1	1.5	1.2	1.6
Weerdsingel Wz	29.1	16.6	3.4	52.4	87.7
	1.2	1.1	2.2	1.5	2.0
Overall ratio[b]	1.2	1.2	1.9	1.5	1.8

[a]Soot expressed in 10^{-5}/m; [b]Overall ratio calculated as the median ratio street vs. urban background location for the individual samples (N PM_{10} ratio=45, N $PM_{2.5}$ ratio=40, N soot ratio=45, N NO ratio=48). Overall ratio tested on significance with the use of the non-parametric signed rank test, and all p-values were below 0.0001.

Elemental composition

Substantial differences between streets were found in the elemental composition of PM_{10} (Table 3). Especially, Cr, Cu and Fe were clearly elevated at all street locations (overall ratio with urban background >2.0). The highest ratios were again observed at the canyon and one-side built streets. Especially Cu differed substantially from a factor 1.5 at the Vleutenseweg to a factor 5.1 at the Haarlemmerweg, both compared to background levels. The contrast of these three elements was much larger than found for Al, Si and Ti, frequently used indicators of soil dust, suggesting a traffic source.

In the $PM_{2.5}$ fraction, only the elements Cu and Fe were consistently and highly elevated at all streets (Table 4). Cr and Ba were not detected sufficiently to be reported neither was Pb (high CV). Mn, Si and Ti are also somewhat higher at most street locations. No consistent difference was found for the other elemental concentrations. The increased concentration ratios for Cu and Fe could be explained as the tail of the coarse particle fraction sampled by the $PM_{2.5}$ cyclone. Alternatively, it could reflect direct tailpipe emission of $PM_{2.5}$. Elemental concentrations in $PM_{2.5}$ samples were much smaller than in the PM_{10} samples, with the exception of Ni, S and V, elements typically associated with the $PM_{2.5}$ fraction.

The contrasts in Fe and Cu were even more pronounced when compared to the simultaneously measured suburban locations, especially in the PM_{10} fraction. Cu at the street locations ranged from a factor 2.1 to 6.5 compared to the matching suburban location (Supplement Tables 3-5).

Table 3 Average PM_{10} elemental concentrations (ng/m^3) at the various streets and median ratio street vs. urban background location in the same city

Urban streets	Round	Al	Ba	Br	Ca	Cl	Cr	Cu	Fe	Mn	Ni	Pb	S	Si	Ti	V	Zn
Haarlemmerweg	A	99.2	17.1	6.6	435.8	1017.3	7.9	63.2	1316.0	18.4	2.9	11.0	1753.3	448.8	21.8	3.4	52.3
		1.3	0.8	1.1	1.3	1.3	3.5	5.1	3.4	1.7	1.2	0.6	1.1	1.2	1.1	0.7	1.4
Hoofdweg	A	71.3	8.9	7.3	365.0	1414.6	4.7	28.4	664.9	11.2	2.8	10.6	1535.4	369.0	21.6	3.6	43.1
		1.3	0.5	1.1	1.2	1.1	2.0	2.4	1.8	1.2	1.2	0.8	1.0	1.3	1.1	0.9	1.3
Stille Veerkade	A	89.8	14.5	9.9	475.4	2148.2	8.5	56.5	1332.4	20.3	5.0	13.5	1841.8	533.7	27.0	6.1	51.6
		2.6	1.3	1.1	1.5	1.5	2.9	3.6	2.8	2.5	1.2	1.4	1.2	1.7	1.7	1.0	1.7
Brugstraat	B	52.3	14.8	6.8	276.3	1553.5	8.7	47.6	1072.4	15.1	3.3	12.6	1153.3	338.9	11.8	3.4	55.1
		3.2	3.8	1.2	1.3	1.2	2.4	3.6	2.9	1.8	1.4	1.1	1.1	1.5	1.3	1.0	1.3
Koningsweg	B	47.4	10.2	6.0	587.3	1104.3	5.1	35.6	908.0	15.1	2.4	11.3	1210.9	423.6	15.1	3.7	67.1
		1.7	2.1	1.1	1.0	1.0	1.5	2.1	2.0	1.4	1.2	1.0	1.1	1.8	1.4	0.9	1.3
HVB	B	42.1	9.1	7.3	354.2	1208.9	5.3	33.1	734.0	12.6	3.3	16.4	1266.8	319.4	14.4	4.9	50.2
		1.1	1.2	1.1	1.2	1.1	2.2	2.6	2.6	1.5	1.1	1.3	1.1	1.3	1.5	1.1	1.3
Vleutenseweg	B	23.9	5.3	5.5	311.0	1081.9	3.7	18.8	439.2	10.7	2.6	7.6	986.5	260.8	12.2	3.8	37.4
		1.0	0.6	0.9	1.1	1.1	1.3	1.5	1.1	1.0	0.9	1.0	1.0	1.2	1.0	0.9	1.0
Weerdsingel Wz	B	33.9	9.4	6.1	331.3	1432.9	5.7	40.7	997.7	15.5	3.3	7.7	1058.0	309.3	15.4	3.8	46.0
		1.2	0.9	1.0	1.1	1.1	2.3	3.1	2.7	1.6	1.1	0.8	1.0	1.4	1.1	0.9	1.1
Overall ratio[a]		1.4	1.2	1.1	1.2	1.2	2.2	2.8	2.5	1.5	1.2	1.1	1.1	1.3	1.3	0.9	1.4
P-value[b]		0.01	0.45	0.04	0.00	0.00	0.00	0.00	0.00	0.00	0.07	0.14	0.02	0.00	0.04	0.07	0.00

[a]Overall ratio calculated as the median ratio street vs. urban background location for the individual samples (N ratio=45); [b]Ratio tested on significance with the use of the non-parametric signed rank test.

Table 4 Average $PM_{2.5}$ elemental concentrations (ng/m^3) at the various streets and median ratio street vs. urban background location in the same city

Urban streets	Round	Al	Br	Ca	Cl	Cu	Fe	Mn	Ni	S	Si	Ti	V	Zn
Haarlemmerweg	A	29.3	4.2	40.4	159.2	9.1	200.1	4.0	1.8	1306.7	86.6	3.4	2.7	24.6
		0.8	1.0	1.3	1.3	3.2	2.2	1.6	1.2	1.0	1.1	0.8	1.1	1.3
Hoofdweg	A	23.8	4.3	36.3	194.7	4.5	116.8	2.4	1.3	1377.9	146.3	3.2	1.9	30.9
		1.1	1.0	1.2	1.1	1.8	1.5	1.0	1.0	1.0	1.8	0.7	0.8	1.1
Stille Veerkade	A	15.1	4.7	36.8	212.7	7.4	159.2	4.0	2.3	1426.6	60.4	2.6	3.7	24.0
		0.9	0.9	1.6	1.3	1.8	1.6	1.3	0.9	1.0	1.2	1.7	1.0	1.2
Brugstraat	B	17.6	2.8	33.4	88.7	5.2	140.4	3.3	1.3	831.1	105.0	1.1	1.7	28.2
		1.3	0.6	1.2	1.0	2.4	1.9	2.0	0.6	1.0	1.4	0.6	0.6	1.1
Koningsweg	B	16.7	2.9	53.4	62.0	6.9	153.0	3.0	1.1	872.8	116.8	1.6	2.4	35.5
		1.4	0.8	1.5	1.0	2.6	1.7	2.1	0.8	1.0	1.5	0.9	1.1	1.3
HVB	B	11.3	3.6	33.2	108.2	5.0	125.7	2.0	1.8	888.8	44.9	0.7	2.8	22.8
		0.9	0.8	0.9	0.9	2.3	2.0	1.2	1.4	1.0	1.1	0.0	1.2	1.0
Vleutenseweg	B	13.9	3.6	37.4	58.2	3.1	97.4	2.7	1.7	765.9	52.5	2.0	2.8	18.2
		1.0	1.3	0.9	1.0	1.4	1.3	1.0	1.0	1.1	1.7	0.9	1.1	1.6
Weerdsingel Wz	B	10.1	5.4	33.4	63.9	5.5	142.2	3.1	1.4	764.4	58.0	1.7	2.5	20.4
		1.1	1.3	1.0	1.0	2.8	2.0	1.6	0.9	1.0	1.4	2.0	1.1	1.5
Overall ratio[a]		1.0	1.0	1.1	1.0	2.3	1.9	1.4	1.0	1.0	1.2	0.8	1.0	1.1
P-value[b]		0.72	0.43	0.15	0.46	0.00	0.00	0.00	0.87	0.64	0.00	0.08	0.87	0.01

[a]Overall ratio calculated as the median ratio street vs. urban background location for the individual samples (N ratio=40); [b]Ratio tested on significance with the use of the non-parametric signed rank test.

Particle size distributions

PNC and particle size distributions, measured in two streets were substantially different from the nearby suburban location (Table 5, Figure 2). PNC was more elevated at the one-sided built street location (Haarlemmerweg: factor 2.4), than at the wider, two-sided built street location (Vleutenseweg: factor 1.3), consistent with the contrast for other components. At the Haarlemmerweg, especially PNC in the 30-100 nm range were significantly elevated (factor 3.2), the typical size range for particles freshly emitted by motorized traffic. Particles at the Haarlemmerweg were generally peaking at 40-50 nm whereas at the suburban location the peak was at a somewhat larger particle diameter (70-80 nm). At the Vleutenseweg and the suburban location, most particles were in the 60 to 70 nm range (Figure 2).

Table 5 Average PNC (p/cm^3) at two streets and median ratio street vs. suburban background location[a]

Urban streets	N	16-30 nm	30-100 nm	100-470 nm	Total
Haarlemmerweg	18	1257	10 727	4207	16 191
		0.8 (0.48)	3.2 (0.00)	2.3 (0.00)	2.4 (0.00)
Vleutenseweg	16	1540	6365	2538	10 443
		1.6 (0.02)	1.4 (0.00)	1.4 (0.08)	1.3 (0.00)

[a]Ratio tested on significance with the use of the non-parametric signed rank test, p-value in brackets.

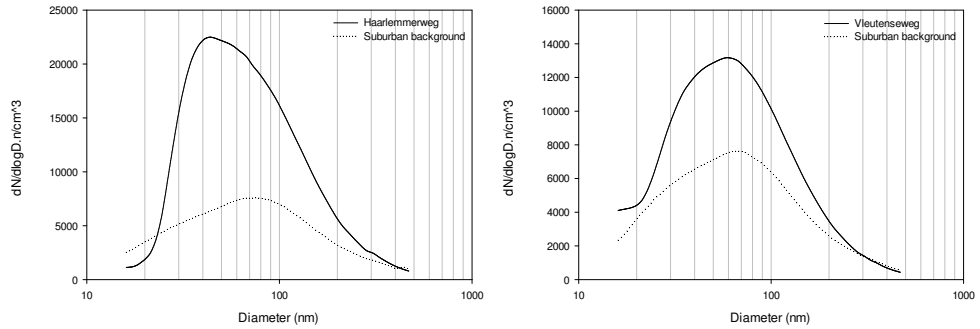

Figure 2 Particle size distribution measured by SMPS at two street locations and the respective suburban background locations. Note that the scale is different

Interpretation of contrasts in concentration of the various pollutants

NO_2 vs. soot, PNC and elemental composition

The contrast in NO_2 between busy roads and background was smaller than the contrasts for soot, PNC and elements indicative of non-tailpipe emissions such as Cr, Cu and Fe. NO_2 has often been used as an indicator for traffic-related air pollution in epidemiological studies and traffic management evaluations[7]. NO_2 is widely used because an air quality guideline exists and it is easy to measure with passive samplers. Some indicative evidence that NO_2 is not representing (current) traffic well comes from a few studies that documents stagnant NO_2 concentrations over time, despite considerable reductions in NO_x concentrations[22,23]. We were not able to distinguish between primary (directly emitted) and secondary NO_2 (formed secondary by NO and O_3). Emission of primary NO_2 is rising due to an increase in diesel cars and their particulate filters in European countries. Our data add to the evidence that also with NO_2, traffic-related contrasts are underestimated. The reported NO_2 contrast may differ from country to country due to the difference in car park, e.g. in the fraction of diesel vehicles emitting higher fractions of primary NO_2.

Elemental composition and non-tailpipe emissions

The contrasts in Cu, Fe and Cr were at least as large as that in PNC and soot. Cu at traffic sites originated mostly from non-tailpipe emissions, in particular from brake wear, together with Sb and Ba[11,24,25]. The contribution of non-tailpipe emissions in PM mass has been recently highlighted[26]. Because of substantial efforts to reduce exhaust emissions, non-exhaust emissions have become more important. Non-exhaust particles typically arise from abrasive sources which include brake wear, tyre wear, road surface abrasion and re-suspension in the wake of passing traffic. Fe has often been associated with brake wear and re-suspension of road dust as well[26]. In addition, Fe could originate from direct tailpipe emissions. The elevated concentrations of Cr at the streets are not totally understood, although Cr may reflect re-suspended road dust or brake lining as well[27]. In addition, Cr can originate from a variety of other (industrial) sources. However, our results suggests a common traffic source as Cr was highly correlated ($R>0.8$) with more traffic-related indicators like NO_x, NO_2, soot, and Cu and Fe. Mn is also higher at all street locations, but to a somewhat lesser extent (factor 1.5). We found only very modestly increased Zn concentrations at the street sites (factor 1.4). Zn may derive from tyre wear, but has multiple atmospheric sources[26], which may have contributed to the relatively small contrast in our study. In none of the cities, large industrial sources were present of the elements reported here. Recent measurements in London also did not show a large contrast between a major road and urban background for Zn[11].

Typically, most elements are somewhat lower as has been found in other recent studies investigating busier traffic roads[10,11,28,29]. Hence sampling was often much closer to the road as well. Comparison of absolute concentrations

is limited given the large differences in sample location, sampling period and time, and monitoring equipment.
Concentration contrasts were also found in the $PM_{2.5}$ fraction, in line with previous studies. Götschi et al.[30] found substantially elevated concentrations of Ca, Cu, Fe, Mn, Ti and soot in $PM_{2.5}$ particles at an Antwerp busy street location, compared to a background site (factor 1.4-1.9). Hueglin et al.[31] reported large contrasts in Fe, Cu and Sb (factor 7-9) between a busy street in Bern and a suburban site in Basel in the PM_{10} fraction. In the $PM_{2.5}$ fraction those contrasts lowered considerably[31]. Johansson et al.[10] reported much higher ratios than in the present study. The largest difference was found for Cu and Sb in the PM_{10} fraction, with on average more than 7 times higher levels in a street canyon compared to an urban background location on a rooftop in Stockholm. Cr was elevated with about a factor 2-3, comparable with our study. Fe was not reported in that study[10]. Harrison et al.[4] reported at roadside sites a strong elevation in EC, organic compounds and Fe-rich dust in comparison to the simultaneously collected urban background samples. Gietl et al.[11] showed that in particular Fe, Cu, Ba and Sb were elevated at a London roadside compared to background locations (factor 5-6).

Particle number contrasts
The contrast in PNC at the two streets where PNC was measured was slightly smaller than observed for soot and concentrations of Cu and Fe (Supplement Tables 3-5). An additional study at the same street in Utrecht (Weerdsingel Wz) also showed slightly smaller contrasts for PNC than for soot[32]. PNC and soot were both more than 3 times higher at the street location compared to the suburban location. Differences between the suburban and urban background location (both the same as in present study) were much smaller and statistically non-significant[32]. Some studies[33] have shown that the spatial gradient near motorways is largest for the nucleation mode particles (<10 nm). The relatively low contrast in the smallest particle range in our study might be the result of coagulation of the smallest particles with the larger chainlike particles. In addition, the instrument collected particles from 16 nm, hence the particles smaller than 16 nm were not sampled. The larger agglomerated particles (with a diameter of some hundreds of nm) have a relatively large surface where nucleation aerosol can easily deposit on by diffusion. Because there is much more of that type of aerosol in street locations, the deposition by diffusion might be enhanced at these locations with high aerosol concentrations.
Apart from the PM composition, other PM characteristics like particle size and surface area may determine the health risks posed by PM as well. Ultrafine particles might play an important role in this association, both because of the large PNC contrast and a rapid decline with increasing distance from the motorway[34]. However, most studies of PNC near major roads were conducted near or close to motorways up to 300 000 motorised vehicles per 24-hour[34-36]. There is much less information about PNC concentration contrasts for less busy urban roads[21,37], to which a significant amount of urban inhabitants are

exposed. The latter studies suggested a modest contrast in PNC at urban streets as well, comparable with soot or NO_x contrasts.

Factors influencing street concentrations

The observation that contrasts with urban background differed substantially between the eight traffic sites is probably explained by the poorer dilution in street canyons; the larger fraction of wind from the road to the sampling location in streets with only one side of the road built and a smaller distance to the road. Traffic intensities were quite similar for all streets apart from the Hoofdweg showing lesser traffic (Table 1). The average speed was also lowest at the canyon streets and the Weerdsingel Wz. The average speed in rush hours dropped significantly, and hardly any difference was shown between average daytime and rush hour speed, consistent with the observed continuous amount of congestion and stop-and-go traffic during daytime. Stop-and-go traffic increases emissions; especially (hard) acceleration contributes to this. Acceleration reduces temporarily the efficiency of catalytic converters[38].

In the sampling period, average temperature was about 8°C (2-18°C). Wind direction was predominantly southwest (5 times), and mean wind speed was 3.6 m/s (2.5-4.6 m/s). Especially, wind direction plays an important role in the large variation between measurements. The hourly weather data from the nearby station de Bilt could not explain the observed contrasts between different streets. Field observations suggested that especially with winds parallel to the street limited contributions of traffic were found. We do not see different patterns of behaviour between different street types. Especially canyon type of streets is known for their enclosed nature leading to a de-coupling of the dispersion within the street canyon from those above the building canopy that determine the background concentrations[39]. At the two streets with adjoining buildings at one side, the wind was never coming from that side. With that wind, contrasts are expected to be even greater due to recirculation and entrapment in the lee of the buildings[39]. There was no information available about subtle weather conditions at the specific locations.

Limitations

A limitation of our sampling schedule with discontinuous measurements spread over a half year is that the absolute concentrations can deviate from the annual average. However, efficient and valid estimates of the spatial contrast between locations can be obtained[15]. We further could not afford to perform particle size monitoring on more than two of the eight streets. Sampling height was generally the same for all but two background locations. Exact sampling height was slightly different between the streets in Utrecht (4-5 m) and the suburban background location (1 m). This was also the case for the Stille Veerkade and the matching urban background location. The reported contrast between those street and the background sites was

probably not substantially affected by this difference in sampling height, as in the well-mixed urban atmosphere, vertical gradients are generally small for sampling sites away from local sources such as major roads. In contrast, near major roads large vertical gradients have been identified[40].

The background locations were chosen away from busy streets (>50 m). Exact distance to nearest major roads at background locations was different. Distance to nearest major road was smallest at the urban background sites for Amsterdam (50 m, 17 000 motorised vehicles per 24-hour), Den Bosch and Utrecht (70 and 100 m, both 16 000 motorised vehicles per 24-hour). For the suburban background locations, distance to nearest major road was somewhat further away (between 100-300 m). Difference in distance to nearest major road has unlikely had more than a minor influence on the reported contrasts, as all the locations were in residential areas surrounded by other buildings.

We measured only the ambient concentrations. Since people spend a large percentage of their time indoors, infiltration into indoor air is important as well for actual exposure[41]. Various studies have investigated indoor/outdoor relationships and identified important determinants such as air exchange rate, building characteristics, filtration and particle size distribution and composition[14,41,42]. Infiltration rates of the different components are different. For example components associated with $PM_{2.5}$ penetrated well into indoor environments with typical percentages of 70-80%, although highly depending on ventilation status. The infiltration of ultrafine particles is associated with higher agglomeration, with more agglomeration reported for the nucleation mode particles compared to particles in the larger end of the ultrafine particle range[43].

Conclusion

Simultaneous measurements of an extensive set of air pollutants at eight moderately busy urban streets and ten urban and suburban background locations, showed that the contrast between busy streets and urban background in NO_2 was substantially less than the contrast found for soot, PNC and elements indicative of non-exhaust emissions, adding to indicative evidence that NO_2 is not representing (current) traffic well. The quantitative contrast for the elements Cr, Cu and Fe (factor 2.2 to 2.8) was even larger than the contrast found for soot and NO_x (factor 1.8), typically indicators of direct combustion emissions. This supports a substantial role for non-exhaust emissions, with brake wear, tyre wear and road dust as the potentially important components. The contrast in PNC measured in two streets was similar to soot. The variability in contrast between streets was as large as that between streets and background locations, with the largest contrast found for two street canyons and two streets with buildings at one side of the street only. Several of the components showing a large contrast have documented health effects, such as soot, PNC and transition metals like Cu and Fe, and may be associated ultimately with various adverse health effects, in addition to PM_{10} effects. Significant underestimation of disease burden may occur when relying too much on the regulated components.

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Supplement

Supplement Table 1 Limits of detection (LOD) and field blanks[a] of the various elemental concentrations (ng/m^3)

	Al	Ba	Br	Ca	Cl	Cr	Cu	Fe	Mn	Ni	Pb	S	Si	Ti	V	Zn
Average PM_{10} blanks	0.0	1.8	0.2	5.0	3.0	0.1	0.3	2.3	0.1	0.1	0.2	1.3	31.3	0.5	0.0	2.9
PM_{10} LOD (3*SD)	0.0	3.7	0.8	5.7	10.9	0.5	0.8	3.1	0.7	0.4	0.9	7.2	63.5	1.4	0.1	8.8
Average $PM_{2.5}$ blanks	0.9	0.9	0.2	10.1	11.3	0.8	2.1	4.0	0.6	0.3	0.0	3.5	17.9	0.7	0.2	5.3
$PM_{2.5}$ LOD (3*SD)	6.4	5.3	0.7	40.5[b]	54.5[c]	1.7	3.6	8.0	1.0	0.7	0.0	5.3	52.2	1.6	0.8	14.9
Laboratory LOD	2.5	1.2	0.4	0.7	2.7	0.5	0.7	0.8	0.6	0.5	0.7	0.9	1.8	0.5	0.4	0.9
% PM_{10} filters >LOD	85	72	100	100	100	100	100	100	100	100	100	100	100	100	99	100
% $PM_{2.5}$ filters >LOD	69	24	99	55	52	43	80	100	99	90	100	100	74	82	87	86

[a]There were 6 field blanks for both PM_{10} and $PM_{2.5}$; [b]The $PM_{2.5}$ LOD was high because of one outlier field blank, otherwise 100%; [c]The $PM_{2.5}$ LOD was high because of one outlier field blank, otherwise 67%.

Supplement Table 2 Coefficients of variability[a] of the various elemental concentrations

	PM_{10} replicates		$PM_{2.5}$ replicates	
	Laboratory (N=11)	**Field (N=10)**	**Laboratory (N=7)**	**Field (N=9)**
Al	8.6 (8)	9.4 (7)	14.3	40.9 (8)
Ba	24.3 (6)	7.5 (4)	ND[b]	ND[b]
Br	6.5	14.9	34.0	13.2
Ca	1.4	7.6	6.8 (4)	9.9 (4)
Cl	6.5	16.4	12.4 (1)	29.3 (5)
Cr	6.5	25.3	ND[b]	ND[b]
Cu	3.9	11.1	9.8 (6)	12.7 (6)
Fe	1.5	7.7	2.0	16.6
Mn	7.9	7.9	22.1 (6)	16.6
Ni	11.6	302.3[c]	11.5 (2)	23.6 (6)
Pb	15.8	22.6	61.1	20.5
S	0.5	7.1	0.8	9.1
Si	1.8 (10)	8.8	2.9 (4)	18.6
Ti	8.7	23.2	32.2 (6)	18.6
V	7.9	12.9	12.2 (5)	19.2 (7)
Zn	3.8	12.5	4.4 (5)	14.1

[a]Coefficients of variability (CV) have been calculated as the square of the absolute differences of the measurements. Then the root of the sum of the values is divided by two times the number of duplicate measurements. CV expressed in %; [b]ND=not sufficiently detectable; [c]CV extremely high because of one outlier duplicate, otherwise CV=20.6.

Supplement Table 3 Median ratio street vs. suburban background location in the same city for different traffic-related air pollutants

Urban streets	**PM_{10}**	**$PM_{2.5}$**	**Soot**	**NO_2**	**NO_x**
Haarlemmerweg	1.3	1.4	2.1	2.0	2.7
Hoofdweg	1.3	1.1	1.8	1.8	2.0
Stille Veerkade	1.4	1.4	2.2	2.1	2.4
Brugstraat	1.2	1.2	2.4	2.4	3.3
Koningsweg	1.2	1.2	2.0	2.1	2.6
HVB	1.1	1.2	1.7	1.7	1.9
Vleutenseweg	1.3	1.0	1.7	1.5	1.7
Weerdsingel Wz	1.5	1.4	3.0	1.8	2.1
Overall ratio[a]	1.3	1.2	2.1	1.9	2.3

[a]Overall ratio calculated as the median ratio street vs. suburban background location for the individual samples (N PM_{10}, $PM_{2.5}$ and soot ratio=44, N NO ratio=48). Overall ratio tested on significance by the use of the non-parametric signed rank test, and all p-values were below 0.0001.

Supplement Table 4 Median ratio street vs. suburban background location for various elemental concentrations (PM_{10} fraction)

Urban streets	Al	Ba	Br	Ca	Cl	Cr	Cu	Fe	Mn	Ni	Pb	S	Si	Ti	V	Zn
Haarlemmerweg	2.8	4.8	1.2	2.0	2.0	3.7	6.0	4.8	2.5	1.4	1.1	1.1	2.0	2.3	1.3	1.5
Hoofdweg	2.9	1.7	1.3	2.0	2.0	2.3	2.8	2.4	1.5	1.6	1.0	1.0	2.1	2.2	1.4	1.3
Stille Veerkade	2.3	2.7	1.1	2.2	2.2	2.7	5.0	4.3	2.1	1.9	1.2	1.2	2.2	2.2	1.3	1.6
Brugstraat	2.0	ND[c]	1.1	1.2	1.2	3.2	6.5	5.8	2.4	1.2	1.1	1.1	1.5	1.1	1.0	1.1
Koningsweg	1.8	ND[c]	1.0	1.1	1.1	2.0	4.7	3.6	1.7	0.9	0.9	1.1	1.3	1.1	0.9	1.2
HVB	1.2	0.2	1.1	1.2	1.3	2.2	3.9	2.7	1.4	1.2	1.2	1.2	1.1	1.3	1.3	1.1
Vleutenseweg	2.1	1.8	1.0	1.5	1.2	1.3	2.1	1.7	1.5	1.1	1.0	1.1	1.4	1.5	1.1	1.4
Weerdsingel Wz	2.0	2.3	1.2	1.6	1.4	2.4	4.2	4.0	2.3	1.4	1.1	1.1	1.8	2.0	1.0	1.8
Overall ratio[a]	2.3	2.2	1.1	1.6	1.4	2.5	4.1	3.3	1.8	1.3	1.1	1.1	1.6	1.5	1.1	1.4
P-value[b]	0.00	0.10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.45	0.00	0.00	0.00	0.29	0.00

[a]Overall ratio calculated as the median ratio street vs. suburban background location for the individual samples (N ratio=44); [b]Ratio tested on significance with the use of the non-parametric signed rank test; [c]ND=not sufficiently detectable.

Supplement Table 5 Median ratio street vs. suburban background location for various elemental concentrations ($PM_{2.5}$ fraction)

Urban streets	Al	Br	Ca	Cl	Cu	Fe	Mn	Ni	S	Si	Ti	V	Zn
Haarlemmerweg	1.2	1.1	2.2	1.0	5.9	3.0	1.8	3.2	1.1	1.5	1.2	2.0	1.2
Hoofdweg	1.0	1.0	2.2	1.0	2.7	1.6	1.1	1.4	1.0	1.2	1.2	2.5	1.1
Stille Veerkade	0.9	1.3	1.5	1.1	2.6	2.3	2.1	1.8	1.2	1.1	0.7	1.7	1.2
Brugstraat	1.2	1.0	1.2	1.0	2.1	3.1	1.9	0.9	1.1	1.7	0.9	0.8	1.4
Koningsweg	1.0	1.0	2.0	0.9	2.4	2.9	2.2	1.0	1.1	1.6	1.2	1.0	1.4
HVB	1.0	1.4	1.4	1.1	1.4	2.7	1.6	1.9	1.1	0.9	0.3	1.2	1.0
Vleutenseweg	1.1	1.1	1.1	0.9	0.8	1.3	1.4	1.1	1.0	1.3	0.9	1.0	1.2
Weerdsingel Wz	1.0	1.9	1.4	1.0	2.7	2.5	1.3	1.1	1.0	1.3	1.0	1.1	1.8
Overall ratio[a]	1.0	1.1	1.5	1.0	2.5	2.7	1.6	1.3	1.1	1.2	1.0	1.2	1.2
P-value[b]	0.28	0.10	0.00	0.86	0.02	0.00	0.00	0.10	0.00	0.02	0.88	0.05	0.01

[a]Overall ratio calculated as the median ratio street vs. suburban background location for the individual samples (N ratio=44); [b]Ratio tested on significance with the use of the non-parametric signed rank test.

Chapter 3

Comparison of short-term exposure to particle number, PM_{10} and soot concentrations on three (sub) urban locations

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Abstract

Background
Recent interest has focused on the health effects of ultrafine particles (UFP) because of the documented toxicity and the larger concentration contrast near motorways of UFP than for PM_{10} or $PM_{2.5}$. There are only few studies that have measured UFP at inner-city streets simultaneously with other PM components. The aim of this study was to compare the contrast of UFP, PM_{10} and soot measured simultaneously at three inner-city locations, namely a moderately busy street (15 000 vehicles per 24-hour), an urban and a suburban background location.

Methods
Simultaneously, measurements of UFP characterised by particle number concentrations (PNC), PM_{10} and soot have been conducted on three locations in and around Utrecht, a medium-sized city in the Netherlands for twenty weekdays in autumn 2008. Measurements were done for six hours during afternoon and early evening.

Results
The mean PNC at the street location was more than 3 times higher than at the two background locations. The contrast was similar for soot concentrations. In PM_{10} concentrations less contrast was found, namely 1.8 times. Average PNC concentrations were poorly correlated with PM_{10} and soot. At the street location, high temporal variation of PNC concentrations occurred within each sampling day, probably related to variations in traffic volumes, high-emission individual vehicles and wind direction. Temporal variation was smaller at the two background locations. Occasional unexplained short-term peaks occurred at the suburban background location. A relatively high correlation between PNC minute values at the two background locations was found, pointing to similar area-wide sources. Typically low correlations were found at the street location, consistent with the dominant local impact of traffic.

Conclusion
A large contrast between two background locations and a moderately busy urban street location was found for PNC and soot, comparable to previous studies of much busier motorways. Temporal variation of PNC was higher at the street location and uncorrelated with background variations.

Introduction

Numerous studies have documented the effects of particulate matter (PM) air pollution on morbidity and mortality from respiratory and cardiovascular diseases[1,2]. Most epidemiological studies have characterised particulate matter air pollution as the mass of particles smaller than 10 µm (PM_{10}) or 2.5 µm ($PM_{2.5}$). PM however is a complex mixture of ultrafine, fine and coarse particles from a variety of sources. There is also an extensive literature that has associated living in close proximity to major roads or traffic density on the street of residence with various health effects including respiratory morbidity[3,4], subclinical markers of atherosclerosis[5] and premature mortality from cardio-respiratory diseases[6,7]. In those studies, little quantitative information is available on differences in traffic-related air pollution concentrations related to proximity to traffic.

One of the components that may be responsible for the observed health effects related to proximity to motorised traffic is ultrafine particles (UFP) because of the documented toxicity[8,9]. A recent expert panel confirmed the potential adverse health effects of UFP[10,11]. In the documentation for the expert panel, the near-roadway studies were used to derive the potential long-term health effects of UFP, based upon the large concentration contrast near major roads[10]. Most studies of UFP near major roads were conducted close to motorways with up to 300 000 motorised vehicles per 24-hour[9,12,13]. In those studies UFP is characterised by particle number concentration (PNC). Zhu et al.[9,14] have shown a large contrast with a rapid decrease in PNC with increasing distance from the motorway. Weijers et al.[15] also found a decrease in PNC as a function of distance from the road. Harrison et al.[12] have measured PNC and PM_{10} simultaneously in Birmingham, UK. While PM_{10} concentrations did not vary much between the three urban sites (20–24 µg/m^3), PNC was around 3 times higher at the site influenced by a motorway 70 m away with 160 000 motorised vehicles passing every 24-hour than at the other two background locations. Other studies also have showed a large PNC contrast and a rapid decline with increasing distance from the motorway[16,19]. Hagler et al.[18] calculated an approximately 5-12% average drop in PNC per 10 m distance from the roadway, based on six studies.

There is much less information about PNC concentration contrasts for less busy urban roads[20,21], to which a significant amount of urban inhabitants are exposed. In a study of Aalto et al.[20] PNC at traffic sites was between 2 and 6 times higher than the concentration at the corresponding urban background sites in Rome, Barcelona and Stockholm, depending on the distance to the road, traffic volume, road configuration and sampling height. In a study in four European cities, residences located less then 10 m from a major street experienced on average more than 2 times higher PNC than residences located further away[21]. In a study in Rome[22] the PNC concentration at a traffic-oriented site was about a factor 2 higher than at the urban background location.

For the interpretation of the near-roadway studies it is also important to compare the PNC contrast of moderately busy urban streets with those of other potentially important components of traffic emissions such as PM_{10} or soot. Several studies have simultaneously measured PM air pollutants in urban streets with high and low traffic intensities[9,12,23-25].
The aim of this study is to compare the concentration contrast of three different PM components (UFP, PM_{10} and soot) of a moderately busy urban street with urban and suburban background locations. The focus is on comparing average concentrations, but we were also interested in the contrast in occurrence of short-term peaks of UFP as health effects of highly variable concentrations and more constant concentrations may differ.

Methods

Study design

Simultaneously, detailed measurements of PM_{10}, soot and PNC have been measured on three locations for 20 weekdays (excluding Fridays) from Oct. 06-Nov. 06 in 2008. Measurements were done for six hours, from 12.30 to 18.30 PM, including evening rush hour. Three locations were selected in and around Utrecht, a medium-sized city in the Netherlands. A moderately busy street location, an urban background location and a suburban background location were chosen to investigate PM concentration differences.

Characteristics of the three monitoring sites

Figure 1 gives an overview of the geographic locations of the three monitoring sites. The street location was a moderately busy two-lane road with about 15 000 motorised vehicles per 24-hour. The samplers were placed on the pavement at the façade of the buildings, at a distance of 4 m from the side of the road. Inlets were placed ~1.5 m above the ground. The road itself was about 6 m wide. The buildings at the other side were separated from the main road by a canal and an additional one-way street (~4500 motorised vehicles per 24-hour). Hardly any shipping took place. The distance between buildings was about 40 m. This street configuration has been shown to be highly exposed because all winds perpendicular to the road result in exposure at the façade[26], due to turbulence induced by the buildings. Also the short distance to motorised traffic combined with the turbulence caused by the traffic increases the exposure to local traffic emissions. Traffic lights were operated at ~100 m from the sampling site.
At the street location, detailed tube traffic counts were done for a week during the measurement campaign. Total average traffic volume was 15 312 motorised vehicles per 24-hour on workdays. Seven percent was characterised as moderately large vehicles (small trucks, vans) whereas 3% was characterised as large vehicles (trucks, buses). There were no scheduled bus routes passing the street. Total traffic volume was fairly constant during

sampling time, ranging from 867 to 967 motorised vehicles per hour. Average speed was 28 km/h during the sampling time.
The background site was located in the city centre of Utrecht but was not in the direct vicinity of busy streets. Nearest major road was about 100 m from the site with a traffic volume of ~16.000 motorised vehicles per 24-hour. Traffic volume on the nearest minor road (at the other side of the building) was about 2286 motorised vehicles per 24-hour. The urban background site was located in the courtyard patio of an elderly home. Samplers were placed on the first floor on a balcony. Measurements were done at 8 m from the ground (1.5 m above a balcony of 6.5 m). Mean distance to the street location was 1.6 km.
The suburban location was in the village of Bilthoven, which is detached from the city of Utrecht. The equipment was placed in the garden of a single family house. Nearest minor road was about 20 m away (610 motorised vehicles per 24-hour). Nearest major road was about 100 m away. Mean distance to the street location in Utrecht city centre was 6.7 km. Inlets were placed ~1.5 m above the ground. The distance to the major motorway (A27) carrying more than 100 000 vehicles per 24-hour was 2.5 km.
None of the locations was near industrial areas, small industries, gas stations or large parking areas.

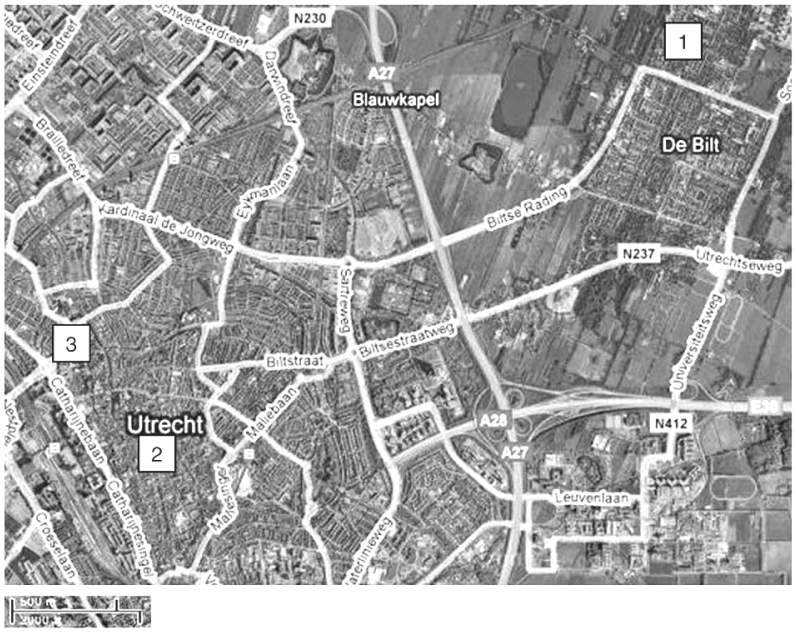

Figure 1 Geographic location of the three measurement sites. 1=suburban background location; 2=urban background location; 3=street location

Exposure measurements

PM_{10} and soot

Six hour average concentrations of PM_{10} were measured with Harvard Impactors operating at 10 liter/minute. PM_{10} was collected on 37-mm 2-µm pore size Teflon filters. Flows were measured with calibrated rotameters. The PM_{10} mass was determined by weighting a filter before and after sampling using a Mettler MT5 micro-balance (Mettler-Toledo, Greifensee, Switzerland) with 1 µg reading precision. Filters were stored at 4°C and were weighted after equilibrating for 24-h in a temperature (20-23°C) and humidity (30-40%) controlled room. Details on the analytic procedures can be found elsewhere[27,28].

After weighing, reflectance of filters was measured and transformed into absorbance, according to Standard Operation Procedures. Absorbance has been shown to be a good indicator for elemental carbon (EC) and soot[27,29]. In this paper we will further use the term soot for the absorbance measurements.

We calculated the EC concentration from the absorbance using a formula derived from collocated measurements in the Netherlands: EC=1.6053*absorption-0.2620[29] to test how much of the contrast in PM_{10} was due to primary soot emissions.

For quality assurance/control field blanks were taken. We took six field blanks by loading a filter cassette of a Harvard Impactor with a filter and leaving it in place for the time of sampling. Mean field blank weight changes were subtracted from all sample weights. Mean blank filter reflectance was accounted for in the calculation of the absorbance.

Particle number concentrations

PNC were measured with a real-time condensation particle counter (CPC model 3007, TSI Inc., MN, USA) for about six hours per sampling day, as after that the CPC 3007 runs out of isopropyl. The CPC 3007 effectively counts particles from 10 nm. Above 100 000 particles/cm^3, the CPC 3007 may systematically underestimate the PNC due to coincidence[13]. We did not make adjustments for the limited number of samples above 100 000 particles/cm^3. Laboratory tests comparing the CPC 3007 and 3022A model at high concentrations showed that up to 200 000 particles/cm^3 no underestimation occurred[30].

Three devices were used for the measurement campaign. The same device was used for each location, apart from four days when devices were switched between the urban and suburban background locations. Before each sampling day a zero check was performed. Also comparability of the three devices was checked overnight for all sampling days. Correlation was high between the devices (R>0.97), although there were systematic differences between the devices that did not show a trend over time. In particular, the device used in the street location gave one average 28% lower readings compared to the

suburban device. The measurements were corrected for these systematic differences.

Traffic counts and meteorological data

Detailed traffic counts were done at the street location for a week in November by making use of a standard Minuteman EVR (Counters & Accessories Ltd, England), which is a two-tube counter. Apart from total traffic counts, speed and classification of the motorised vehicles has been registered as well. No detailed tube traffic counts were done at the other locations. Therefore, motorised vehicles were also counted by hand several times for fifteen minutes at the three locations between 10.00 AM and 15.00 PM, at low traffic hours. Traffic counts were converted to 24-hour estimates by using the same approach as other studies[31].

Data on hourly temperature, relative humidity, wind speed and wind direction were obtained from the Royal Dutch Meteorological Institute (KNMI) station de Bilt, located less than 5 km from Utrecht.

Data management and analysis

Before statistical analyses were performed, the quality of the measurements was assessed. For PM_{10} and soot, three samples were excluded for analysis because of power supply problems, inaccurate time and flow registrations and deviating sampling times (at least 2/3 of the indicated time period): all from the suburban location. Also one urban background sample has been excluded for analysis because of power supply problems, leaving 56 samples available for analysis. At the first measurement day all three PNC samples were missing for analysis because of inadequate data uploading. Eight PNC samples were excluded from analysis due to equipment failure and power problems supply, all but one from the suburban location, leaving 49 samples in total available for analysis. PNC data were checked for unreliable measurements and outliers using previously described procedures[32]. Outlier values were defined as measurements that varied more than a factor ten between successive seconds. Total number of minute averages ranged from 288 to 497 per sampling day, after checking the PNC data for unreliable measurements and outliers. No outlier values were detected with the outlier procedure.

The average reflection of the field blank filter (N=6) was 101.3 (SD=1.4). The limit of detection (LOD), calculated as three times the SD of the blanks divided by the average sampling volume was 0.42 10^{-5}/m for soot. None of the soot samples were below this limit. The average PM_{10} weight of the field blank filter (N=6) was 8.5 µg (SD=12.4) and the matching LOD was 9.4 µg/m^3. Measurements of six days were below the detection limit of PM_{10}, all but one obtained from the suburban background location. Values below the PM_{10} LOD were retained rather then replaced by a standard value. There were no negative PM_{10} values.

Descriptive statistics were calculated for all measurements. PNC were calculated for one-minute mean and median values. Averages were calculated

per sampling day and plotted. To obtain a valid comparison between locations we first have calculated the difference per day. Median ratios were based on those daily differences. Non-parametric Spearman correlation coefficients were calculated to examine relationships between the different PM metrics. For PNC we also evaluated one-minute patterns per location averaged over all measurement days.

Results

Six-hour average concentration patterns

The distribution of PM_{10}, soot and PNC concentration for the different locations are given in Table 1. Figure 2 showed graphically the mean concentrations per measurement day for the three locations. Concentrations were lowest at the suburban and urban background location, and significantly highest at the street location. Relative differences varied between the PM components: the mean PM_{10} concentration at the street location was 1.8 and 1.5 times higher than at the suburban and urban background location, respectively. The mean PNC at the street location was 3.5 times higher than at the suburban background location. The contrast was similar for soot concentrations (street 3.8 and 3.0 times higher at the street location compared to suburban and urban background location). Urban background concentrations were only slightly higher than at the suburban location and none of these differences were statistically significant. The 5.1 10^{-5}/m contrast in soot between street and urban background median concentration was translated into an 8 $\mu g/m^3$ contrast in EC. This implied that approximately 70% of the contrast in PM_{10} was due to primary soot emissions. The ratio between street and background locations was similar for the six-hour mean PNC and six-hour median PNC. We calculated the six-hour median PNC as this is less affected by individual high peaks related to e.g. high-emission individual vehicles. The similarity of the ratios suggested that the large contrast was mostly due to total traffic and not related to a few individual vehicles. Figure 2 illustrated the general pattern observed during the measurement campaign: a large temporal variation with the highest mean concentrations of PNC and soot at the street location on every measurement day. PNC was poorly correlated with PM_{10} and also soot, although this analysis was hampered by the small amount of samples (Table 2).

One-minute PNC concentration patterns

Figure 3 showed the average time pattern of PNC for the three locations. PNC did not change systematically with time of day at the street location, consistent with the lack of trend in traffic volumes within the (afternoon) sampling period for this street. For the other locations a slight increase was found with time of day, especially at the urban background location, probably reflecting the broad evening rush hour peak resulting from traffic in the wider

urban area. At the street location this pattern was probably not detectable because of the dominance of the constant local traffic emissions.
Figure 4 showed time patterns for three typical measurement days by location type (street and background). The street location was characterised by very large temporal variability of one-minute values, probably a combination of short-term fluctuations of wind direction and speed, traffic volumes and emission per vehicle. Field observations suggested that especially with winds parallel to the street limited contributions of traffic were found. At the street location, on all but four sampling days, the highest one-minute values exceeded 100 000 particles/cm^3. Overall, there were 142 observations (2%) above 100 000 particles/cm^3, with eighteen (0.3%) observations above 200 000 particles/cm^3. At the urban and suburban background zero and two minute values exceeded 100 000 particles/cm^3, respectively.
The urban and suburban background locations were generally characterised by more slowly changing temporal variability, with typical clear patterns within the sampling period. Although at some measurement days, considerable temporal variability was also visible at the background sites, see for example measurement day sixteen in Figure 4. The sites were located away from any major road therefore it is unlikely local traffic explained the variability. On most sampling days temporal patterns of one-minute values were similar for the suburban and urban locations, as indicated by the relatively high correlation between the two locations (Table 3). These patterns are likely explained by the influence of area-wide sources or subtle variations in weather conditions. The hourly weather data from the nearby station de Bilt could not explain the observed patterns within sampling periods. There was typically very low correlation with the values at the street location. In the sampling period, average temperature was 12°C (5-20°C). Wind direction was predominantly south (seven times) and west (six times). Mean wind speed was 3.2 m/s (1.1-5.7 m/s).
More short-term peaks were found at the suburban background compared to the urban background location. These typically lasted for a few consecutive minutes, possibly indicating a very local source, suggesting that sources of UFP within urban areas are not completely understood.

Table 1 Distribution of PM_{10} ($\mu g/m^3$), soot ($10^{-5}/m$) and PNC (p/cm^3) at a street, urban and suburban background location

	Location	N	P5	P25	Median	Mean (SD)	P75	P95	Ratio[a]	P-value
PM_{10}	Street	20	21	30	35	40 (14.83)	45	66	1.8	0.02
	UB[b]	19	11	19	23	31 (19.23)	39	61	1.1	0.28
	SUB[c]	17	6	9	17	24 (23.95)	32	56	1	
Soot	Street	20	3.9	5.9	7.1	6.7 (1.4)	7.6	8.4	3.8	0.00
	UB[b]	19	1.0	1.2	2.0	2.3 (1.2)	2.7	4.7	1.2	0.26
	SUB[c]	17	0.6	1.0	1.8	1.8 (1.1)	2.0	3.8	1	
Mean PNC	Street	19	23 877	29 042	37 988	38 635 (10 946)	45 703	62 302	3.5	0.00
	UB[b]	18	8486	11 832	13 522	14 094 (3639)	15 291	22 325	1.2	0.77
	SUB[c]	12	4566	12 048	12 771	13 289 (4201)	15 524	21 852	1	
Median PNC	Street	19	22 304	27 731	36 368	37 073 (10 593)	43 902	60 234	3.4	0.00
	UB[b]	18	8491	11807	13 513	14 077 (3629)	15 278	22 324	1.2	0.72
	SUB[c]	12	4112	11949	12 707	13 080 (4243)	14 934	21 914	1	

[a]Ratio is median ratio. Median of the N daily differences (ratios) is not necessarily the same as the differences between the overall medians in the table;
[b]UB=urban background; [c]SUB=suburban background.

Table 2 Correlation coefficients[a] among six-hour concentrations of different PM component per location

	Location	Soot	PM_{10}
PM_{10}	Street	0.34 (0.14)	
	UB[b]	0.85 (0.00)	
	SUB[c]	0.78 (0.00)	
Mean PNC	Street	0.49 (0.03)	-0.39 (0.10)
	UB[b]	-0.01 (0.98)	-0.01 (0.96)
	SUB[c]	0.46 (0.13)	0.17 (0.60)

[a]Spearman correlation coefficients, p-values in brackets;
[b]UB=urban background; [c]SUB=suburban background.

Table 3 Distribution of correlation coefficients[a] of one-minute PNC concentrations between locations

Location	SUB[c]	Street
Street	0.19 (0.0–0.6)	
UB[b]	0.49 (0.0–0.9)	0.19 (0.0–0.7)

[a]Spearman correlation coefficients (median), min and max values in brackets;
[b]UB=urban background; [c]SUB=suburban background.

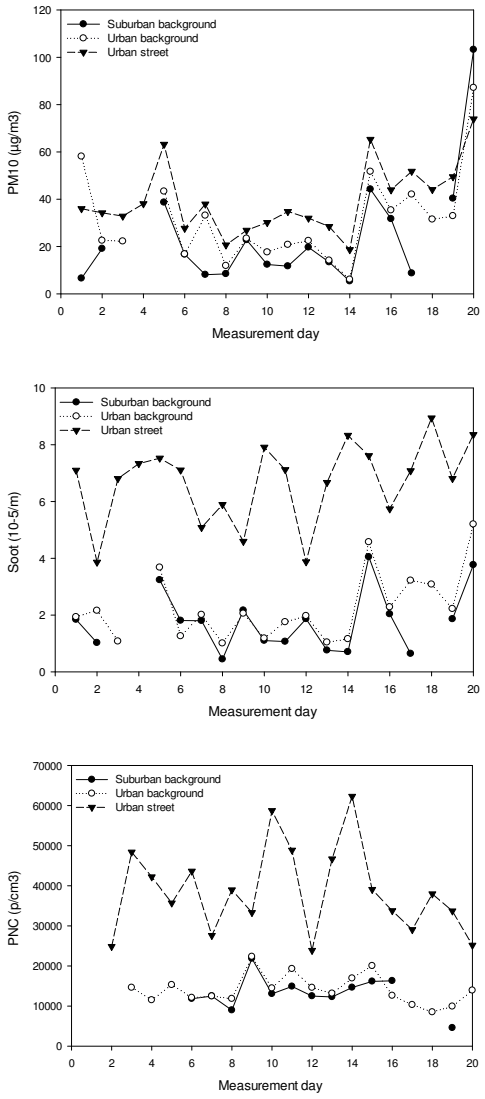

Figure 2 Average concentrations of the three PM components per location per measurement day

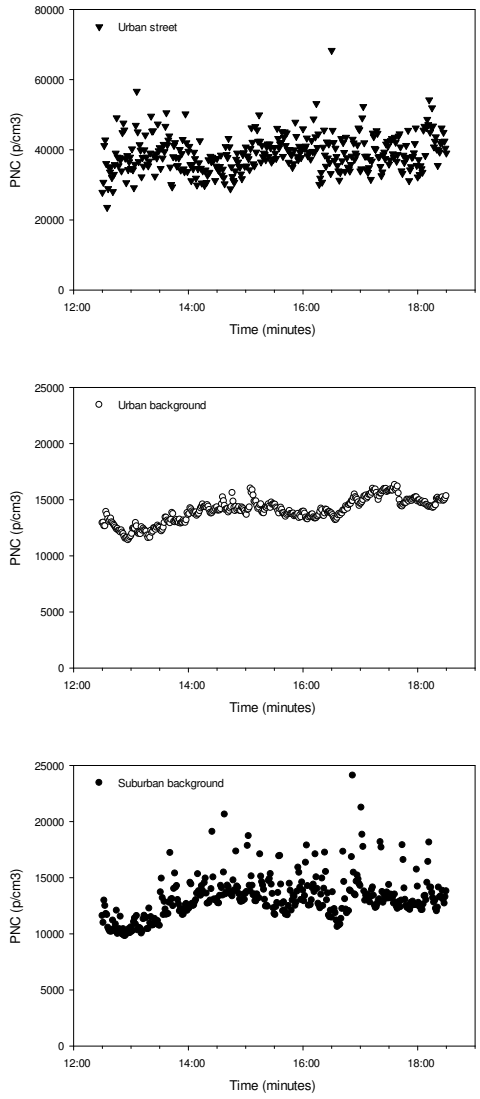

Figure 3 Afternoon pattern of one-minute PNC per location averaged over all measurement days. Note that the scale of the y-axis differ

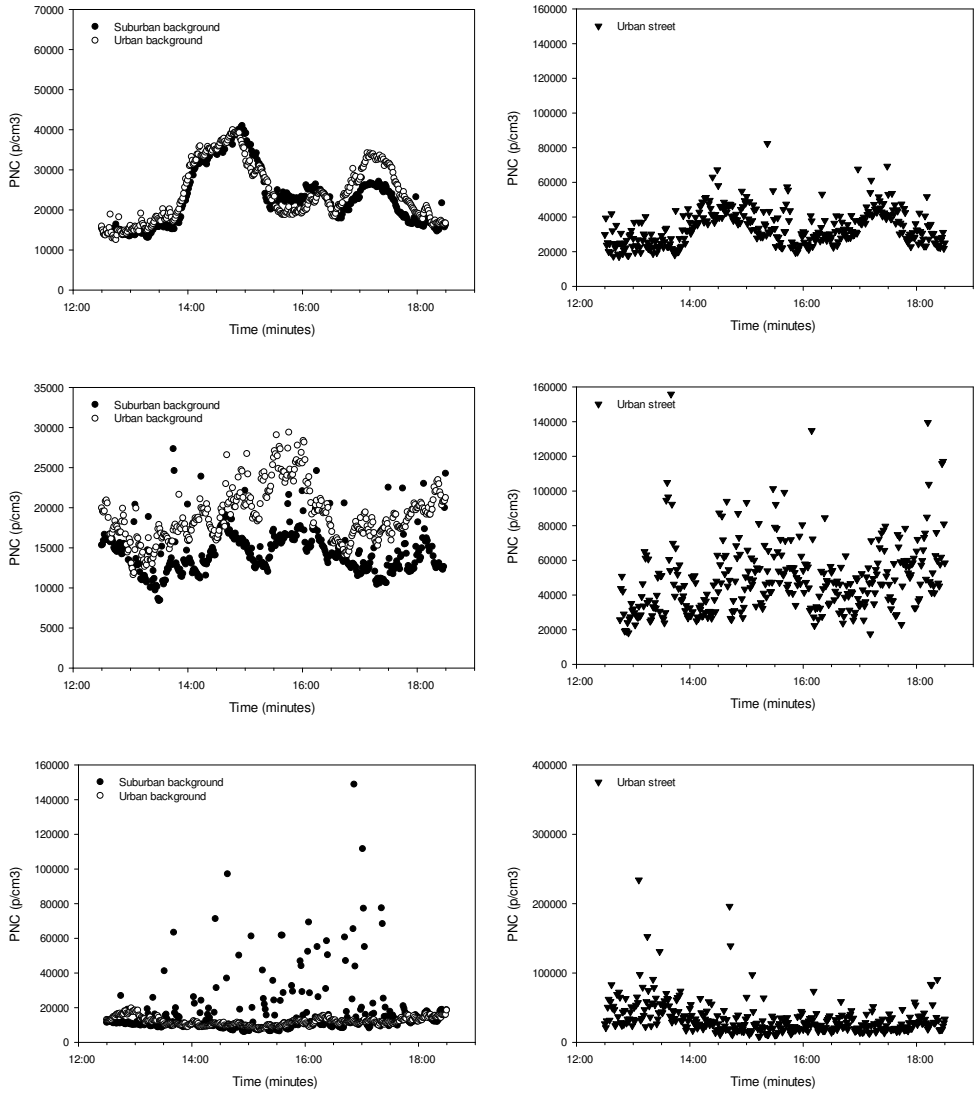

Figure 4 Afternoon pattern of one-minute PNC per location for three typical measurement days (day 9, 11 and 16, respectively). Note that the scale of the y-axis differ

Discussion

Average concentrations of PNC concentrations and soot measured at a moderately busy urban street were more than 3 times higher than simultaneously measured concentrations at urban and suburban background locations. The contrast for PM_{10} concentrations was much smaller with a ratio of 1.5 between the street and urban background location. Within each sampling day, high temporal variability of PNC was found for the street location reflecting varying local traffic influences. For the background locations, smaller and more gradual temporal variability was found probably reflecting area-wide source contributions and meteorology. The street and background locations thus varied both in average concentration and in short-term variability of concentrations.

Comparison of concentration contrasts with previous studies

We have found 3.5 and 3.0 times higher PNC concentrations at a busy street compared to the urban and suburban background location, respectively. Traffic volume was only moderate (15 032 vehicles per 24-hour). Most previous studies were conducted near substantially busier roads, such as motorways. Our study adds that significant exposures occurred at more frequently occurring moderately busy inner-city roads.

Zhu et al.[9,14] measured PNC near or close to motorways up to 300 000 motorised vehicles per 24-hour in Los Angeles, USA, and showed an exponentially decrease in PNC with distance downwind of motorways. The concentration at the nearest site (17 m downwind from the motorway) was about a factor 4 higher than at the background site (300 m downwind from the motorway). It was suggested that both atmospheric dispersion and coagulation play important roles in the rapid decrease of PNC and the change in particle size distribution[9]. The largest contrast was found for the smallest size fraction. Hardly no contrast between sites was seen in PM_{10} concentrations in Birmingham, UK, while PNC at the site 70 m to a major motorway with 160 000 vehicles per 24-hour passing was around 3 times that of the two more background locations[12], similar to our results. PNC concentrations during morning rush hours at very short distance from a freeway in Raleigh, USA, were more than 20 times higher than upwind concentrations[18]. There is much less information about PNC concentration contrasts for less busy urban roads[20,21], to which a significant amount of urban inhabitants are exposed. These studies reported a 2 to 6-fold increase of PNC at traffic sites compared to corresponding urban background sites[20,21]. PNC concentrations were thus substantially increased near motorways and at short distances from (moderately) busy urban roads. Differences in the relative contrast and especially absolute concentrations between studies can be related to the sampling location (e.g. traffic volume, traffic composition, street configuration, distance to the road), and differences in study design. Important differences between studies included the sampling period and

monitoring equipment. Some studies compared 24-hour average concentrations[20,21], whereas other studies reported morning rush hour contrasts[18]. Our sampling time, the afternoon and early evening, will likely be associated with higher than 24-hour average concentrations but lower than the morning rush hour concentrations, as on average daily patterns of PNC are characterised with a sharp increase in particles during morning rush hour, a lower evening peak and low concentrations during late evening and night time[22,32].

Several studies have shown that the spatial gradient near motorways is largest for the smallest particles[14,17]. Studies have used different instruments to measure total or size-selective PNC, including SMPS, CPC3007 and Ptrak. These instruments differ in the lower particle size effectively counting. We used the CPC3007 which starts detecting particles from 10 nm. We therefore largely missed the smallest particles, resulting in lower absolute concentrations and lower contrasts compared to studies effectively counting nanoparticles (<10 nm). The contrast may also be underestimated, because at high particle number counts, the instrument underestimates counts due to coincidence[13]. We tested the instruments in another study[30] before the sampling campaign, and found no evidence for underestimation of the readings of a CPC 3022A until 200 000 particles/cm^3. Very few concentrations above 200 000 were observed in our study. Compared to studies using the PTRAK (counting from 20 nm), our contrast related to monitoring is larger.

Our results confirmed the increasing evidence that proximity to traffic is much better reflected in contrasts in PNC or soot concentrations than in PM_{10} and $PM_{2.5}$ concentrations[21,23-25]. In those studies 1.2 to 1.4 times higher PM_{10} concentrations have been found at busy streets than at background locations. About 70% of the contrast in PM_{10} was due to differences in EC, suggesting that the major part is due to tailpipe emissions rather than re-suspended coarse particles. In an earlier study it was found that 50% of the difference in PM_{10} between a kerbside traffic location and urban background was due to differences in EC[24]. The smaller distance to traffic is probably the reason for the larger fraction of re-suspended particles in that study. This also illustrated the sensitivity of measurement results to the exact site characteristics.

In the present study similar contrast was found for soot (ratio 3.0 to 3.8), largely due to soot emissions from diesel-powered vehicles. Besides practically all trucks, 17% of the passenger cars have diesel engines in the Netherlands[33]. During our sampling time, traffic was standing still for traffic lights, given the relatively stable traffic volumes per hour and the relatively low average speed. Stop-and-go traffic increases emissions of soot and PNC; especially (hard) acceleration contributes to this. Acceleration reduces temporarily the efficiency of catalytic converters[34].

In other studies EC (soot) or BS concentrations were 1.8 to 2.6 times higher at major streets than at background locations[23-25]. Also in a study of Hoek et al.[35] $PM_{2.5}$ concentrations were on average 1.2 times higher at traffic sites than at urban background sites, but soot at traffic sites were between 1.3 to 1.6 times higher than background. Another study conducted in Harlem, New York, comparing sidewalk sites with low and high traffic counts, found that eight

hour measurements of elemental carbon varied 4-fold across sites, whereas for $PM_{2.5}$ this was only 1.3 times[36]. Relative differences between studies can probably be explained by differences in locations (traffic volumes, speed, composition of the roads), sampling methods (amount of samples, sampling time and height, different devices, different sampling distance to the road), and meteorological conditions.
The concentration contrasts of various traffic-related air pollutants is of importance in the interpretation of studies that have shown increased cardio-respiratory morbidity and mortality related to living near major roadways[37]. It has been argued that ultrafine particles may play a role in this association, partly based upon the large contrast in concentration[10]. However, the contrast of other components (e.g. elements, organic compounds) can be even higher. Therefore, new studies should focus on an extensive measurement set of traffic-related air pollutants at various urban roads simultaneously, to which a significant amount of urban inhabitants are exposed. An important issue is the contribution of primary exhaust emissions and non-tailpipe emissions related to brake- and tyre wear and re-suspended road dust[38]. Further work on identifying sources of ultrafine particles at background locations would be useful as well.

Short-term variability of PNC
The analysis of one-minute PNC values showed that the street location not only had higher average concentrations than the background locations, but also showed more short-term variability. This may have consequences for potential adverse health effects, though there is currently no empirical data comparing human responses to a constant exposure or a variable exposure with the same average.
The high temporal variability of minute values at the street location can probably be explained by the variability in wind direction and speed, traffic volumes and emissions of passing vehicles. High-emission vehicles may explained some of the peaks but comparison of the ratio of the six-hour mean and median PNC values at the street location with the background suggested that their contribution to the overall concentrations in the street is limited, and that the large contrast was mostly due mostly to total traffic. Westerdahl et al.[13] measured PNC on the motorway. They showed far more short-term variability of PNC, with a continuously sequence of very high PNC peaks (up to 800 000 particles/cm^3). Individual high-emission vehicles were probably responsible for those sharp peaks, although the origin of many peaks were not readily identifiable[13]. Traffic participation studies involving cyclists and pedestrians[30,39-42] have also found high short-term peaks of on-road PNC concentrations. PNC peaks in these studies were attributed to passing high-emission vehicles.
The occurrence of some moderately elevated short-term peaks at the suburban background was not anticipated, as we selected a location in a residential area without local sources in the vicinity. It suggests that we currently do not completely understand sources of UFP within urban areas. It

is unlikely an artifact of the instrument as we did not identify outliers using our data cleaning procedure. The pattern of the peaks with elevations during a few minutes also supports that it was not an artifact. Until now, the two dominant sources of UFP in ambient urban air are direct combustion emissions from sources like motorised vehicles or power plants and secondary particles generated by the photochemical or physical processes in the atmosphere[13,43,44]. It seems unlikely that secondary particle formation explains the peaks in the weather conditions prevailing in the present study. The site was located away from any major road, and therefore traffic could not explain the peaks either. For indoor environments the most common UFP source is cooking with a gas stove[45]. Results could be influenced by this indoor source, since sampling took place close to the kitchen (5 m) and people were actually at home during sampling. Because the peaks were not present on each sampling day and not related with time of day, this explanation seems unlikely. Other neighborhood sources might include use of fire places.

Limitations

The present study was performed within one month in the autumn season. Also sampling time was relatively short (~six hours). It is therefore possible that the exact magnitude of the differences found between the different locations would have been somewhat different if other time periods had been measured as well, as especially PNC are higher in winter. It is however unlikely that the ranking of differences observed between PM components would have been different.

Exact sampling height was slightly different between the street (1.5 m) and the urban background site (8 m). The reported contrast between the street and the urban background was probably not substantially affected by this difference in sampling height, as in the well-mixed urban atmosphere, vertical gradients are generally small for sampling sites away from local sources such as major roads. In contrast, near major roads large vertical gradients have been identified[46]. When we had lowered the sampling height at the urban background location to 1.5 m, PM_{10} concentrations might have been slightly higher due to re-suspension of soil dust, though 1.5 m is generally considered sufficient to prevent this. Sampling height was identical for the street and the suburban location. Hence, this contrast was not affected by difference in sampling height.

Conclusion

Similar high contrasts in average concentration between a moderately busy street (15 000 vehicles per 24-hour) and urban and suburban background sites were found for soot and PNC. The mean PNC and soot concentration was more than 3 times higher than at the two background locations. The contrast for PM_{10} concentrations was much smaller with a ratio of 1.5 between the street and the urban background location, adding to the evidence that PM_{10} does not reflect the impact of motorised traffic emission well. This study documented that substantial contrasts in PNC and soot exposure occurred near moderately busy roads, in addition to previously documented contrasts near motorways. Short-term variability of PNC was also much higher at the street location and uncorrelated with background variations. Though less frequent, at the background locations high one-minute PNC values were observed as well, suggesting that sources of ultrafine particles are not yet fully understood.

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Chapter 4

Contrasts in oxidative potential and other particulate matter characteristics collected near major streets and background locations

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Abstract

Background
Measuring the oxidative potential of airborne particulate matter (PM) may provide a more health-based exposure measure by integrating various biologically relevant properties of PM into a single predictor of biological activity. We aimed to assess the contrast in oxidative potential of PM collected at major urban streets and background locations; to assess the association of oxidative potential with other PM characteristics; and to assess the oxidative potential in different PM size fractions.

Methods
Measurements of PM_{10}, $PM_{2.5}$, soot, elemental composition and oxidative potential of PM were conducted simultaneously in samples from eight major streets and ten urban and suburban background locations in the Netherlands. Six one-week measurements were performed at each location over a six-month period in 2008. Oxidative potential was measured as the ability to generate hydroxyl radicals in the presence of hydrogen peroxide in all PM_{10} samples and a subset of $PM_{2.5}$ samples.

Results
The PM_{10} oxidative potential of samples from major streets was 3.6 times higher than at urban background locations, exceeding the contrast for PM mass, soot and all measured chemical PM characteristics. The contrast between major streets and suburban background locations was even higher (factor 6.5). Oxidative potential was highly correlated with soot, Ba, Cr, Cu, Fe and Mn. Oxidative potential of PM_{10} was 4.6 times higher than the oxidative potential of $PM_{2.5}$, when expressed per volume unit and 3.1 times higher when expressed per mass unit.

Conclusion
The oxidative potential of PM near major urban roads was highly elevated compared with urban and suburban background locations, and the contrast was greater than that for any other measured PM characteristic.

Introduction

Numerous epidemiological studies have documented the effects of particulate matter (PM_{10}, $PM_{2.5}$) air pollution on morbidity and mortality from respiratory and cardiovascular diseases[1]. Although studies often have measured PM concentrations on a mass basis, proximity to traffic is much better reflected by other PM characteristics, such as soot or particle number concentrations, than by PM mass levels[2,3]. In addition, characteristics such as surface area, acidity, and particle composition (including transition metals and hydrocarbons content) are likely to affect PM toxicity[4-6]. Oxidative stress has been suggested as an important underlying mechanism of action by which exposure to PM may lead to adverse health effects[6]. Oxidative stress results when the generation of reactive oxygen species (ROS), or free radicals, exceeds the available antioxidant defenses. ROS can damage membrane lipids, proteins and DNA, which can result in cell death via either necrotic or apoptotic processes[1,6]. High levels of oxidative stress lead to inflammatory response via activation of various transcription factors and stimulation of cytokine production[4,6]. Inflammation plays an important role in respiratory and cardiovascular diseases that have been associated with PM.

Oxidative potential of PM integrates various biologically relevant properties, including size, surface and chemical composition of PM. Therefore it may provide a more health-based exposure measure than PM mass alone, and may be a better measure of the biologically effective dose that drives adverse health effects[7]. There is no consensus regarding the most appropriate assay to measure the oxidative potential of PM[8]. Studies have made use of various assays, including the ability of PM to induce hydroxyl radicals[9,10], the ability of PM to deplete antioxidants[11] and the promotion of electron transfer measured by the consumption of dithiothreitol (DTT)[12,13].

Several studies have shown increased cardio-respiratory morbidity and mortality related to living near major roadways[1]. Candidate pollutants that may be responsible include ultrafine particles, soot and polycyclic aromatic hydrocarbons, partly based upon the large contrast in concentration[14]. A more biologically relevant measure such as the oxidative potential may help in the interpretation of these studies. To our knowledge no studies have included a systematic comparison of oxidative potential between PM collected near major urban streets and background locations. A few studies have compared the oxidative potential of PM and other PM characteristics. Künzli et al.[15] compared oxidative potential with other $PM_{2.5}$ characteristics measured at twenty different European locations and reported that oxidative potential varied by one order of magnitude both in time and in space, and that it was not well correlated with other PM characteristics. Other studies on the oxidative potential of outdoor PM have focused on the potential for PM to generate oxidative stress[16] and on the difference in oxidative potential between different PM fractions or PM sources[9,11,13,17,18].

Previously we reported a substantial contrast between moderately busy streets and background locations for elemental concentrations of Cr, Cu and

Fe (factor 2 to 3), and for soot and ultrafine particles (factor 1.9), whereas the contrast was small for PM_{10} and $PM_{2.5}$ mass concentrations[19]. Because the chemical analyses conducted did not provide information regarding bioavailability of elements we aimed to evaluate whether these chemical differences were found in oxidative potential.

The present study was conducted within the framework of a study to evaluate the air quality and health benefits of low emission zones directed at heavy duty vehicles in several Dutch cities. Our study includes comprehensive measurements of air quality and of the health status of populations within the study area before and two years after the policy was implemented[19]. In this paper we report the oxidative potential of PM measurements at major urban streets and urban and suburban background locations collected at baseline in 2008. We aimed to assess the contrast in oxidative potential of PM collected at major urban streets and background locations; to assess the association of oxidative potential with other PM characteristics; and to assess the oxidative potential in different PM size fractions.

Methods

Air sampling campaign

PM_{10}, $PM_{2.5}$, soot, elemental composition and oxidative potential of PM were measured in samples collected at eighteen locations in the Netherlands: eight major streets in five cities, five urban background locations (one in each city), four suburban background locations (one of which was used as a comparison location for two nearby cities), and one urban background location in the center of the Netherlands where continuous measurements were made throughout the study period. At each location six weekly samples were collected over a six-month period beginning in June 2008[19]. Each street and its corresponding urban and suburban background location were measured simultaneously during the same week. For budgetary reasons, locations were measured in two rounds, with two cities included in one round and the remaining three in the other (Supplement Table 1). As noted above, continuous measurements were made during the whole study period at a central urban background location, which were used to adjust discontinuous measurements at the other locations to account for temporal variations in background concentrations. The eight street locations had a traffic intensity varying between approximately 10 000 and 19 000 motor vehicles passing every 24-hour (Supplement Table 2). The five urban background locations were located in the city centres but not in the direct vicinity of major streets. The four suburban background locations were in villages near the selected cities.

Elemental composition and other PM characteristics

Exact methods and results of the analysis of elemental composition and other PM characteristics have been published previously[19]. Briefly, PM_{10} and $PM_{2.5}$ were collected gravimetrically on Teflon filters using PM_{10} personal samplers (MSP Corp., Shoreview, MN, USA) and $PM_{2.5}$ GK2.05 cyclones (BGI Inc., Waltham, MA, USA) at all locations. Soot content of all PM_{10} filters was measured using a Smoke Stain Reflectometer (model M43D; Diffusion Systems, London, UK) and transformed into absorption coefficients. In total 105 (92%) PM_{10} filters and 104 (91%) $PM_{2.5}$ filters were available for analysis. For quality assurance, twelve field blank filters and nineteen duplicate measurements also were collected. All measurements were above the detection limit (LOD), with a coefficient of variation (CV; a measure of precision) <10%. All filters were analysed with energy dispersive X-ray fluorescence spectrometry (ED-XRF) at Cooper Environmental Services, Portland, USA. In this paper we report elemental concentrations derived from PM_{10} filters. All elements were above the LOD in all samples except for Al, Ba and V (72-99% of samples>LOD). The CVs for duplicate measurements were <25%.

Oxidative potential

The oxidative potential of PM was measured with the electron paramagnetic resonance (EPR) assay, which uses conditions similar to those in the lungs[7,10]. Besides the EPR assay, other acellular assays exists such as the DTT assay and the antioxidant depletion assay, which are sensitive to slightly different PM characteristics[8]. We chose the EPR assay because it required relatively little material, its costs are relatively low and it has been used in other large air pollution studies[e.g.15].

After ED-XRF analyses, oxidative potential was assessed in all PM_{10} samples and a subset of $PM_{2.5}$ samples (because of budget restraints) in the laboratory of the Province of Limburg, The Netherlands. All $PM_{2.5}$ samples from two sampling weeks in October and December 2008 were included. To our knowledge, effects of XRF analysis on oxidative potential have not been investigated, but XRF analyses will likely not change the composition or valence state of the material on the samples, and is therefore not expected to affect oxidative potential.

The EPR assay measures oxidative potential based on the ability of PM to generate ·OH in the presence of H_2O_2, as described in detail elsewhere[10,15]. Briefly, PM suspensions were prepared from the Teflon filters. Laboratory blank filters were treated similarly and used as controls in the experiments. Generation of ·OH by particle suspensions was studied in the presence of H_2O_2 and the spin trap 5,5-dimethyl-1-pyrroline-N-oxide (DMPO). For the ·OH analyses, 50 µl of the particle suspension was mixed with 50 µl H_2O_2 and 100 µl DMPO. The mixture was incubated in the dark and shaken continuously at 37°C before being filtered through a 0.1 µm pore filter. The clear filtrate was measured with a Miniscope MS100 EPR spectrometer (Magnettech, Berlin, Germany), all under standard conditions. The oxidative potential of each PM sample was calculated from the sum of total amplitudes of the DMPO-·OH quartet signal, and expressed as the total amplitude in arbitrary units divided by the sampled air volume (m^3). Unless stated otherwise ·OH is expressed per volume of air, because this reflects exposure more as encountered by the airways under real-life conditions compared to expression per mass. Mean field blank readings (mean 704 range: 377-1063, all in arbitrary units per m^3) were subtracted from all measurements. The average ratio of the blank readings to the samples was 0.09. All but one oxidative potential measurement were above the limit of detection (>725 per m^3), calculated as three times the standard deviation of the field blank readings. The one value below the limit of detection was retained. The CV of ten oxidative potential duplicate measurements was 19%.

Data management and analysis

We adjusted our measurements for temporal variation using the central reference location. Specifically, we determined the difference between the average values measured over the six-month study period at the central reference location and the value measured during each sampling week at the same location, and then added or subtracted this value from the observed

measurements for samples from the other locations during corresponding weeks[20]. Because this procedure has been applied in previous studies to PM, soot and NO_2, but not elemental composition and oxidative potential, we first compared how measurements of the central reference location correlated with other locations, measured simultaneously. Pearson correlation coefficients were first calculated among samples collected at each location separately, and then summarised by site type (suburban, urban and major urban street) with the use of the median. Correlation coefficients from an individual location had limited precision because they were based upon at most six data points.

Spatial variation

We calculated ratios between major streets and matching background locations for the different PM characteristics. Median ratios were reported per major street (N=6). In addition an overall median ratio was given (N=48). We calculated the median because it is less influenced by outlier values. The overall median ratio was tested for statistical significance with the non-parametric signed rank test.

Association between oxidative potential and other PM characteristics

Pearson correlation coefficients were calculated between location-specific median and averages of the PM characteristics as well as for the individual measurements. Scatterplots were made and Cooks distance was calculated to investigate the extent to which the correlation analyses were driven by outliers.

To investigate which PM_{10} characteristics were associated with oxidative potential variability, we first used univariate mixed models based on individual measurements, using the natural logarithm of oxidative potential as the dependent variable. We identified the PM characteristic that best predicted oxidative potential by comparing the Akaike Information Criterion (AIC) values of the univariate models, and then added other PM characteristics (with p-value<0.10) to that model to determine if they reduced the AIC further (i.e. improve model fit).

Results

Temporal variation

High temporal variability in the oxidative potential of individual PM_{10} samples for all locations is evident when expressed per volume. This variability is not explained by the variability in absolute particle mass collected, as the oxidative potential expressed per mass showed a similar pattern (Figure 1).
The oxidative potential of samples collected at the urban and suburban background locations was highly correlated with simultaneously measured values for samples collected at the central urban background reference location (R=0.81 and R=0.98, respectively; Table 1). Median correlations between urban and suburban locations and the central reference location for corresponding weeks were also high for PM mass and other PM characteristics. In contrast, median correlations between measurements at the street locations and the central reference location were lower for oxidative potential (R=0.61) and other traffic-related indicators such as soot, Cu and Fe, but not for $PM_{2.5}$ and S, which are driven primarily by long-range transport of PM from other sources.

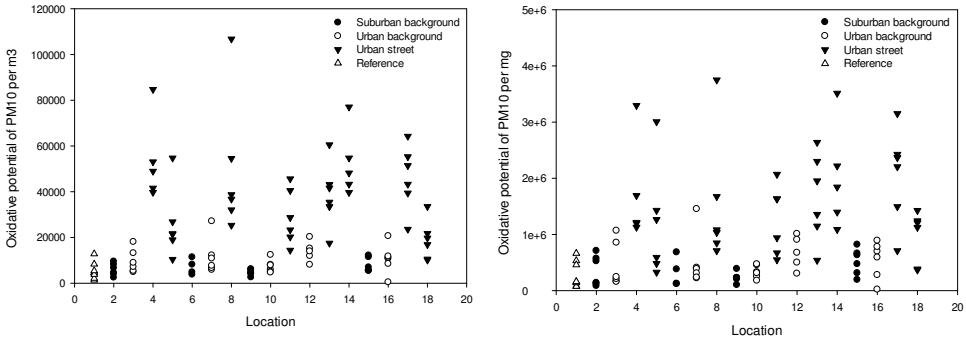

Figure 1 Oxidative potential of PM_{10} measurements per location[a] expressed per volume (m^3) and mass (mg) unit, respectively (N=18, 4 to 6 samples per location)

[a]Locations: 1, central reference location (Utrecht University); 2, suburban Amsterdam; 3, urban background Amsterdam; 4 and 5, urban streets Amsterdam; 6, suburban The Hague; 7, urban background The Hague; 8, urban street The Hague; 9, suburban Den Bosch + Tilburg; 10, urban background Tilburg; 11, urban street Tilburg; 12, urban background Den Bosch; 13 and 14, urban streets Den Bosch; 15, suburban Utrecht; 16, urban background Utrecht; 17 and 18, urban streets Utrecht.

Table 1 Median correlations[a] between PM characteristics measured simultaneously at the central urban background location and other locations

Locations	N	·OH	PM_{10}	$PM_{2.5}$	Soot	Cu	Fe	S
Urban streets	8	0.61	0.92	0.95	0.72	0.40	0.46	0.94
UB[b]	5	0.81	0.92	0.99	0.97	0.90	0.91	0.94
SUB[c]	4	0.98	0.97	0.97	0.97	0.93	0.73	0.97
All	17	0.81	0.92	0.97	0.90	0.85	0.58	0.94

[a]Pearson correlation coefficients were first calculated among samples collected at each location separately (N=4-6 measurements per location) and then summarised with the use of the median, either by site type or all locations together; [b]UB=urban background; [c]SUB=suburban background.

Spatial variation

Oxidative potential of PM_{10} was on average 3.6 times higher at the street locations than at the corresponding urban background locations in each city (Table 2). This contrast was larger than observed for the transition metals Fe and Cu (2.5 and 2.8 times higher, respectively), or for soot and ultrafine particles (1.9 times higher for both). There was relatively little spatial contrast in the regulated PM mass metrics PM_{10} and $PM_{2.5}$ (ratio of 1.2).

The highest oxidative potential contrasts were found for two streets that were classified as street canyons (Stille Veerkade and Brugstraat, which are narrow streets with adjoining high buildings on both sides) and two streets with buildings at one side of the street only (Haarlemmerweg and Weerdsingel Wz) (Table 2) This was consistent with the higher soot, Cr, Cu and Fe contrasts previously reported for the same streets[19].

The contrast in oxidative potential between matched urban and suburban locations was also substantial (1.8 times higher at the urban locations). The oxidative potential of PM_{10} from the major streets was 6.5 times higher than that of the matched suburban locations (Supplement Table 3).

Table 2 Median PM characteristics at the different urban streets and median ratio between street and corresponding urban background[a]

Cities	Urban streets	·OH[b]	PM_{10}[c]	$PM_{2.5}$[c]	Soot[d]	Cr[e]	Cu[e]	Fe[e]
Amsterdam	Haarlemmerweg	50 200	28.6	19.1	3.9	8.9	63.0	1205.7
		6.8	1.2	1.2	2.1	3.5	5.1	3.4
Amsterdam	Hoofdweg	23 500	24.1	15.0	2.8	5.9	27.1	606.7
		2.6	1.1	1.0	1.7	2.0	2.4	1.8
The Hague	Stille Veerkade	38 600	32.5	17.2	4.3	9.8	52.8	1180.6
		4.2	1.3	1.2	2.7	2.9	3.6	2.8
Den Bosch	Brugstraat	49 500	32.7	18.1	3.6	7.5	45.9	975.2
		4.0	1.3	1.3	2.1	2.4	3.6	2.9
Den Bosch	Koningsweg	38 900	30.0	17.5	3.0	5.9	35.1	750.4
		2.5	1.1	1.2	1.9	1.5	2.1	2.0
Tilburg	HVB	24 000	31.5	17.5	2.5	6.0	35.0	666.1
		3.5	1.1	1.1	1.5	2.2	2.6	2.6
Utrecht	Vleutenseweg	18 500	25.0	15.5	2.1	4.8	20.6	387.8
		1.6	1.0	1.1	1.5	1.3	1.5	1.1
Utrecht	Weerdsingel Wz	48 100	28.2	17.2	3.6	7.1	42.0	971.0
		4.1	1.2	1.1	2.2	2.3	3.1	2.7
	Overall ratio[f]	3.6	1.2	1.2	1.9	2.2	2.8	2.5

[a]Data are median concentrations and median ratios with corresponding urban background. Concentrations adjusted for temporal variation using data from the central reference location. Streets in Amsterdam and The Hague and corresponding background locations were measured during the same weeks. Den Bosch, Tilburg and Utrecht (street and background locations) were simultaneously measured during a different set of weeks; [b]Oxidative potential of PM_{10} in arbitrary units per m^3; [c]PM_{10} and $PM_{2.5}$ in $\mu g/m^3$; [d]Soot in $10^{-5}/m$; [e]Elements in ng/m^3; [f]Overall ratio tested on significance with the use of the non-parametric signed rank test, and all p-values were below 0.0001.

Oxidative potential of different PM size fractions

The oxidative potential of PM_{10} was 4.6 times higher than the oxidative potential of $PM_{2.5}$ measured simultaneously in a subset of filters when expressed per volume unit, and 3.1 times higher when expressed per mass unit across all samples tested (Figure 2). These median ratios were statistically significantly different from unity (p-value<0.0001). Note that the model equations in Figure 2 described average ratios together with estimated intercepts. The correlations between the oxidative potentials of PM_{10} and $PM_{2.5}$ were 0.83 and 0.77 when expressed per volume and mass unit, respectively.

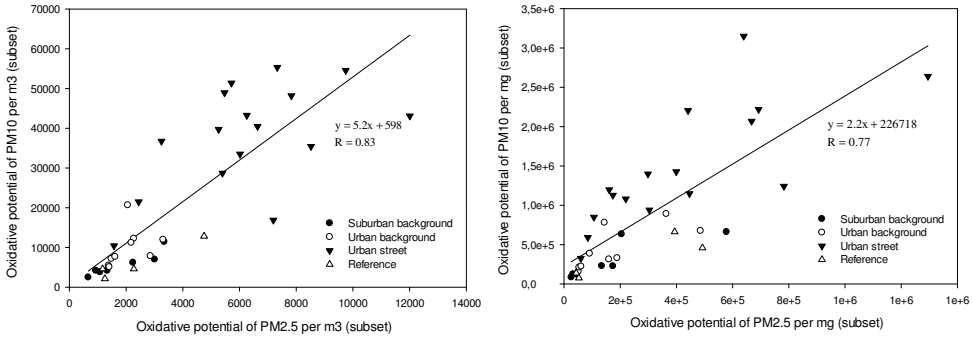

Figure 2 Association between oxidative potential of $PM_{2.5}$ and PM_{10} samples measured simultaneously expressed per volume (m^3) and mass (mg) unit, respectively (N=36)

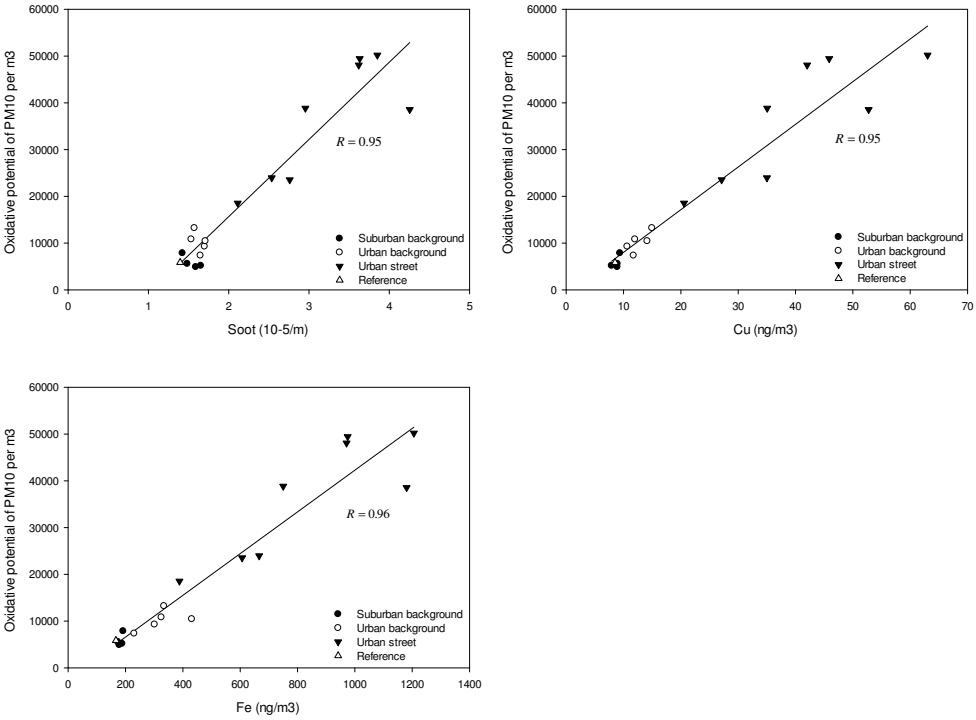

Figure 3 Associations of median oxidative potential of PM_{10} and soot, Cu and Fe concentrations per location, corrected for temporal variation

Association between oxidative potential and other PM characteristics

High correlations (R>0.80) were found between median and average oxidative potential and soot, Ba, Cr, Cu, Fe and Mn in the PM_{10} fraction for all locations (Figure 3). Further analyses were hampered by small sample size. Across all individual samples oxidative potential was highly correlated with soot, Ba, Cr, Cu, Fe and Mn (Table 3). After excluding outlier correlations did not change substantially. PM_{10} and $PM_{2.5}$ measurements were not significantly associated with oxidative potential. Results were comparable when comparisons were limited to samples collected during the same sampling weeks.

When we analysed the oxidative potential of PM_{10} using mixed models based on individual measurements, with location treated as a random effect, the amount of Cu in PM_{10} explained most of the variability in oxidative potential in an one-pollutant model (Supplement Table 4). A two-pollutant model that included Cu and PM_{10} mass had a smaller AIC than the model with Cu alone. In this two-pollutant model an interquartile range (IQR) increase in Cu (22 ng/m^3) was associated with a 176% increase in oxidative potential, whereas an IQR increase in PM_{10} (13 $\mu g/m^3$) was associated with a 35% decrease in oxidative potential. Because the correlation between Cu and PM_{10} was 0.51, the negative PM_{10} slope is unlikely because of collinearity. Cu was highly correlated with soot (R=0.90), Ba (R=0.64), Cr (R=0.84), Fe (R=0.95) and Mn (R=0.74), therefore it was not possible to estimate the independent associations of those with oxidative potential of PM_{10}.

Table 3 Pearson correlation coefficients between oxidative potential (·OH) of PM_{10} and other PM characteristics (N=97 for ·OH, $PM_{2.5}$ and 105 (all others), elements derived from PM_{10} filters)

PM characteristics[a]	·OH	P-value
PM_{10}	0.20	0.05
$PM_{2.5}$	0.08	0.43
Soot	0.73	0.00
Al	0.12	0.21
Ba	0.58	0.00
Br	-0.01	0.88
Ca	0.22	0.02
Cl	0.07	0.46
Cr	0.73	0.00
Cu	0.89	0.00
Fe	0.84	0.00
Mn	0.61	0.00
Ni	0.22	0.02
Pb	-0.01	0.89
S	-0.04	0.64
Si	0.19	0.05
Ti	0.15	0.13
V	0.01	0.90
Zn	0.30	0.00

Discussion

The oxidative potential of PM_{10} collected at eighteen locations spread over the Netherlands varied substantially in time and space. On average, the oxidative potential of PM collected at major urban streets (expressed per volume unit) was 3.6 times higher than simultaneously measured PM at urban background locations. This exceeded contrasts for other PM characteristics such as soot and transition metal contents. The oxidative potential contrast between measurements collected near major urban streets and suburban background locations was even higher (factor 6.5). The oxidative potential of PM_{10} was 4.6 times greater than that of $PM_{2.5}$ measured simultaneously in a subset of filters when expressed per volume unit, and 3.1 times greater when expressed per mass unit. Oxidative potential was highly correlated with soot, Ba, Cr, Cu, Fe and Mn.

Spatial variation

Previously we reported substantially higher concentrations of soot, ultrafine particles, and the transition metals Cr, Cu and Fe at the same moderately busy streets compared with background locations[19]. This reflects local exhaust emissions as well as non-exhaust traffic emissions such as Cu and Fe from brake- and tyre wear. The current paper shows that this contrast is even higher for oxidative potential. This is important, because there is growing evidence that the ability of PM to cause oxidative stress may be an important mechanism for the adverse health effects of particles[1,8].

We used the EPR assay to characterise the oxidative potential of PM. The EPR assay is especially sensitive to transition metals driving ·OH generation mechanisms via the Fenton reaction, which involves the reduction of H_2O_2 by a transition metal ion[21]. Although transition metals influence oxidative potential as measured by the EPR assay, the finding of a high contrast in total transition metal concentrations obtained by ED-XRF does not necessarily imply a high contrast in oxidative potential because ED-XRF measurements do not reflect the valence state or bioavailability of the metals, which can influence oxidative potential substantially. Shi et al.[10] showed a high ability for Cu (II), V (II), V (V), and Fe (II), and less ability for Fe (III) and Ni (II) to generate oxidative potential of PM.

The contrast in oxidative potential of PM from major urban streets and urban background locations may have been greater than corresponding contrasts in the measured concentrations of transition metals because of differences in the concentrations of bioavailable (i.e. soluble) metals between the two site types. Alternatively, other unmeasured pollutants near major roads may affect oxidative potential for example ultrafine particles. Some studies have shown that ultrafine particles have higher oxidant potential than larger PM fractions[13,22,23]. These studies used the DTT assay to measure oxidative potential, which is more sensitive to organic compounds that are concentrated in the ultrafine fraction of PM than the EPR assay, which is

driven by chemical composition rather than by particle size. However, in a subset of two streets (Haarlemmerweg, Vleutenseweg) ultrafine particles showed a smaller contrast at urban streets compared with matching suburban locations than for Cr, Cu and Fe[19]. Finally, it is also possible that differences in oxidative potential of PM from different locations may reflect additive or synergistic effects of different components of PM[24].

Only a few field studies have systematically compared oxidative potential of PM from different locations[15,21], and none have compared oxidative potential of PM from major urban streets with urban background locations. A study in the western part of Germany found approximately 1.7 times higher ·OH generation in PM from three urban locations compared with a more rural location[21], which is very close to the contrast we found between urban and suburban background locations (1.8 times higher for urban vs. suburban locations). Künzli et al.[15] reported a 9.6-fold contrast in mean oxidative potential measured as ·OH generation between different European cities. Most urban sites can be classified as urban background locations. The mean oxidative potential for the second lowest and second highest cities varied by a factor of 3.1, very close to the variability in our study, which covered a much smaller geographic area. This suggested that oxidative potential varied substantially, both on a large and on a fine spatial scale.

Association between oxidative potential and other PM characteristics

We reported high correlations between the oxidative potential of PM_{10} and the concentration of the transition metals Ba, Cr, Cu, Fe, Mn and soot in the PM_{10} fraction. This is consistent with previous observations that transition metals drive ·OH generation via the Fenton reaction, which involves the reduction of H_2O_2 by a transition metal ion[21]. High correlations between oxidative potential and transition metals such as Cu and Fe have been reported previously based on different assays[11,21,25]. The correlations between oxidative potential and transition metals in our study (e.g. R=0.89 and 0.84 for Cu and Fe, respectively) were higher than in the Künzli et al.[15] study (R=0.39 and 0.45 for Cu and Fe), possibly because of the greater diversity of locations included in that study. In that study transition metal concentrations were also measured with ED-XRF. Differences between studies may reflect differences in the bioavailability of the metals for Fenton reactions.

In the present study, the amount of Cu explained variability in oxidative potential of PM_{10} best in univariate mixed models. However, because of the high correlation between Cu and soot, Ba, Cr, Fe and Mn we could not further disentangle independent contributions. Nawrot et al.[26] investigated the oxidative potential of $PM_{2.5}$ in relation to other PM characteristics using the same data as the Künzli et al.[15] study. In their study, 716 filters were available for analysis from twenty different locations that were each sampled over at least twelve months. Across the twenty locations, elements that explained most of the variation in oxidative potential were Fe (positive), Cl and Na (inverse)[26]. In our study, Fe was also significantly and positively correlated with oxidative potential of PM_{10}, whereas Cl and Na concentrations

were not related to oxidative potential. In fifteen locations Nawrot et al.[26] showed a positive relation between oxidative potential and at least one transition metal (i.e. Cu, Fe, Mn, Pb, Ti and V). Associations between $PM_{2.5}$ mass concentrations and oxidative potential varied in that study, with positive associations in eight locations, no association in ten locations, and inverse associations in two locations. We found an inverse relation between oxidative potential and PM_{10} mass in a model that included Cu. An effect of sulphate - a major component of PM mass - may partly explain this relation, because sulphate is able to modify transition-metal-catalyzed oxidative reactions by scavenging ·OH to yield less reactive inorganic radicals[27].
We also found a positive relationship between oxidative potential and soot. Künzli et al.[15] reported a much lower correlation between annual average ·OH and soot (R=0.16, compared with an R=0.73 in our study). Their study was not designed to look at traffic pollution effects specifically, because most of the sites in that study are urban background locations. Temporal correlations between ·OH and soot were low to moderate as well, with the highest correlation among samples collected in Reykjavik, Iceland (R=0.50). Correlation between soot and oxidative potential determined with the DTT assay in another study was also high (R=0.89)[13].

Oxidative potential of different PM size fractions

We found a significantly higher oxidative potential in PM_{10} then in $PM_{2.5}$ samples expressed per equal volume and mass basis. This comparison was based on a subset of the data (34%) and the association might have been different when all study samples were assessed. The average PM_{10} oxidative potential for samples collected in the two weeks when oxidative potential was measured in $PM_{2.5}$ as well was 20 900 per m^3, which is similar to the average value of 21 800 per m^3 for the other four measurement weeks. The ratio of $PM_{2.5}$ to PM_{10} mass was somewhat higher for the two-week subset than for the other measurement weeks (0.67 compared with 0.53), mostly due to higher S and Si contributions to $PM_{2.5}$ mass. The overall ratio of Cu in $PM_{2.5}$ and Cu in PM_{10} was 0.16 for the selected period and 0.15 for the non-selected period. For Fe these ratios were 0.22 and 0.18. Therefore it seems unlikely that the differences in the oxidative potential of PM_{10} and $PM_{2.5}$ would have varied substantially if all samples had been measured.
It is likely that chemical composition, rather than PM size, is responsible for differences in the oxidative potential of PM_{10} and $PM_{2.5}$. Total transition metals were especially present in the coarse fraction of PM_{10}. Other studies have also reported that coarse particles rather than $PM_{2.5}$ had the highest oxidative potential on a per unit mass basis[9,21]. Differences between studies are partly related to the assay used; the EPR assay used in the present study being more sensitive to transition metals, whereas the DTT assay used by others is more sensitive to organic components such as polycyclic aromatic hydrocarbons and quinones. The high oxidative potential of coarse particles based on the EPR assay supports the hypothesis that coarse particles may be able to induce oxidative stress, and that ultimately this may lead to adverse

health effects. Epidemiological studies have found associations between daily variation in coarse particle concentrations and a range of adverse respiratory and cardiovascular health effects[28]. Our study adds to the evidence that these effects may be due to similar mechanisms as hypothesised for fine and ultrafine particles.

Limitations

An important limitation is that we used only one assay to measure oxidative potential of PM. How the EPR assay relates to oxidative potential as measured by other assays is not clear. All laboratory tests of oxidative potential are sensitive to slightly different panels of metals or organic PM characteristics such as quinones and endotoxins[8]. The EPR assay used here is not sensitive to quinones or endotoxins, in contrast with assays such as the antioxidant depletion assay[29]. Künzli et al.[15] used two different assays to measure oxidative potential of PM; the EPR and the antioxidant depletion assay. The correlation between the EPR and the antioxidant depletion assay results varied between 0.18 and 0.65 for the different cities, showing poor to only moderate agreement between assays.

Like most other studies investigating oxidative potential of PM, PM was first collected on Teflon filters. Redox active volatile compounds may not be fully captured by these filters. Coarse and fine PM recovery of filters was estimated to be around 80-90%, based on other studies[9,15,21]. Exact recovery is unknown, and depends on the exact composition of the sampled PM.

The significance of PM oxidative potential for health at present is known only from experimental studies on biomarkers of early biological effects[9,21]. Almost no epidemiological studies have linked high levels of oxidative potential of particles to adverse health effects, such as respiratory or cardiovascular morbidity and mortality. In a recent panel study, Delfino et al.[30] related oxidative potential - using the alveolar macrophage ROS assay - to airway and systematic inflammation in 60 elderly people. The oxidative potential effects on IL-6 and exhaled NO were comparable with the effects of traffic pollutants, such as black carbon and ultrafine particles[30]. The use of different oxidative potential assays, difficulties to assess exposure because the oxidative potential of PM varies substantially in time and space, and the high costs of the different assays currently hampers wide-scale use. Further studies that evaluate the relationship of oxidative potential of PM using different assays and public health are needed[8,15].

Conclusion

The oxidative potential of PM_{10} collected at major streets was 3.6 times higher than that of simultaneously measured PM_{10} from urban background locations, and 6.5 times higher than PM_{10} from suburban locations. These contrasts exceeded corresponding spatial contrasts in other PM characteristics, including concentrations of transition metals such as Cu and Fe. Oxidative potential was highly correlated with soot, Ba, Cr, Cu, Fe and Mn. The oxidative potential of PM_{10} was 4.6 times greater than the oxidative potential of $PM_{2.5}$, measured simultaneously in a subset of filters when expressed per volume unit, and 3.1 times greater when expressed per mass unit.
Given the large contrast in oxidative potential of PM between moderately busy streets versus background locations, and some indications that oxidative potential may be more directly biologically relevant than other measures of exposure, oxidative potential may serve as a better indicator to assess and control adverse health effects of traffic-related PM.

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Supplement

Supplement Table 1 Exact air sampling schedule

Cities	Location	MEASUREMENT WEEK[a]												
		1	2	3	4	5[b]	6[b]	7	8	9[b]	10[b]	11	12	
Amsterdam	Haarlemmerweg	X			X		X		X		X		X	
	Hoofdweg	X			X		X		X		X		X	
	UB[c]	X			X		X		X		X		X	Round A
	SUB[d]	X			X		X		X		X		X	
The Hague	Stille Veerkade	X			X		X		X		X		X	
	UB[c]	X			X		X		X		X		X	
	SUB[d]	X			X		X		X		X		X	
Den Bosch	Brugstraat		X	X		X		X		X		X		
	Koningsweg		X	X		X		X		X		X		
	UB[c]		X	X		X		X		X		X		
	SUB[de]		X	X		X		X		X		X		
Tilburg	HVB		X	X		X		X		X		X		Round B
	UB[c]		X	X		X		X		X		X		
	SUB[de]		X	X		X		X		X		X		
Utrecht	Vleutenseweg		X	X		X		X		X		X		
	Weerdsingel		X	X		X		X		X		X		
	UB[c]		X	X		X		X		X		X		
	SUB[d]		X	X		X		X		X		X		
	Central reference	X	X	X	X	X	X	X	X	X	X	X	X	

[a]Measurements performed in 2008 and beginning of 2009: 1, June 12–June 19; 2, June 24–July 01; 3, Sept. 1–Sept. 08; 4, Sept. 11–Sept. 18; 5, Sept. 29–Oct. 06; 6, Oct. 30–Nov. 06; 7, Nov. 10–Nov. 17; 8, Nov. 20–Nov. 27.; 9, Dec. 01–Dec. 08; 10, Dec. 11–Dec. 18; 11, Jan. 12–Jan. 19; 12, Jan. 22–Jan. 29; [b]In this week oxidative potential of $PM_{2.5}$ was also measured; [c]UB=urban background; [d]SUB=suburban background; [e]One suburban background location was used for two nearby cities (Den Bosch, Tilburg).

Supplement Table 2 Detailed characteristics of the different urban streets

Cities	Urban streets	Traffic intensity	Fraction[a]		Speed (km/h)		Road type	Distance road axis
		Per 24-h	Middle[b]	Heavy[c]	Per 24-h	In rush hours		
Amsterdam	Haarlemmerweg	15 253	0.03	0.02	41	38	Adjoining buildings on one side	11
Amsterdam	Hoofdweg	9774	0.01	0.06	41	41	Adjoining buildings on two sides	16
The Hague	Stille Veerkade	17 438	0.05	0.02	34	32	Canyon	11
Den Bosch	Brugstraat	17 896	0.05	0.05	32	28	Canyon	9
Den Bosch	Koningsweg	17 138	0.05	0.03	46	39	Adjoining buildings on two sides	14
Tilburg	HVB	18 812	0.03	0.07	51	49	Adjoining buildings on two sides	12
Utrecht	Vleutenseweg	13 553	0.06	0.05	39	36	Adjoining buildings on two sides	15
Utrecht	Weerdsingel Wz	14 831	0.06	0.03	35	31	Adjoining buildings on one side	9

[a]Fraction of total traffic intensity; [b]distance between wheel axes 3.5-7 m; [c]distance between wheel axes >7 m.

Supplement Table 3 Median ratio streets and urban background locations vs. suburban background locations

Cities	Locations	·OH
Amsterdam	Haarlemmerweg	11.6
	Hoofdweg	4.2
	UB[a]	1.8
The Hague	Stille Veerkade	8.7
	UB[a]	2.1
Den Bosch	Brugstraat	11.6
	Koningsweg	8.2
	UB[a]	3.1
Tilburg	HVB	5.3
	UB[a]	1.5
Utrecht	Vleutenseweg	2.2
	Weerdsingel Wz	6.1
	UB[a]	1.6
	Overall ratio streets	6.5
	P-value[b]	0.00
	Overall ratio UB[a]	1.8
	P-value[b]	0.00

[a]UB=urban background; [b]Overall ratio tested on significance with the use of the non-parametric signed rank test.

Supplement Table 4 Relation between oxidative potential and other PM characteristics - results of mixed model analysis

	IQR[a]	Beta	SE[b]	P-value	AIC[c]
One-pollutant model[d]					
PM_{10}	13	-0.0100	0.0068	0.0867	232.7
$PM_{2.5}$	14	-0.0100	0.0072	0.1235	220.8
Soot	1.4	0.2600	0.0717	0.0005	220.7
Ba	9	0.0241	0.0105	0.0235	229.9
Cu	22	0.0368	0.0040	<0.0001	198.6
Cr	3	0.0734	0.0299	0.0163	227.8
Fe	471	0.0012	0.0002	<0.0001	226.1
Mn	7	0.0066	0.0126	0.6016	234.1
S	834	-0.0002	0.0001	0.0132	237.9
Two-pollutant model[d]					
Cu	22	0.0462	0.0040	<0.0001	181.8
PM_{10}	13	-0.0335	0.0063	<0.0001	

[a]IQR=interquartile range; [b]SE=standard error; [c]AIC=Akaike Information Criterion (the lower, the better model fit); [d]To estimate percentage change per IQR of a pollutant the following formula can be used: $((e^{IQR*beta}) -1)*100$.

Chapter 5

Impact of low emission zones and local traffic policies on ambient air pollution concentrations

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Abstract

Background
Evaluations of the effectiveness of air pollution policy interventions are scarce. This study investigated air pollution at street level before and after implementation of local traffic policies including low emission zones (LEZ) directed at heavy duty vehicles (trucks) in five Dutch cities.

Methods
Measurements of PM_{10}, $PM_{2.5}$, soot, particle number concentration (PNC), NO_2, NO_x and elemental composition of PM_{10} and $PM_{2.5}$ were conducted simultaneously at eight streets, six urban background locations and four suburban background locations before (2008) and two years after implementation of the policies (2010). The four suburban locations were selected as control locations to account for generic air pollution trends and weather differences.

Results
All air pollution concentrations were lower in 2010 than in 2008. For traffic-related pollutants including soot, NO_x and elemental composition (Cr, Cu, Fe) the decrease did not differ significantly between the intervention locations and the suburban control locations. Only for $PM_{2.5}$ reductions were considerably larger at urban streets (30%) and urban background locations (27%) than at the matching suburban control locations (20%). In one urban street where traffic intensity was reduced with 50%, soot, NO_x and NO_2 concentrations were reduced substantially more (41, 36 and 25%) than at the corresponding suburban control location (22, 14 and 7%).

Conclusion
With the exception of one urban street where traffic flows were drastically reduced, the local traffic policies including LEZ were too modest to produce significant decreases in traffic-related air pollution concentrations.

Introduction

Numerous studies have documented effects of outdoor air pollution on morbidity and mortality from respiratory and cardiovascular diseases[1]. To protect public health, the European Union has set air quality standards of which the standards for particulate matter (particles smaller than 10 μm (PM_{10}) and smaller than 2.5 μm ($PM_{2.5}$)) and nitrogen dioxide (NO_2) are the most important. These standards are still exceeded especially near busy roads. In response to this, substantial policy measures have been taken to reduce air pollution. Europe-wide policies include emission standards for motor vehicles and definition of national emission ceilings. In addition, local policies have been implemented to reduce air pollution concentrations at busy urban roads. Examples of local traffic policy measures include congestion charging directed to all passenger cars entering the inner-city, and low emissions zones (LEZ) designed to limit traffic of old vehicles in city centres. To date, LEZ have been implemented in 152 cities in nine EU countries, and are being considered in other cities worldwide[2].

There is very little evidence of the effectiveness of these local policies in reducing air pollution and ultimately improving public health. From a cost effectiveness standpoint, evaluation of these policies is of interest, as questions arise whether they are worth pursuing[3]. Early examples of intervention studies include an evaluation of a steel mill closure in Utah Valley[4], a ban on coal sales in Dublin, Ireland[5], and regulations to reduce sulphur in fuel in Hong Kong[6]. Recently, research in the effectiveness of policy interventions has increased. The US-based Health Effects Institute (HEI) has promulgated a research programme to investigate interventions[3]. Studies include the traffic policies taken at the Summer Olympic Games in Atlanta[7] and the sale ban of coal in multiple Irish cities[8]. To evaluate policies in the Accountability programme of HEI, a full chain approach has been used where policies are assessed at different stages including drivers, emissions, concentrations, exposures, doses and health effects. Studies typically compare air pollution and/or population health before and after implementation of a policy intervention. One lesson from previous studies is the importance of including control locations to account for temporal trends, unrelated to the policy. An illustration of this is the re-evaluation of the Friedman et al.[9] finding of significant decreases in ozone (~30%), and associated health effects in relation to the traffic policy measures during the 1996 Summer Olympic Games in Atlanta, USA. Peel et al.[7] showed that changes in air pollution were also occurring at the time in other cities throughout the Southeast of USA, where temporary traffic restrictions policies were not in place.

Previous studies have evaluated the effectiveness of local traffic policy measures using modelled instead of measured air pollution concentrations. Examples of modelled evaluations of (local) traffic policy measures include the congestion charging zones in London[10] and Stockholm[11] and the LEZ established in Rome[12]. The recent extensive evaluation of the London

congestion charging zone illustrates that approaches based upon modelling and measurement can differ substantially[13,14]. Modelled emissions of PM_{10} and NO_x dropped with about 20%, resulting in small reductions in modelled annual average concentrations (0.8 and 2.2 $\mu g/m^3$ in PM_{10} and NO_x). Measurements of the single roadside monitor and three urban background sites within the zone did not show any relative change due to the policy for NO_x, and some evidence for a decrease in PM_{10} (one urban background site available)[14].

We started a study to evaluate the impact on air quality and population health of local traffic policies including LEZ directed at heavy duty vehicles in several Dutch cities. Before the implementation the expected impacts of the LEZ have been modelled. Models predicted widely varying estimates for the concentration reductions, up to 1.5 for PM_{10} and 4 $\mu g/m^3$ for NO_2, due to the LEZ[15]. Our study is on a much more local scale that any other traffic policy evaluation before: the main focus is on street level. The study includes comprehensive measurements of air quality at street level before (2008) and two years after the implementation of the policies (2010).

The aim of this paper is to document changes in air pollution in relation to local policies including the LEZ directed at heavy duty vehicles. Air pollutants studied were PM_{10}, $PM_{2.5}$, particle number concentration (PNC), soot, NO_2, NO_x and elemental composition of PM_{10} and $PM_{2.5}$. Our hypothesis was that the LEZ would affect traffic-related components like soot and NO_x more than PM_{10} and $PM_{2.5}$, since the latter are more general air quality indicators with major contributions from secondary components, such as nitrates and sulphates. It is well-documented that proximity to traffic is much better reflected in contrasts in particle number concentrations (PNC) or soot than in PM_{10} and $PM_{2.5}$ levels[16]. We further hypothesised that reductions would be more pronounced at major urban streets than at urban background locations within the LEZ. The study also included an evaluation of changes in population health which will be reported separately. Results of the measurements before the LEZ implementation have been published previously[17,18].

Methods

Study design

Measurements of PM_{10}, $PM_{2.5}$, soot, NO_2, NO_x and elemental composition of PM_{10} and $PM_{2.5}$ were conducted simultaneously at eighteen locations in the Netherlands before (2008) and two years after the implementation of the policies (2010) (Figure 1). Measurements were done at eight major streets and five urban background locations (one in each of the five cities) located within the LEZ. An additional urban background location was selected as a reference location to adjust for temporal variation (not in the LEZ). We further include four suburban background locations as control locations. One suburban location was used for two nearby cities (Den Bosch, Tilburg). The suburban background locations were in villages (~30 000 inhabitants) near the selected cities (10-30 km). In addition, we measured PNC at only four of these locations (two urban streets, two suburban background locations). LEZ policies will potentially affect pollution concentrations in major streets more than at urban background locations. We compare street and urban background locations with simultaneously measured suburban background locations (likely not affected by the LEZ) to distinguish 2010–2008 concentration trends attributable to the LEZ, from those attributable to other factors such as generic emission reductions, the economic downturn since 2008 and the weather. Sampling was done within the same seasons in the second half of 2008 and 2010, with identical equipment and at exactly the same location.

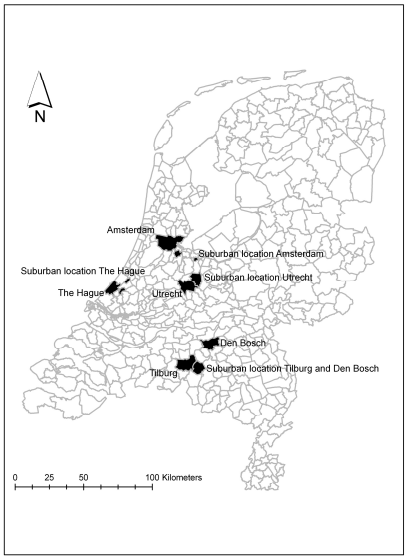

Figure 1 Geographical location of the measurement sites in the Netherlands

Traffic policy measures

The main policy we evaluated was the LEZ. From July 2007 to October 2008 the LEZ was gradually implemented in several Dutch cities. The aim of the policy was to comply with EU air quality standards for PM_{10} and NO_2. The major component of the policy was directed at forbidding old trucks to enter the LEZ in the inner-city. Initially, only EURO-0 and EURO-I trucks were forbidden, whereas EURO-II and EURO-III trucks were only allowed when retrofitted – if filters were available. Given the relatively small amount of particle traps available on the market at that time, EURO-II and EURO-III trucks were largely tolerated till 2008/2009. Since 2010 all EURO-II trucks were forbidden as well, and EURO-III trucks were only allowed if retrofitted with particulate filters and if not older than eight years. The EURO-0 and EURO-I emission standards applied to the type approval tests of trucks that entered the market before 1992. The EURO-II to EURO-VI standards applied to type approval of new truck engines from 1996, 2000, 2005, 2008 and 2013 onwards, respectively. The emission standards progressively decreased NO_x and PM emissions with about a factor 2 (NO_x) to 8 (PM), comparing EURO-IV with EURO-II trucks, due to the increasingly stringent EURO standards (Supplement Table 1). Emission factors estimated from EURO standards - related to the average type of trucks and average Dutch urban driving conditions - reduced with about the same order of magnitude (Supplement Table 2, for NO_x only). Enforcement was tightened with cameras and special staff since 2010, with high fines for drivers entering illegally. Percentage enforcement ranged from 48-66% of the less strict policy in 2008, to 85-97% in 2010 (Table 1). LEZ in the cities cover only a small area of the cities (up to ~10%). However, their impact is larger as (cleaner) trucks with destination city centre will also pass through other areas. Apart from the LEZ other traffic policies measures were introduced in the same period (Table 1). In The Hague inner-city a traffic circulation plan was implemented in 2009 aimed at reducing traffic flows at hotspots including the street we studied.

Air sampling campaign

At each location, six weekly samples were collected spread over two six-month periods in 2008 and 2010 respectively. For logistical reasons, locations were measured in two rounds. All locations within the same city and the nearby suburban control location were measured simultaneously in the same sampling round. An additional urban background location was selected in Utrecht – located in the centre of the country - as a reference location where continuous measurements were made during the whole study period. Therefore we could adjust discontinuous measurements at the other locations for temporal variations in background concentrations.

Sampling locations were exactly the same in all but one location between the two time periods. The urban background location for 2010 in Utrecht was about 100 m away from the 2008 location because of renovation work at the former location, but remained an urban background location. Sampling height was similar. Sampling and analysis methods have been published before[17,18].

Table 1 Traffic policies implemented during study period

Cities	Urban streets	Traffic policies	Implementation date	Enforcement LEZ[b]
Amsterdam	Haarlemmerweg	LEZ[a]	09-10-2008	Camera enforcement from 2009
Amsterdam	Hoofdweg	LEZ[a]	09-10-2008	Enforcement ~66% (2008); ~97% (2010)
The Hague	Stille Veerkade	LEZ[a]	16-04-2008	Camera enforcement from 2010
		Traffic circulation plan	20-11-2009	Enforcement ~72% (2008); ~95% (2010)
Den Bosch	Brugstraat	LEZ[a]	01-09-2007	With the use of personnel
		Deal with local transport company not to drive	24-11-2010	Enforcement ~48% (2008); ~85% (2010)
Den Bosch	Koningsweg	LEZ[a]	01-09-2007	
		Deal with local transport company not to drive	24-11-2010	
Tilburg	HVB	LEZ[a]	01-09-2007	With the use of personnel
		Extension of the LEZ	01-01-2010	Enforcement ~82% (2008); ~83% (2010)
Utrecht	Vleutenseweg	LEZ[a]	01-07-2007	With the use of personnel
Utrecht	Weerdsingel Wz	LEZ[a]	01-07-2007	Enforcement unknown (2008); ~85% (2010)

[a]Low emission zone (LEZ) directed at old trucks (see section traffic policy measures); [b]Enforcement percentages derived from local municipalities and based on different count methods.

Air pollutants

Briefly, PM_{10} and $PM_{2.5}$ were collected gravimetrically on Teflon filters using PM_{10} personal samplers (MSP Corp., Shoreview, MN, USA) and $PM_{2.5}$ GK2.05 cyclones (BGI Inc., Waltham, MA, USA) at all locations. After weighting, reflectance of PM_{10} filters was measured and transformed into absorption coefficients. Absorbance has been shown to be a good indicator for elemental carbon (EC) and soot[19,20]. In this paper we will further use the term soot for the absorbance measurements. From the two campaigns, there were 105 (92%) and 94 (82%) PM_{10} filters and 104 (91%) and 92 (81%) $PM_{2.5}$ filters available for analysis, respectively. We had to exclude PM_{10} and $PM_{2.5}$ concentrations from the first sampling week of the post campaign because a problem occurred in the pre-weighing of the filters. The soot reading of those filters was valid since they were not dependent on the mass weightings. In addition we excluded all measurements from the second sampling week at the Stille Veerkade in The Hague because unexpectedly, the street was closed because of road construction.

NO and NO_2 concentrations were measured with OGAWA passive samplers (OGAWA & Company Inc., FL, USA) with pre-coated NO_2 and NO collection pads. They were placed at the same height near the PM equipment. Sampling and analysis were performed according to procedures reported before[21]. There were no missing samples.

All filters were analysed with energy dispersive X-ray fluorescence spectrometry (ED-XRF) at Cooper Environmental Services, Portland, USA. We focused in this paper on those elements which show the largest contrast between streets and background locations in the 2008 campaign (Cr, Cu, Fe).

All pollutants showed good agreement and almost all samples were above limit of detection (LOD) (Supplement Table 3). Percentage of samples above LOD ranged from 96 (Cr) to 100% ($PM_{2.5}$, NO_2, Cu, Fe). Values below the LOD were retained.

PNC have been measured at two street locations and the matching suburban locations in two sampling rounds. Particle size distributions were measured using a Scanning Mobility Particle Sizer (SMPS) comprising a TSI Model 3071 Electrostatic Classifier and Model 3022 Condensation Particle Counter. Eighteen and 16 (pre) and 33 and 21 paired measurement days (post) were available for analysis in the Amsterdam and Utrecht round. No correction was made for the diffusion losses in both campaigns.

Traffic characteristics and meteorological data

Automated traffic counts were done at the street locations for one week in November 2008 and 2010 by making use of a standard Minuteman EVR (Counters & Accessories Ltd, England), which is a two-tube counter. Classification (light, middle and heavy) of traffic has been registered as well. We performed manual counts as well, since it is known that two-tube counters have difficulties to distinguish large vans from small trucks, resulting in a disproportional large fraction of especially middle traffic[20]. Therefore, vehicle count and classification of motorised vehicles were also registered

manually for 30 minutes at low traffic hours (at least two times per period). This approach to calculate total traffic has been applied successfully before[21]. Classification middle and heavy vehicles in EURO classes was derived from local governments (pre) and from a modelled evaluation of the LEZ (post)[22]. Classification was available on city level only, and was based on different count methods and different years for the cities studied.
Data on 24-hour standard meteorological parameters were obtained from the Royal Dutch Meteorological Institute, station the Bilt, located in the middle of the Netherlands (<5 km from Utrecht) to characterise weather conditions.

Data management and analysis
We adjusted our measurements for temporal variation using the central reference location. Specifically for both campaigns, we determined the difference between the average value (N=12 weeks, spread over a six-month period) and the value measured during each sampling week at the reference site, and then added or subtracted this value from the measured concentrations at the other locations during the same sampling week[23]. At both campaigns, correlations between the reference site and other background sites was high for $PM_{2.5}$, soot, NO_2, NO_x and Cu (R>0.90). For Cr and Fe correlations were somewhat lower (R>0.60). The PM_{10} post campaign correlation was lower (R=0.60) than during pre campaign (R>0.90), probably because of a local source of especially coarse particles at the reference location. This local source does not result in high Cr, Cu and Fe concentrations at the reference location. Therefore, for the post PM_{10} temporal correction, we have made use of the $PM_{2.5}$ measurements at the reference site, which correlated better with the PM_{10} measurements at the other locations (R>0.80). Adjusted averages and absolute changes were calculated for the different pollutants. T-tests and Wilcoxon tests were performed to statistically test the differences between pre and post measurements. We next tested whether the pre to post changes at the urban streets and urban background locations were statistically different from the changes at the matching suburban locations. For this purpose, we subtracted the individual measurement at the suburban site from the measurement made simultaneously at the urban site. Wilcoxon tests were added to the T-tests to test sensitivity to outlier values. We reported significance of T-tests and noted when the Wilcoxon test produced a different result.
Meteorological conditions can differ between the two time periods. Since we measured street and matching urban and suburban location in both periods simultaneously, the 2010-2008 comparisons between street, urban and suburban concentrations remain valid. Particularly for locally generated pollutants, the difference between a street and a suburban background site may differ with dispersion conditions, such as wind speed. We therefore performed additional regression analyses in which the difference between street and corresponding suburban background of all individual measurements was adjusted for the average wind speed during the period.

Results

Pre and post intervention traffic characteristics

At the individual street level, total traffic intensity was consistently reduced with 11 to 51% at two urban streets with both count methods (Hoofdweg and Stille Veerkade) (Table 2). The most striking reduction in traffic intensity took place at the Stille Veerkade in The Hague (decrease of ~6000 to 9000 vehicles per 24-hour). This was in line with the implementation of the traffic circulation plan in The Hague, aimed to reroute traffic from busy streets to other 'less busy' streets. Based upon the manual counts, at the urban and suburban background locations, total traffic intensity was below 1250 vehicles per 24-hour. Differences between 2008 and 2010 were below 300 motorised vehicles per 24-hour at the background locations.

Classification (light, middle and heavy) of the motorised vehicles has been recorded as well. Fraction middle (e.g. small trucks) and heavy traffic (e.g. large trucks, buses) was quite comparable before and after the policies at all but two streets. Only at the Stille Veerkade, the fraction of middle traffic reduced with 1 to 3%. At the Vleutenseweg, the fraction of heavy traffic increased with 2 to 3%. In absolute terms there was a substantial decrease in middle and heavy traffic of 120 (Koningsweg), 211 (Haarlemmerweg) and 284 large vehicles per 24-hour (Stille Veerkade) after the policy implementation. Absolute increases in middle and heavy traffic were observed at the Hart van Brabantlaan (HVB) (106) and Vleutenseweg (294). Generally, the fraction of middle but not heavy traffic was somewhat lower with hand counts than with tube counts (Table 2).

The LEZ policies were designed to prohibit old trucks only. EURO-0 and EURO-I trucks almost disappeared in 2010. EURO-II and EURO-III trucks without retrofit were still present in 2010 (~18%), although officially forbidden (Table 3). The absolute decrease of EURO-0, I, II and III without retrofit trucks per day ranged from about 150 (HVB) to 530 per 24-hour (Haarlemmerweg) from baseline values of 240 and 650 trucks in 2008 (Supplement Table 4).

Pre and post intervention weather

Temperature, relative humidity, barometric pressure, precipitation and sunshine duration did not differ significantly between the 2008 and 2010 campaign (Supplement Table 5). Only wind speed was significantly lower in the 2010 campaign than in the 2008 campaign. Wind direction was predominantly south-southwest (180°-240°) in the 2008 campaign and more fluctuating in the post campaign. When we compare weather characteristics within the same round the difference in especially wind speed occurred in one round only (Round B: Utrecht, Den Bosch, Tilburg), illustrating the need for 1) correction of temporal variation with the use of the central reference location, and 2) the importance to adjust the 2010-2008 concentration differences for differences in wind speed. We did not have more specific meteorological data available, such as mixing height.

Table 2 Detailed traffic data of the different urban streets

Cities	Urban streets	Traffic intensity per 24-h					
		Tube			Manual		
		Pre	Post	%	Pre	Post	%
Amsterdam	Haarlemmerweg	15 253	15 314	0.4	15 347	14 204	-7.4
Amsterdam	Hoofdweg	9774	8375	-14.3	9434	8420	-10.7
The Hague	Stille Veerkade	17 438	8471	-51.4	14 082	8402	-40.3
Den Bosch	Brugstraat	17 896	18 170	1.5	18 623	18 572	-0.3
Den Bosch	Koningsweg	17 138	16 876	-1.5	16 429	16 937	3.1
Tilburg	HVB	18 812	19 010	1.1	18 517	20 037	8.2
Utrecht	Vleutenseweg	13 553	11 158	-17.7	10 836	10 653	-1.7
Utrecht	Weerdsingel Wz	14 831	15 045	1.4	14 569	14 569	0.0
Average		15 587	14 053	-9.8	14 730	13 974	-5.1

Cities	Urban street	Middle fraction[ab]				Heavy fraction[ac]			
		Tube		Manual		Tube		Manual (buses)	
		Pre	Post	Pre	Post	Pre	Post	Pre	Post
Amsterdam	Haarlemmerweg	0.03	0.03	0.04	0.03	0.02	0.02	0.02 (0.00)	0.02 (0.01)
Amsterdam	Hoofdweg	0.01	0.01	0.03	0.03	0.06	0.06	0.04 (0.03)	0.05 (0.04)
The Hague	Stille Veerkade	0.05	0.02	0.03	0.04	0.02	0.01	0.02 (0.00)	0.01 (0.01)
Den Bosch	Brugstraat	0.12	0.09	0.01	0.02	0.05	0.04	0.05 (0.02)	0.04 (0.03)
Den Bosch	Koningsweg	0.05	0.04	0.02	0.02	0.03	0.02	0.05 (0.03)	0.04 (0.03)
Tilburg	HVB	0.03	0.03	0.02	0.02	0.07	0.07	0.05 (0.04)	0.05 (0.04)
Utrecht	Vleutenseweg	0.06	0.04	0.03	0.03	0.05	0.05	0.11 (0.07)	0.14 (0.07)
Utrecht	Weerdsingel Wz	0.06	0.04	0.03	0.03	0.03	0.03	0.01 (0.00)	0.01 (0.01)
Average		0.05	0.04	0.03	0.03	0.04	0.04	0.04 (0.02)	0.04 (0.03)

[a]Fraction of total traffic intensity; [b]distance between wheel axes 3.5-7 m (tube); [c]distance between wheel axes >7 m (tube).

Table 3 Fraction middle and heavy traffic (e.g. trucks) in EURO-classes before[a] and after[b] the introduction of LEZ – city level

Cities	EURO-0		EURO-1		EURO-II - retrofit		EURO-II + retrofit		EURO-III - retrofit		EURO-III + retrofit		EURO-IV		EURO-V	
	Pre	**Post**	**Pre**	**Post**	**Pre**	**Post**	**Pre**	**Post**	**Pre**	**Post**	**Pre**	**Post**	**Pre**	**Post**	**Pre**	**Post**
Amsterdam	0.01	0.00	0.03	0.01	0.02	0.04	0.22	0.01	0.42	0.14	0.02	0.27	0.12	0.17	0.16	0.36
The Hague	0.02	0.00	0.05	0.01	-	0.05	0.27[c]	0.01	-	0.09	0.49[c]	0.29	0.09	0.14	0.04	0.39
Den Bosch	0.12	0.01	0.04	0.00	0.22	0.07	0.02	0.02	0.24	0.12	0.22	0.28	0.06	0.16	0.11	0.32
Tilburg	0.02	0.00	0.03	0.01	0.14	0.06	0.05	0.01	0.19	0.13	0.22	0.22	0.17	0.22	0.18	0.35
Utrecht	0.02	0.00	0.02	0.02	0.17	0.06	0.04	0.02	0.17	0.07	0.28	0.29	0.15	0.20	0.15	0.34
Average	0.04	0.00	0.04	0.01	0.14[d]	0.06	0.08[d]	0.01	0.26[d]	0.11	0.19[d]	0.27	0.12	0.18	0.13	0.35

[a]Before based on individual municipality data, and originated from different years before introduction of the LEZ (not particularly 2008); [b]After based on Goudappel[22] for the year 2010 (Table B2.1). Note that fraction does not necessarily add up to 1 because of rounding; [c]No distinction made with or without retrofit; [d]Average calculated excluding The Hague.

Pre and post intervention air pollution concentrations

Concentration differences at urban streets

For all pollutants concentrations were lower in 2010 than in 2008 (Table 4). For PM_{10}, $PM_{2.5}$ and NO_x differences were statistically significant. The 2010-2008 decreases did not differ significantly between the average of the major streets and the suburban background locations for the traffic-related pollutants soot, NO_2 and NO_x. This suggests no measurable effect from the implementation of the LEZ (Table 4). We also found no difference in 2010-2008 trend of PM_{10} - when averaged. We did find a substantial reduction in $PM_{2.5}$ concentrations, which was significantly larger (5.4 $\mu g/m^3$: 31%) at the urban streets than at the matching suburban background locations (2.7 $\mu g/m^3$: 20%). The $PM_{2.5}$ reduction at street locations was significantly larger than the urban background $PM_{2.5}$ reduction (3.9 $\mu g/m^3$: 27% (p-value: 0.08)) (Table 4).

There were remarkable differences between the observed changes in pollution concentrations pre and post the policies at individual urban streets. The most striking decrease in soot, NO_2 and NO_x was found at the Stille Veerkade, where apart from the LEZ other local traffic policies were implemented resulting in a substantial decrease in total traffic intensity. Soot concentrations dropped with 41% (1.74 10^{-5}/m) and NO_2 concentrations with 25% (13.4 $\mu g/m^3$), which was significantly more than the decrease at the corresponding suburban background (22 and 7%: 0.34 10^{-5}/m and 1.7 $\mu g/m^3$, respectively). On the other hand a 35% increase in soot (0.72 10^{-5}/m) was found at one street (Vleutenseweg) which was larger than the soot increase at the matching suburban location (16%: 0.22 10^{-5}/m). NO_2 but not NO_x also increased at that street compared to suburban background levels (Table 4). In this street the fraction of heavy traffic increased. Both streets in Amsterdam also showed decreases in especially soot and NO_x, though these changes were not significantly different from suburban background. Effects were stable after excluding the Stille Veerkade where multiple local traffic policies took place. Only NO_2 decreased significantly more at the suburban background than at the urban streets. The latter effect disappeared again when the trend was adjusted for differences in wind speed (Supplement Table 6).

A low wind speed during the measurements was related to a larger difference between the street and suburban background locations for all pollutants except for PM_{10} and $PM_{2.5}$. When trends were corrected for wind speed differences, all pollutants decreased more at the street compared to suburban decreases, but generally far from statistically different for all but $PM_{2.5}$ (Supplement Table 6), as observed in the uncorrected analysis presented in Table 4-5.

Total PNC was increased at the Vleutenseweg as well, but the trend did not differ from the suburban trend (Table 6). Particularly the total amount of smallest particles (16-30 nm) was more increased at the Vleutenseweg than at the matching suburban location, in line with the increase in soot and NO_2. At the Haarlemmerweg also the smallest particles increased more than at the suburban location, whereas the larger particles (100-470 nm) decreased significantly more than at suburban background (not significant with Wilcoxon

test). PNC data was not available for the other streets. Comparison of PNC with other air pollution indicators is limited, because of the relatively few measurement days available for analysis.

Elemental concentration differences at urban streets

Cr, Cu and Fe elemental concentrations derived from PM_{10} samples were not different, on average, between 2008 and 2010 (Table 5). The significant Fe increase at the streets compared to the average suburban increase after excluding the Stille Veerkade was due to an outlier value. Also no difference was found in Cr, Cu and Fe derived from $PM_{2.5}$ samples (data not shown).

On the individual street level there were substantial decreases in Cu and Fe found at the Stille Veerkade, which was significantly larger than suburban background decreases (not significant with T-tests). Also at the Haarlemmerweg a substantial decrease was found in Cu which was different from suburban trend. This was also visible in Fe. Again, at the Vleutenseweg, Cu and Fe were almost twice as high as before. Also at the HVB an increase in Cu and Fe was visible which was significantly higher than the matching suburban trend (Table 5). Apart from a decrease in sulphur (a major fraction of especially $PM_{2.5}$), no consistent difference was found in other measured elements that was different at the streets compared to suburban trend, and not already seen in urban background concentration differences.

Concentration differences at urban background

$PM_{2.5}$ decreased borderline significantly more (27%) at the urban background compared to suburban locations (20%) (p-value T-test; 0.10). This decrease in $PM_{2.5}$ at urban background was not significant when corrected for wind speed (Supplement Table 6).

Table 4 Average concentrations of different pollutants before (2008) and after (2010) introduction of the LEZ

Cities	Locations	PM_{10}			$PM_{2.5}$			Soot		
		Pre	Post	Abs dif	Pre	Post	Abs dif	Pre	Post	Abs dif
Amsterdam	Haarlemmerweg	27.5	26.2	-1.3#	17.8	12.3*	-5.5#	3.92	3.42	-0.50
Amsterdam	Hoofdweg	22.4	25.4	3.0	15.1	10.8*	-4.3	2.58	2.34	-0.24
The Hague	Stille Veerkade	32.2	27.9	-4.3	19.4	11.8**	-7.6	4.26	2.52**	-1.74##
Den Bosch	Brugstraat	32.9	25.2*	-7.7	18.0	12.0**	-6.0	3.66	3.43	-0.23
Den Bosch	Koningsweg	30.2	24.4**	-5.8	17.4	10.6**	-6.9#	2.84	2.34**	-0.50#
Tilburg	HVB	29.4	21.9**	-7.5	17.2	11.6**	-5.6	2.37	2.40	0.04
Utrecht	Vleutenseweg	25.9	24.1	-1.7	15.4	11.8**	-3.7	2.06	2.78**	0.72#
Utrecht	Weerdsingel Wz	29.1	28.0	-1.1	16.6	13.4**	-3.1	3.36	3.63	0.27
	Average	28.6	25.3**	-3.3	17.2	11.8**	-5.4##	3.10	2.85	-0.25
	Average[a] (N=7)	28.1	25.0**	-3.1	16.8	11.8**	-5.1##	2.93	2.89	-0.04
	Amsterdam UB	23.2	21.2	-2.1	13.3	9.5**	-3.9#	1.66	1.45**	-0.20
	The Hague UB	23.2	21.4	-1.8	14.1	10.3*	-3.7	1.67	1.39*	-0.28
	Den Bosch UB	26.6	21.4	-5.2	14.8	11.3*	-3.5	1.61	1.44	-0.17
	Tilburg UB	27.0	20.6**	-6.4	16.6	10.8**	-5.8	1.56	1.42	-0.14
	Utrecht UB	25.6	21.3**	-4.3	14.7	12.0**	-2.8	1.54	1.71	0.17
	Average	25.1	21.2**	-4.0	14.7	10.8	-3.9	1.61	1.48**	-0.13
	Amsterdam SUB	18.1	19.9	1.8	13.1	11.3	-1.8	1.49	1.38	-0.11
	The Hague SUB	23.5	18.9	-4.6	12.4	10.6	-1.7	1.58	1.23*	-0.34
	Den Bosch – Tilburg SUB	26.4	18.7**	-7.7	15.3	11.1**	-4.2	1.52	1.34	-0.18
	Utrecht SUB	21.5	18.6	-2.9	14.6	11.4*	-3.2	1.31	1.53	0.22
	Average	22.4	19.0	-3.3	13.8	11.1**	-2.7	1.48	1.37	-0.11

Table 4 Continued

Cities	Locations	NO_2			NO_x		
		Pre	Post	Abs dif	Pre	Post	Abs dif
Amsterdam	Haarlemmerweg	54.3	50.9	-3.4	100.5	83.3	-17.1
Amsterdam	Hoofdweg	47.5	41.4	-6.1	75.6	61.5	-14.1#
The Hague	Stille Veerkade	54.1	40.7*	-13.4##	109.9	70.4*	-39.5#
Den Bosch	Brugstraat	53.9	53.3	-0.5	99.3	104.6	5.2
Den Bosch	Koningsweg	44.2	39.3	-4.9	78.3	56.7**	-21.6##
Tilburg	HVB	36.6	37.9	1.3##	60.6	55.5	-5.1
Utrecht	Vleutenseweg	41.7	44.1	2.5##	70.3	66.7	-3.6
Utrecht	Weerdsingel Wz	52.4	52.7	0.3	87.7	91.5	3.8
	Average	48.1	45.1	-2.9	85.3	73.8**	-11.4
	Average[a] (N=7)	47.2	45.7	-1.5#	81.8	74.3	-7.5
	Amsterdam UB	34.4	30.7*	-3.7	49.1	43.6	-5.4
	The Hague UB	32.1	28.1	-4.0	52.4	41.5	-10.9
	Den Bosch UB	27.6	25.7	-1.9##	42.4	35.8**	-6.5
	Tilburg UB	31.6	27.1**	-4.5	45.9	34.4**	-11.5
	Utrecht UB	34.2	31.3	-2.9##	48.8	44.5	-4.2##
	Average	32.0	28.6	-3.4	47.7	40.0**	-7.7
	Amsterdam SUB	25.5	22.0*	-3.5	35.2	34.7	-0.5
	The Hague SUB	24.3	22.6	-1.7	39.8	34.5	-5.3
	Den Bosch – Tilburg SUB	23.0	18.5**	-4.5	33.1	25.5*	-7.6
	Utrecht SUB	30.2	21.8**	-8.4	45.3	34.3**	-10.9
	Average	25.8	21.2**	-4.5	38.3	32.3**	-6.1

Table 5 Average concentrations of Cr, Cu and Fe from PM_{10} filters before (2008) and after (2010) introduction of the LEZ

Cities	Locations	Cr			Cu			Fe		
		Pre	Post	Abs dif	Pre	Post	Abs dif	Pre	Post	Abs dif
Amsterdam	Haarlemmerweg	5.0	5.7	0.7	62.5	47.6*	-14.9#	1293.5	1080.6	-212.9#
Amsterdam	Hoofdweg	3.5	4.0	0.5	28.0	27.9	-0.1	738.4	745.8	7.4
The Hague	Stille Veerkade	7.3	4.7	-2.6	56.2	29.0*	-27.2	1405.9	913.0	-492.9
Den Bosch	Brugstraat	10.0	6.9	-3.1	47.5	53.3	5.8	1008.1	1292.3	284.2
Den Bosch	Koningsweg	6.4	4.3**	-2.1	35.9	29.0	-6.9	834.5	757.6	-76.9
Tilburg	HVB	6.6	5.8	-0.8	33.4	46.0*	12.6#	660.5	1024.5**	364.0#
Utrecht	Vleutenseweg	5.0	4.0	-1.0	19.1	37.4**	18.3##	365.6	816.1**	450.5##
Utrecht	Weerdsingel Wz	7.0	6.7	-0.3	41.0	52.2	11.2	924.2	1256.2	332.0
	Average	6.3	5.2	-1.0	39.8	40.3	0.5	893.1	981.4	88.3
	Average[a] (N=7)	6.1	5.3	-0.8	37.4	41.7	4.3	816.2	989.8**	173.6#
	Amsterdam UB	1.5	1.9	0.4	11.3	12.2	0.9	569.6	385.9	-183.7
	The Hague UB	1.6	2.3	0.7##	14.5	11.6	-2.9	474.6	375.6	-99.0
	Den Bosch UB	4.3	2.4**	-1.9	14.4	11.2**	-3.2	323.7	361.6	37.9
	Tilburg UB	3.7	2.4*	-1.3	11.4	10.7	-0.7	220.4	312.0*	91.6
	Utrecht UB	3.7	2.4**	-1.3	12.3	13.6	1.3	326.6	425.4	98.8
	Average	2.9	2.3	-0.6	12.7	11.9	-0.8	385.0	372.1*	-12.9
	Amsterdam SUB	0.8	1.2	0.4	9.5	9.4	-0.1	344.9	267.7	-77.2
	The Hague SUB	5.1	1.2**	-3.9	6.9	8.6	1.7	295.5	245.4	-50.1
	Den Bosch – Tilburg SUB	3.5	2.1**	-1.4	8.7	7.4	-1.3	178.5	246.7	68.2
	Utrecht SUB	3.7	2.0**	-1.7	9.1	9.8	0.7	182.1	270.0**	87.9
	Average	3.1	1.6	-1.5	8.7	8.8	0.1	246.1	257.5**	11.4

Concentrations are average concentrations adjusted for temporal variation using data from the central reference location. Air pollutants in $\mu g/m^3$ apart from soot ($10^{-5}/m$) and Cr, Cu and Fe (ng/m^3). Overall averages calculated from individual samples. UB=urban background. SUB=suburban background.

[a]After excluding the Stille Veerkade where multiple traffic policies were implemented.

*Significant difference pre and post at the 0.10 level.

**Significant difference pre and post at the 0.05 level.

#Significant difference pre and post between street and matching suburban location at the 0.10 level.

##Significant difference pre and post between street and matching suburban location at the 0.05 level.

Significance was tested with T-tests.

Table 6 Average PNC (p/cm^3) before (2008) and after (2010) introduction of the LEZ

Locations	16-30 nm			30-100 nm			100-470 nm			Total		
	Pre	Post	Abs dif	Pre	Post	Abs dif	Pre	Post	Abs dif	Pre	Post	Abs dif
Haarlemmerweg	1257	5009**	3752##	10 727	9651	-1076	4207	2919**	-1288#	16 191	17 579	1388
Vleutenseweg	1540	5009**	3469##	6365	8612	2247	2538	2790	252	10 443	16 410**	5967
Amsterdam SUB	1054	1371	317	3574	4102	528	2211	1789	-422	6839	7263	424
Utrecht SUB	1098	2047**	949	3661	5448**	1787	1851	2445	594	6611	9941**	3330

Concentrations are average particle number concentrations (p/cm^3) and can be compared within the same sampling round only (Haarlemmerweg with matching Amsterdam SUB, and Vleutenseweg with Utrecht SUB). SUB=suburban background.

*Significant difference pre and post at the 0.10 level.

**Significant difference pre and post at the 0.05 level.

#Significant difference pre and post between street and matching suburban location at the 0.10 level.

##Significant difference pre and post between street and matching suburban location at the 0.05 level.

Significance was tested with T-tests.

Discussion

The LEZ policies directed at heavy duty vehicles (trucks) did not substantially change concentrations of traffic-related pollutants (e.g. soot, NO_2, NO_x, Cu, Fe) at street sites more than at suburban background sites outside of the LEZ selected as control locations. Street and urban background $PM_{2.5}$ concentrations did decrease more than suburban background $PM_{2.5}$. In one street - where in addition to the LEZ- traffic intensity was reduced, soot, NO_x and NO_2 concentrations decreased significantly more than at the corresponding suburban background location.

Lack of a LEZ effect on traffic-related air pollutants

At baseline, concentrations of Cr, Cu and Fe, soot and PNC were 2-3 times higher at the busy streets of this study than at the urban background locations[18]. After the LEZ introduction, these differences were very similar. The changes over time were much smaller at all three site types (street, urban and suburban background locations) than the contrasts between them. It is well-documented that proximity to traffic is much better reflected in contrasts in particle number concentrations (PNC) or soot than in PM_{10} and $PM_{2.5}$ levels[16]. We therefore hypothesised that the LEZ would affect traffic-related components like soot and NO_x more than the more general air quality indicators PM_{10} and $PM_{2.5}$. We did not find a LEZ effect on these traffic-related indicators. There can be various explanations for this: (1) the impact of the LEZ may have been too small to measure. The LEZ only affects trucks and not private cars and vans. The absolute decrease in the number of old trucks was relatively small. The LEZ further affects only a small portion of the city, though the actual area affected is larger than the formal LEZ, typically located in the wide city centre; (2): recent data suggest that differences in emissions of especially NO_x between the various EURO-classes are much smaller than anticipated when the policy was formulated[24]. Especially real-world emissions of EURO-V trucks are about a factor 3 higher than initially estimated (Supplement Table 2). This seems also be the case for real-world emissions of PM[25]; (3): some LEZ effect may have been present already in the baseline measurements, since the LEZ was gradually implemented from July 2007 to October 2008. However, this is unlikely a major issue, since regulations of which types of trucks were allowed became more stringent and enforcement was tightened after baseline; (4): another potential reason is that the actual effect of the LEZ may counteract or coincide by other (policy) changes over time which increases traffic-related air pollutants. We asked local governments about specific traffic policies implemented, which could potentially affect street as well as urban background (Table 1). This list basically includes not much more than the LEZ apart from the traffic circulation plan in The Hague, in addition to city-level policies such parking costs and stimulation of cycling. A possible change over time includes the relative increase of new diesel cars in the average car park, causing rising

emissions of especially soot. The introduction of LEZ can cause side effects in itself as well. It is known that retrofitting vehicles cause higher primary NO_2 emissions[26]; (5): because of the economic crisis that hit the Netherlands in 2008, pollution emissions may have decreased everywhere, making it even harder to detect a small LEZ-related change; (6): our sampling campaign may have been too short to detect small changes related to the LEZ as significant; (7): different subtle weather conditions may have blurred a LEZ effect. We minimised weather effects since we measured each street with its matching urban and suburban location simultaneously. In addition we corrected street-suburban trends for differences in wind speed, and results were similar. That wind speed decreases concentrations at street locations relatively more than concentrations at background locations have been reported before[27], which basically is the result of greater dilution in pollutants that are produced locally (the more traffic-related components).

LEZ effect on $PM_{2.5}$?

We found a higher reduction in $PM_{2.5}$ between 2008 and 2010 at both the urban streets as well as the urban background locations compared to suburban background locations. Since we did not find a LEZ effect on other more traffic-related indicators (e.g. soot), and that the $PM_{2.5}$ reductions were almost as large at the matching urban background locations, it is not very likely that this reduction reflects an effect of the LEZ. Elemental composition data from the $PM_{2.5}$ fraction did not provide an explanation.

We may have missed some important traffic-related indicator within the $PM_{2.5}$ fraction. Although we did measure elemental carbon (EC) as absorbance (soot), we were not able to measure organic carbon (OC) including quinones and polycyclic hydrocarbons in this study. OC may originate from a wide range of sources, including combustion processes (including wood combustion), re-suspension of road dust and secondary organic aerosol, and is therefore not that traffic-specific. Total amount of OC within the $PM_{2.5}$ fraction can be quite substantial (up to 11%)[28]. Although the fact that OC is potentially a substantial fraction of the $PM_{2.5}$, this could not explain the reported large reductions in $PM_{2.5}$ solely (an extra reduction of 2.6 ug/m^3 in $PM_{2.5}$ compared to suburban background).

The $PM_{2.5}$ reduction coincided with a substantial elemental reduction in sulphur at the urban streets. The reduction in sulphur is probably not due to the LEZ, since sulphur originates mostly from long-range (transboundary) transport of multiple pollution sources.

Previous evaluations on LEZ

Many cities implemented congestion charging or LEZ including London, Stockholm and Rome. Those zones are directed to trucks, buses, all old vehicles, or all vehicles entering the inner-city[2]. There are a few evaluation studies on LEZ or similar zones, often based on model predictions instead of measurements. Johansson et al.[11] modelled rooftop concentration reductions

in the zone up to 7 and 12% for PM_{10} and NO_x along four densely trafficked streets in Stockholm compared to a baseline scenario without the zone. In Rome, traffic is restricted in the inner-city. Cesaroni et al.[12] modelled reductions between 10 and 23% in PM_{10} and NO_2 concentrations in the most affected areas of Rome. Tonne et al.[10] estimated the impacts of the congestion charging zone introduced in the inner-city of London, UK, which charges for travel into central London and reduced traffic volumes considerable, and air pollution concentration reductions were estimated to be on average 0.2 $\mu g/m^3$ for PM_{10} and 0.7 $\mu g/m^3$ for NO_2. In a more recent extensive evaluation of the congestion charging zone, including measured data, effects remained relative small[13,14]. Within the zone, they modelled a net decline of 2.2 $\mu g/m^3$ in annual average NO_x concentration and a decline of 0.8 $\mu g/m^3$ in PM_{10}. In addition modelled NO_2 was slightly increased (0.6 $\mu g/m^3$), probably due to an increase in particle traps in diesel buses at the same time. When they made use of measured data, no policy related change was seen in the road-site monitors, where effects would be most pronounced. The effects of the policy were more evident on the few urban background monitor locations within the zone compared to control areas. PM_{10} and NO concentrations declined with about 10 to 35%, whereas NO_2 increases with 2 to 20%[13]. In Kopenhagen, Denmark, the LEZ directed to old trucks and buses was evaluated with the use of measurements along one busy street and one urban background site. An extra reduction was seen in $PM_{2.5}$ of about 0.7 $\mu g/m^3$ (5%), compared to background trend. Effects on other pollutants including NO_x and EC were smaller[29]. In Germany 41 cities implemented LEZ banning old vehicles (both cars and trucks) from city centres[2], but results on the effects were not published yet.

Modelling studies are hampered by assumptions about for example emission factors which may not be correct, as in the present study. Policy evaluation by measurements will encounter difficulties as well, particularly by other developments unrelated to the policy. Examples in our study included construction work in the neighbourhood of the The Hague street and an increase in the fraction of heavy traffic in one of the Utrecht streets. By design, in intervention studies the key challenge is to distinguish between subtle air pollution changes in time due to the policy and other time-varying factors, such as meteorological conditions or other more generic policies affecting larger areas which can influence air pollution concentrations as well[3]. One method to capture this partly is the inclusion of proper control areas not affected by the policy under investigation. In our study we included control locations not affected by the LEZ. It is not straightforward to select proper control areas. In our study the ideal control would have been major streets in cities not affected by the LEZ. As the LEZ was a policy applied in many Dutch cities, these streets were not easily available.

Conclusion

The LEZ policies directed at old heavy duty vehicles (trucks) did not substantially change concentrations of traffic-related pollutants (e.g. soot, NO_2, NO_x, Cu, Fe) at street sites more than at suburban background sites outside of the LEZ. Old trucks are only a small part of the average car park. Effects of the LEZ were too modest to produce significant decreases in traffic-related air pollution concentrations. Street and urban background $PM_{2.5}$ concentrations did decrease more than suburban background $PM_{2.5}$, which could be also related to other changes over time. In one street - where in addition to the LEZ - traffic intensity was reduced, soot, NO_x and NO_2 concentrations decreased significantly more than at the corresponding suburban background location.

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Supplement

Supplement Table 1 EU Emission standards for heavy duty diesel engines (>3.5 tonnes) (g/kWh)

	EURO-0	EURO-I	EURO-II	EURO-III	EURO-IV	EURO-V
PM	-	0.61 / 0.36	0.25 / 0.15	0.13 / 0.10	0.02	0.02
NO_x	-	8.0	7.0	2.0 / 5.0	3.5	2.0

Derived from http://www.dieselnet.com/standards/eu/hd.ph.

Supplement Table 2 Estimated NO_x emission factors (g/km) for EURO-0 to EURO-V heavy duty trucks (>20 tonnes) under different conditions[a]

	EURO-0	EURO-I	EURO-II	EURO-III	EURO-IV	EURO-V
Estimated from EURO standards	20	14.5	15.5	12	7	4.5
New measurements under real-world conditions	21	13.5	14.5	15.5	15	15

[a]Data is estimated from Figure 2 in Velders et al.[24] Emission factors are estimated from the reduction steps in EURO emission standards and those based on new measurements that were carried out under real-world driving conditions along city streets. The emission factors relate to the average type of trucks and average driving conditions in the Netherlands.

Supplement Table 3 Limits of detection[a] (LOD) and coefficients of variability (CV) of the various air pollutants

	PM_{10}	$PM_{2.5}$	Soot	NO_2	NO_x	Cr	Cu	Fe
LOD (3*SD) Pre[b]	3.5	0.5	0.2	3.6	10.1	0.5	0.8	3.1
LOD (3*SD) Post[c]	8.1	1.9	0.09	2.2	8.9	1.1	1.5	13.4
Laboratory LOD Pre	-	-	-	-	-	0.5	0.7	0.8
Laboratory LOD Post	-	-	-	-	-	0.1	0.1	0.1
% >LOD Pre	100	100	100	100	99	100	100	100
% >LOD Post	99[d]	100[d]	100	100	100	96	100	100
CV[e] (%) Pre	5.6 (10)	7.2 (9)	6.8 (9)	4.1 (21)	9.8 (21)	25.3 (10)	11.1 (10)	7.7 (10)
CV[e] (%) Post	1.3 (8)[f]	4.2 (8)[g]	1.5 (10)	4.6 (31)	6.7 (31)	35.1 (9)	7.1 (10)	6.7 (10)

[a]LODS of PM_{10}, $PM_{2.5}$ and NO_x in µg/m^3. Soot in 10^{-5}/m. Elements in ng/m^3 and derived from PM_{10} filters; [b]There were 6 field blanks for both PM_{10} and $PM_{2.5}$ and 19 (1 unexplained outlier excluded) for NO_x; [c]There were 6 field blanks for PM_{10} and 5 field blanks for $PM_{2.5}$ (1 PM_{10} and 1 $PM_{2.5}$ filter excluded because of wrong pre-weightings) and 20 for NO_x; [d]Percentages are reported after deleting sampling week 1 because of wrong pre-weightings; [e]CV values has been calculated as the square of the absolute differences of the measurements. Then the root of the sum of the values is divided by two times the number of duplicate measurements; [f]Two duplicate filters excluded because of wrong pre-weightings in sampling week 1; [g]One duplicate filter excluded because of wrong pre-weightings in sampling week 1.

Supplement Table 4 Estimated absolute amount of total and old trucks per 24-hour

Cities	Urban streets	Abs amount of trucks per 24-h[a]			Abs amount of old trucks per 24-h[b]		
		Pre	Post	Abs dif	Pre	Post	Abs dif
Amsterdam	Haarlemmerweg	921	568	-353	645	114	-531
Amsterdam	Hoofdweg	377	337	-40	264	67	-197
The Hague	Stille Veerkade	704	336	-368	486	54	-432
Den Bosch	Brugstraat	745	557	-188	477	123	-354
Den Bosch	Koningsweg	657	508	-149	421	112	-309
Tilburg	HVB	556	601	45	239	90	-149
Utrecht	Vleutenseweg	759	1065	306	387	202	-184
Utrecht	Weerdsingel Wz	583	437	-146	297	83	-214

[a]Absolute amount of trucks are estimated by combining the fraction middle and heavy traffic (without buses) with total traffic derived from manual counts;
[b]Old trucks defined as EURO-0, I, II and III without retrofit. Data on EURO-classes derived from city level data, see Table 3 main article.

Supplement Table 5 Averages of different meteorological variables during measurement weeks (N=12)

Meteorological variables				Round A (Amsterdam, The Hague)			Round B (Utrecht, Den Bosch, Tilburg)		
	Pre	Post	Abs dif	Pre	Post	Abs dif	Pre	Post	Abs dif
Average temperature (°C)	8.5	9.2	0.7	6.9	8.8	1.9	10.2	9.6	-0.6
Minimum temperature (°C)	5.0	5.3	0.3	3.2	5.3	2.1	6.8	5.4	-1.4
Maximum temperature (°C)	11.8	12.7	0.9	10.2	12.2	2.0	13.3	13.2	-0.1
Average relatively humidity (%)	85.2	85.8	0.6	86.5	84.2	-2.3	84.0	87.4	3.4
Average air pressure (hPa)	1010.8	1013.6	2.8	1011.2	1013.2	2.0	1010.5	1014.1	3.6
Prevailing wind direction[a] (weeks)									
NNE (0°-60°)	-	-	-	-	-	-	-	-	-
E (60°-120°)	2	2	-	2	2	-	-	-	-
SSE (120°-180°)	-	1	-	-	-	-	-	1	-
SSW (180°-240°)	6	2	-	1	1	-	5	1	-
W (240°-300°)	1	1	-	-	-	-	1	1	-
NNW (300°-360°)	-	2	-	-	-	-	-	2	-
Fluctuating wind direction	3	4	-	3	3	-	-	1	-
Average wind speed (m/s)	3.6	3.2	-0.4	3.2	3.5	0.3	4.0	2.8**	-1.2
Maximum hourly wind speed (m/s)	5.4	4.7*	-0.7	5.0	5.0	0	5.8	4.3**	-1.5
Maximum wind gust (m/s)	9.9	8.7*	-1.2	9.2	9.3	0.1	10.7	8.1**	-2.6
Sum precipitation duration (hr)	18.9	15.0	-3.9	13.9	18.1	4.2	23.8	11.8	-12.0
Sum precipitation amount (mm)	24.9	13.9	-11.0	19.9	17.7	-2.2	29.8	10.2*	-19.6
Sum sunshine duration (hr)	29.4	24.7	-4.7	30.3	26.6	-3.7	28.4	22.8	-5.6

[a]Prevailing wind direction was classified in one of the sectors if the wind came for half the time or more from that direction. Otherwise, it was classified as fluctuating wind direction; *Significant difference pre and post at the 0.10 level; **Significant difference pre and post at the 0.05 level; Significance was tested with T-tests.

Supplement Table 6 Results of 2010-2008 air pollution concentration trends after wind speed adjustment

	Unadjusted			Adjusted for wind speed		
	Estimate	**SE**	**P-value**	**Estimate**	**SE**	**P-value**
Urban streets and matching suburban background locations						
PM_{10}	0.44	0.88	0.62	-0.58	0.94	0.54
$PM_{2.5}$	-2.42	0.70	0.00	-2.82	0.75	0.00
Soot	-0.11	0.17	0.50	-0.29	0.18	0.11
NO_2	2.04	1.83	0.27	-0.60	1.95	0.76
NO_x	-4.89	5.22	0.35	-11.37	5.62	0.05
Cr	-0.17	0.51	0.74	-0.89	0.55	0.11
Cu	1.77	3.47	0.61	-4.80	3.60	0.19
Fe	84.04	72.68	0.25	-54.14	75.29	0.47
After excluding one street (Stille Veerkade)						
PM_{10}	0.59	0.86	0.49	-0.54	0.94	0.56
$PM_{2.5}$	-1.79	0.56	0.00	-1.97	0.62	0.00
Soot	0.005	0.17	0.98	-0.15	0.19	0.44
NO_2	3.79	1.92	0.05	1.26	2.19	0.57
NO_x	-0.98	5.42	0.86	-7.45	6.21	0.23
Cr	-0.01	0.52	0.99	-0.66	0.59	0.26
Cu	4.69	3.48	0.18	-1.30	3.82	0.73
Fe	124.09	71.53	0.09[a]	-3.78	78.23	0.96
Urban background locations and matching suburban background locations						
PM_{10}	-0.45	0.98	0.65	-1.14	1.06	0.29
$PM_{2.5}$	-0.82	0.50	0.10	-0.76	0.54	0.17
Soot	-0.01	0.05	0.91	-0.02	0.06	0.80
NO_2	1.12	0.83	0.18	0.43	0.90	0.64
NO_x	-1.34	1.60	0.40	-2.31	1.74	0.19
Cr	0.23	0.26	0.39	-0.12	0.28	0.66
Cu	-0.20	0.69	0.78	-0.51	0.78	0.51
Fe	-23.46	35.90	0.52	-71.80	38.45	0.07[b]

[a]After deleting one outlier value: estimate = -104.3, not significant (p-value: 0.14); [b]After deleting one outlier value: estimate = -17. 64, not significant (p-value: 0.48).

Chapter 6

Respiratory effects of a reduction in outdoor air pollution concentrations

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Abstract

Background
Air pollution has been associated with respiratory health effects. There is little direct evidence that reductions in air pollution related to abatement policies lead to actual improvement in respiratory health. We assessed whether a reduction in (traffic policy-related) measured air pollution concentrations was associated with changes in respiratory health.

Methods
Air pollution concentrations and respiratory health were measured in 2008 and 2010 at eight busy urban streets and at four suburban background control locations. Respiratory function was assessed twice in 661 residents by spirometry and measurements of airway resistance. NO in exhaled air was measured as a marker for airway inflammation.

Results
Air pollution concentrations were lower in 2010 than in 2008. The concentration declines varied between locations, with the largest decline observed in a street with a large reduction of traffic intensity. In regression analyses adjusted for important covariates, there were significant associations between decreases in concentrations of soot, NO_2, NO_x, Cu and Fe and increases in FVC (~1% increase per IQR decline). Airway resistance decreased with a decline in PM_{10} and $PM_{2.5}$ (9% per IQR), although somewhat less consistent. No associations were found with exhaled NO. Results were largely driven by one street where traffic-related air pollution showed the largest reduction. FEV_1 and FVC improved with 3 to 5% in residents of this street compared to suburban background residents, accompanied with a suggestive reduction in airway resistance.

Conclusion
Reductions in air pollution may lead to small improvements in respiratory function.

Introduction

Over the past decades, numerous studies have documented adverse respiratory health effects of traffic-related air pollution[1]. Respiratory effects are not limited to vulnerable subgroups including children and elderly, but have been found in healthy adult populations as well[2]. Most evidence is based on cross-sectional comparisons and cohort studies that investigated association between (changes in) health status within individuals and baseline air pollution[2]. Only a few studies specifically investigated the effects of changes in air pollution over time and changes in respiratory health within individuals. Downs et al.[3] reported that a reduction of 10 $\mu g/m^3$ PM_{10} over 11 years of follow-up slowed the annual rate of FEV_1 decline by 9% in Swiss adults. In a subsample of the large Children's Health Study[e.g.4] of children who moved away, Avol et al.[5] showed an improvement in lung function growth among those who moved to cleaner areas within one to three years.

Standard health impact assessment of air pollution abatement policies[e.g.6,7] relied on existing concentration-response functions from observational studies and often modelled exposure changes, thus making implicit assumptions that causal effects will apply 'in reverse' when exposure is reduced. Since the striking results of Clancy et al.[8] that after the Dublin coal ban, both air pollution and death rates dropped substantially, interest in intervention type of studies is increasing. The US-based Health Effects Institute (HEI) has promulgated a programme to investigate the effects of interventions[9]. Studies include the traffic policies taken at the Summer Olympic Games in Atlanta[10], and the sale ban of coal in multiple Irish cities[11].

Intervention studies have the potential to give more direct evidence than observational studies, and therefore may contribute substantially to the causality debate. Moreover because of the high costs of many air pollution abatement policies, questions have arisen about the public health gains of these policies[9]. Intervention studies are challenging, particularly to disentangle potential effects of a policy from autonomous trends, and to have sufficient contrast in exposure.

We performed a study to evaluate the air quality and health effects of local traffic policies including low emission zones (LEZ) directed at heavy duty vehicles in several Dutch cities. The study includes comprehensive measurements of air quality and population respiratory health at street level before (2008) and two years after the implementation of the policies (2010). A detailed description of the policies and effects of local policies on air pollution concentrations have been reported separately[12]. In this paper we assessed whether changes in air pollution concentrations are related to changes in respiratory health status within two years time.

Methods

Study design

Measurements of air pollution and respiratory health were conducted at 12 locations in the Netherlands before (2008) and two years after the policy implementations (2010).

In all cities the LEZ was evaluated, directed at forbidding old heavy duty vehicles to enter the LEZ, typically the city center. In one city, additionally a traffic recirculation plan designed to reduce concentrations at hotpots was evaluated. Measurements were done at eight busy urban streets in five Dutch cities and four suburban background locations near the selected cities. Suburban background locations were chosen as control areas, likely not affected by the policy under investigation. Respiratory function was assessed twice by spirometry and interrupter airway resistance. The interrupter airway resistance was added because it required minimal cooperation and can therefore be used in young children as well[13]. In addition, NO in exhaled air was measured as a marker for airway inflammation.

We reported before that with the exception of one urban street where traffic flows were drastically reduced, there was no difference in 2010-2008 concentration trend at the urban streets compared to the suburban background trend[12]. Since considerable variability in air pollution reductions was present, we focused on the association of respiratory health changes and concentration changes. We additionally analysed the street with the large change in air pollution separately.

The study has been approved by the Medical Committee of Utrecht University Medical Centre. Signed informed consent forms were obtained from all volunteers or their parents (children).

Study population

All residents of the specific streets were invited to participate by a letter and a reminder. We include both children (aged>4 years) as well as elderly since they may be particularly susceptible. Age of four was chosen as those children would be able to perform at least one respiratory health test (interrupter airway resistance) adequately. For the spirometry test children had to be at least six years of age. No other in- or exclusion criteria were formulated.

Assessment of exposure to air pollution

PM_{10}, $PM_{2.5}$, soot, NO_2, NO_x and elemental composition of PM was measured at eight urban streets in five cities and four suburban background locations (one of which was used as a comparison location for two nearby cities). The four suburban background locations were in villages (~30 000 inhabitants) near the selected cities (10-30 km). An additional background location was selected as a reference location to adjust for temporal variation, since measurements were divided in two rounds because of logistical reasons. All streets within the

same city and the nearby suburban location were measured simultaneously in the same sampling round. At each location, six weekly samples were collected spread over two six-month periods in 2008 and 2010 respectively, with identical equipment and at exactly the same location. Temporally adjusted average concentrations were used in subsequent analyses[12].

Briefly, PM_{10} and $PM_{2.5}$ were collected gravimetrically on Teflon filters using PM_{10} personal samplers (MSP Corp., Shoreview, MN, USA) and $PM_{2.5}$ GK2.05 cyclones (BGI Inc., Waltham, MA, USA). Soot content of all PM_{10} filters was measured using a Smoke Stain Reflectometer (model M43D; Diffusion Systems, London, UK) and transformed into absorption coefficients. All filters were analysed with energy dispersive X-ray fluorescence spectrometry (ED-XRF) at Cooper Environmental Services (Portland, OR, USA). We focused in this paper on those elements which showed the largest contrast between streets and background locations in the 2008 baseline campaign (Cr, Cu, Fe). NO_2 and NO_x concentrations were measured with OGAWA passive samplers (OGAWA & Company Inc., FL, USA). Sampling and analysis methods have been published before[14].

Exposure was measured in the same street where participants lived. The maximal distance between exact sample location and residents' home was about 500 m.

Traffic intensity at the busy streets ranged between 10 000 and 19 000 vehicles per 24-hour, and at follow-up did not differ more than 1000 vehicles per 24-hour, apart from one urban street (Stille Veerkade) where traffic reduced with about 50% (baseline traffic: 17 000 vehicles per 24-hour). Traffic at the suburban locations was only modest (<1250 vehicles per 24-hour) and did not differ more than 300 vehicles per 24-hour at follow-up.

Respiratory health measurements

Baseline and follow-up tests were performed in January-February 2009 and December-March 2011, following the majority of the exposure measurements. Tests were performed within two to four measurement days per location at the same place and close to where the people lived. For the follow-up of a few people (8%) who were unable to attend otherwise, we performed the tests at their home.

FVC, FEV_1 and PEF were measured using the same EASYONE Spirometer (ndd Medical Technologies, Zurich, Switzerland). Calibration was checked daily. Tests were performed according to European Respiratory Society criteria[15]. The majority of the tests (80%) were carried out by the same technician. The fractional concentration of exhaled NO was measured using the NIOXMINO (Aerocrine AB, Solna, Sweden) as a marker of airway inflammation. Exhaled NO tests were performed before spirometry, as it has been suggested that forced expiration may influence exhaled NO levels[16]. Four devices were used, three for the baseline (98% with the same device), and one for the post examination. Airway resistance was measured using the MicroRint (Micro Medical Ltd, Kent, UK), which used the interrupter technique[13]. Rint was measured during expiration, with occlusion of the airway at peak expiratory

flow, and occlusion occurred randomly. Rint values were excluded when instructions were not followed (e.g. no tidal breathing, leakage, hampered expiration) or when interruption was not at the peak of expiratory flow (visual check). Median values of five to ten approved manoeuvres were used in the analyses. We aimed at using one device for all measurements. However, during the baseline measurements the initial device broke down after three measurement days, another device gave unreliable flow checks after four measurement days and therefore another (new) device was used. For the follow-up, only the latter device was used.
Study participants filled in a questionnaire at baseline and at follow-up on prevalence of respiratory symptoms and on important confounder data including detailed smoking history and housing characteristics. Questionnaires were filled in by a parent under the age of twelve at baseline.

Data analysis
Broadly, the same data analysis was performed as in the study of Downs et al.[3] Absolute change in health outcome between the post and the pre campaign was the dependent variable, standardised for the exact follow-up time for each participant. Absolute change in average air pollution concentration (2010-2008) was the independent variable. We performed separate analyses for all pollutants. Slope and standard errors (SE) were multiplied by the interquartile range (IQR) and divided by mean baseline value and then multiplied by 100 to obtain a percentage change. We specified two-pollutant models when possible given the often high correlation between pollutants.
Covariates selected a priori in our basis models included age, gender, amount of cigarettes, level of education, having a cold at the time of examination, and the difference of the exact time of examination to account for diurnal fluctuation in respiratory health. Under the age of twenty-five, the highest level of education of the parents was used. We tested how to control for some of those covariates with the use of the Akaike Information Criterion (AIC). We especially assessed how to account for different time trends related to age, given that our study population included both children and adults. For spirometry, the best model fit included age in four categories (<=12, 13-17, 18-29, 30+ yrs). For the Rint two linear splines over the age intervals <=12 and older than 12 years gave the best model fit. Adding more splines (e.g. 12-17, 18-29, 30-64, 65+) did not improve model fit. For the exhaled NO age as a continuous variable provided the best model fit. These models were fit without air pollution in the model. We included the difference in height as a continuous variable as well in the spirometry models. For the Rint analyses we corrected for device and technician at follow-up as well in the basis models. We did not correct for technician in spirometry, since the majority of the tests were performed by one technician. Correction for technician led to unstable results, because the different technicians visited only one or a few locations. Also the different devices to measure exhaled NO were not evenly distributed over all locations. In sensitivity analyses we restricted the analysis

to tests performed by one technician (spirometry), or with the same device (exhaled NO).
In our extensive models we additionally included smoking status (yes, former, never), passive smoking, carpet in the bedroom, animals in the home, gas cooking, mould in the home and occupational exposure to gases (all 0/1 variables). In addition we accounted for the difference in temperature and ambient background NO_2 concentration between the two examinations to take into account short-term effects of either weather and air pollution. These two variables were 24-hour averages of the day before the examination took place and were taken from the nearest fixed site monitor.
For the Rint extensive models we included technician at baseline as well. All covariates were derived from the baseline questionnaire. We observed that most covariates did not change much within the two year time period. Sensitivity analysis was done excluding participants who changed smoking status (N=26) between the two examinations. Additional subgroup analyses were: excluding children under the age of eighteen because of small sample size (N=54) and excluding influential observations (1% of observations with the highest Cooks distance). Finally we analysed the one street in The Hague with the largest air pollution concentration reduction and suburban background locations separately.
We did not include baseline air pollution in the models, since the study period was relatively short (two years) and the correlation between baseline and changes in air pollutants was often high.
We used simple regression models instead of mixed-model regression analyses with an adjustment for clustering of residuals within locations since the latter did not give a better model fit based upon AIC. Effect estimates were considered significant when p-values were below 0.05.

Results

In total 853 people agreed to participate in the study in 2008. The response rate was low (~10%), and variable across locations. We have no information about the non-responders. 746 (87%) participants completed baseline examination. The most important reason for early drop-out was that they were not available at the baseline examination days. 661 participants (89%) were followed-up two years after baseline examination. Reasons for later drop-out include 27 persons moved, 9 persons unable to reach, 3 persons died, 15 persons unable to participate due to severe sickness, and 31 persons unwilling to participate any further. In total 640 (97%, exhaled NO), 585 (89%, spirometry) and 497 (75%, Rint) subjects were available for analysis after data cleaning. Most missing values were present in Rint analyses since data from two measurement days (71 participants) were removed because the device broke down, and because of another 37 participants where no baseline RINT test was done at all because of the break down. In all analyses, we removed 4 people who where moved in between and 6 pregnant women.
Study participants were on average 49 years of age at baseline, typically never smokers (46%), a relatively high education level (54%) and a paid job (47%) (Table 1). Based on the baseline questionnaire, 20% had current symptoms of wheeze, 12% had had a diagnosis of asthma and 21% had taken lung medicines recently.

Changes in air pollution concentrations
Most air pollution concentrations declined in the second half of 2010 compared to 2008 (Table 2). Average decreases ranged between 10% (PM_{10}, soot, NO_2) and 30% ($PM_{2.5}$), a relatively modest decrease (Figure 1). Table 2 also illustrated the variability in decline across locations. One urban street (Stille Veerkade) in The Hague where traffic flow was substantially reduced, showed substantially larger air pollution decreases than the other streets and suburban background locations, especially in soot (41%) (Table 2, Figure 1). With the exception of this street, trends did not differ significantly between the busy streets and the suburban control locations, as previously reported[12].
Spearman correlations were high for changes in the traffic-related air pollutants including soot, NO_2, NO_x, and the elements Cu and Fe ($R>0.7$), preventing an analysis of the independent effect of these pollutants. Correlation of the traffic indicators with $PM_{2.5}$ was lower ($R=0.20$-0.52). There was no correlation between traffic indicators changes and changes in PM_{10} ($R=-0.19$-0.13). Correlation between PM_{10} and $PM_{2.5}$ changes was 0.40.
The decrease in air pollution concentration in our study period was also visible in PM_{10} and NO_2 concentrations from fixed site monitors -having continuous data in contrast to our design - throughout the whole country, both on urban and regional sites (data not shown).

Table 1 Characteristics[a] of the study participants

	Participants (640)
Age (yr)	49.2 (4.4-87.5)
Female (%)	54.3
Height baseline (cm)	170 (104-202)
Difference[b] in height (cm)	1 (-1-17)
Smoking status (%)	
Never smoked	46.1
Current smoking	14.7
No. of cigarettes per day for current smokers	10.8 (1-25)
Education level (%)	
Low	25.6
Middle	20.0
Having a cold (%)	
Baseline	8.9
Follow-up	9.1
Time difference[b] (hr:min)	-00:13 (-10:16-10:20)
Passive smoking indoors (%)	9.2
Carpet in the bedroom (%)	47.0
Pets indoors (%)	33.9
Gas cooking (%)	86.9
Mould indoors (%)	29.5
Workplace exposure to dust or fumes (%)	6.4
Having a paid job (%)	46.9
Difference[b] in outdoor temperature (°C)	1.3 (-8.8-17.2)
Difference[b] in outdoor NO_2 concentrations ($\mu g/m^3$)	-5.5 (-57.4-30.7)
Current wheeze (%)	20.2
Current dyspnea (%)	14.8
Ever doctor-diagnosed asthma (%)	12.2
Doctor-described lung medicine use in previous yr (%)	21.2
Baseline health measurements	
Exhaled NO (ppb)	21.4 (<5-152)
Airway resistance (kPa/l/s), N=497	0.35 (0.06-1.24)
FVC (ml), N=585	3963 (1169-7558)
FEV_1 (ml)	3008 (412-6079)
PEF (ml/s)	7749 (1136-15124)

[a]Mean characteristics from the baseline questionnaire (min-max), unless stated otherwise; [b]Difference between the follow-up and the baseline examination.

Table 2 Baseline average air pollution concentrations and distribution of 2010-2008 air pollution changes[a] (N=12)

	Baseline (range)	2010-2008 changes					
		Min	P25	Mean	Median	P75	Max
PM_{10}	26.6 (18.1-32.9)	-7.7	-6.6	-3.3	-3.6	-1.2	3.0
$PM_{2.5}$	16.0 (12.4-19.4)	-7.6	-5.8	-4.5	-4.3	-3.2	-1.7
Soot	2.58 (1.31-4.26)	-1.74	-0.42	-0.22	-0.21	0.13	0.72
NO_2	40.6 (23.0-54.3)	-13.4	-5.5	-3.5	-3.5	-0.12	2.5
NO_x	69.6 (33.1-109.9)	-39.5	-15.6	-9.7	-6.4	-2.1	5.2
Cr	5.3 (0.8-10.0)	-3.9	-2.3	-1.3	-1.2	0.1	0.7
Cu	29.8 (6.9-62.5)	-27.2	-4.1	-0.01	0.3	8.5	18.3
Fe	686.0 (178.5-1405.9)	-492.9	-77.1	57.0	37.8	308.1	450.5

[a]Corrected for temporal variation by the use of a central reference location. Air pollutants are in $\mu g/m^3$, apart from soot ($10^{-5}/m$) and elements (ng/m^3). Elements derived from PM_{10} filters.

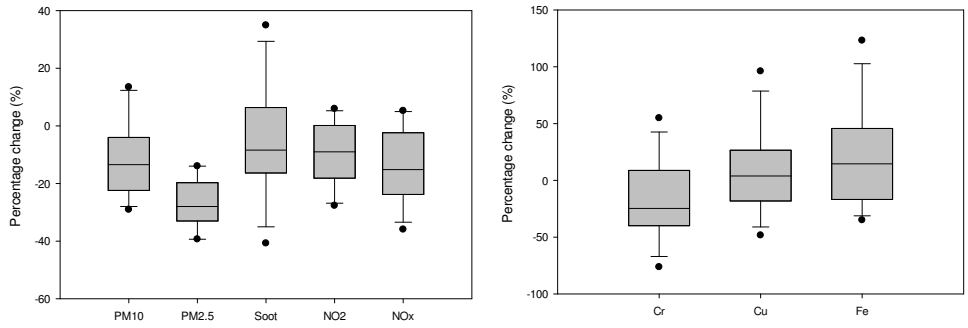

Figure 1 Distribution of changes (%) in average adjusted air pollution concentrations at street level (2010-2008) (N=12). Note that the scale of the y-axis differ

Changes in respiratory effects

In regression analyses adjusted for important covariates, there were significant associations between decreases in traffic-related air pollutants (soot, NO_2, NO_x, Cu and Fe) and improvements in FVC (Table 3). Per IQR decline in traffic-related air pollution concentration over a two year period a small improvement of about 1% in FVC (i.e. 40 ml) was found. Effect estimates were stable in both confounder models (basis, extensive), and differences between unadjusted and adjusted models were relatively small. After excluding participants with changing smoking status results did not change. Results were also stable after excluding the 1% most influential observations (based upon Cooks distance) and after excluding children (data not shown). When only participants were selected with both tests performed by the same technician, effects reported above were stable for soot (estimate -0.73 per IQR, p-value: 0.03), but were less negative for the other traffic-related air pollutants, and lost statistical significance. No significant association was seen with FEV_1, although associations were in the expected direction (Supplement Table 1). In addition, no associations were found with the peak expiratory flow (PEF), which is more variable within an individual than FVC and FEV_1 (data not shown). There were significant associations between a decrease in PM_{10} and $PM_{2.5}$ and a decrease in airway resistance (Table 3), although somewhat less consistent. No associations were seen with exhaled NO, a marker for airway inflammation (Table 3). There was insufficient variability to allow any meaningful analysis of self-reported respiratory symptoms (data not shown). The associations were largely driven by residents at the one street (Stille Veerkade) where traffic-related air pollution was drastically reduced. In those residents a significant FVC improvement of 5% was found (i.e. 199 ml), as well as a 3% FEV_1 improvement (i.e. 90 ml) compared to all four suburban background residents (Table 4). Similar effects were found when compared to the one matching suburban background residents. Effects were similar in the subgroup where both tests were performed by the same technician, or when excluding current smokers. Airway resistance was substantially decreased as well in those residents, but reached statistical significance only in the basis confounder model. The small group of residents in The Hague (N=31) did not differ from the suburban population in important confounder variables, outdoor temperature and short-term air pollution, although there were some more current smokers, which was the case for all street residents (Supplement Table 2).

In two-pollutant models with soot (as a marker for traffic-related air pollution) and PM_{10}, FVC effects remained similar. In addition airway resistance effects remained as well in the PM_{10} and $PM_{2.5}$ models including soot. Also in a two-pollutant model with PM_{10} and $PM_{2.5}$, separate airway resistance effects of both particle sizes remained (data not shown).

After exclusion the street residents in The Hague and the matching suburban background residents, FVC effects largely disappeared and airway resistance effects became inconsistent with significant effects in both directions (Supplement Table 3).

Table 3 Association between changes in average air pollution concentrations at street level and changes in respiratory health of residents[a] (forced vital capacity (FVC), airway resistance and exhaled NO, respectively)

		Unadjusted			Adjusted - Basis			Adjusted - Extensive		
	IQR	% change	SE	P-value	% change	SE	P-value	% change	SE	P-value
Forced vital capacity (FVC) (N=585)										
PM_{10}	5.5	0.67	0.60	0.27	0.29	0.52	0.57	0.47	0.53	0.38
$PM_{2.5}$	2.6	-0.63	0.56	0.26	-0.64	0.48	0.18	-0.87	0.55	0.12
Soot	0.55	-0.82	0.39	0.04	-0.82	0.33	0.01	-0.83	0.35	0.02
NO_2	5.4	-1.43	0.47	0.00	-1.03	0.40	0.01	-1.03	0.46	0.02
NO_x	13.5	-1.10	0.50	0.03	-1.00	0.43	0.02	-0.91	0.44	0.04
Cr	2.4	-0.87	0.68	0.20	-0.78	0.58	0.18	-0.80	0.60	0.18
Cu	12.6	-1.30	0.41	0.00	-0.95	0.35	0.01	-0.88	0.37	0.02
Fe	385.2	-1.60	0.58	0.01	-1.21	0.50	0.02	-1.11	0.51	0.03
Airway resistance (N=497)										
PM_{10}	5.5	9.96	3.04	0.00	6.68	3.31	0.04	9.41	3.67	0.01
$PM_{2.5}$	2.6	9.17	2.84	0.00	7.66	2.79	0.01	4.66	3.36	0.17
Soot	0.55	-0.60	2.11	0.78	2.41	2.31	0.30	-0.06	2.57	0.98
NO_2	5.4	-1.65	2.65	0.54	0.99	2.82	0.72	-0.69	3.38	0.84
NO_x	13.5	-2.40	2.54	0.35	0.86	2.57	0.74	-1.54	2.67	0.56
Cr	2.4	-0.53	3.45	0.88	6.60	3.47	0.06	9.45	3.58	0.01
Cu	12.6	-3.87	2.27	0.09	-1.00	2.38	0.67	-4.10	2.62	0.12
Fe	385.2	-8.38	3.19	0.01	-3.36	3.45	0.33	-7.94	3.97	0.05
Exhaled NO (N=640)										
PM_{10}	5.5	-1.51	3.81	0.69	-0.33	3.83	0.93	-0.05	3.92	0.99
$PM_{2.5}$	2.6	-4.97	3.50	0.16	-3.86	3.48	0.27	-5.92	4.03	0.14
Soot	0.55	-1.82	2.44	0.46	-2.21	2.41	0.36	-2.27	2.51	0.37
NO_2	5.4	-1.74	2.95	0.56	-3.24	2.95	0.27	-1.21	3.32	0.72
NO_x	13.5	-4.46	3.18	0.16	-5.56	3.16	0.08	-4.84	3.22	0.13
Cr	2.4	-0.72	2.67	0.79	-0.13	2.67	0.96	-1.36	2.97	0.65
Cu	12.6	-3.25	2.60	0.21	-4.03	2.59	0.12	-3.44	2.68	0.20
Fe	385.2	-3.03	3.64	0.41	-4.58	3.63	0.21	-3.92	3.74	0.30

[a]Estimates are calculated from linear regression with change (2010-2008) in health outcome as dependent variable and change in air pollution concentration as independent variable in one-pollutant models. Slope and SE were multiplied by the interquartile range (IQR) and divided by mean baseline value of the health outcome (See Table 1) and then multiplied by 100 to obtain a percentage change. Expected direction is a negative (FVC) and positive percentage (airway resistance, exhaled NO).

Table 4 Differences in respiratory health changes (%) between the street with the largest reduction in air pollution and residents at four suburban background locations with a more modest air pollution change[a]

	Unadjusted			Adjusted - Basis			Adjusted - Extensive		
	% change	SE	P-value	% change	SE	P-value	% change	SE	P-value
				Spirometry (N=293)					
FVC	4.26	1.77	0.02	4.53	1.52	0.00	4.92	1.69	0.00
FEV_1	2.57	1.72	0.13	3.05	1.31	0.02	3.00	1.45	0.04
PEF	0.50	2.17	0.82	1.48	1.90	0.44	2.43	2.10	0.25
				Airway resistance (N=261)					
	-1.60	8.56	0.85	-20.41	9.21	0.03	-15.34	11.34	0.18
				Exhaled NO (N=321)					
	-2.10	12.84	0.87	-4.19	12.49	0.74	5.29	13.80	0.70

[a]Estimates are calculated from linear regression with change (2010–2008) in respiratory health as dependent variable and living at the street (yes/no) with the largest air pollution reduction as independent variable. Slope and standard error (SE) divided by mean baseline value of the health outcome and then multiplied by 100 to obtain a percentage change. Expected direction is a positive percentage for FVC, FEV_1, and PEF, and a negative direction for airway resistance and exhaled NO. Mean baseline values: 3989 ml (FVC); 3028 ml (FEV_1); 7844 ml/s (PEF); 0.34 kPa/l/s (airway resistance); 22.3 ppb (exhaled NO).

Discussion

Small improvements in respiratory function were found after a modest decrease in outdoor air pollution concentrations within two years time. Per IQR decline in air pollution concentration over a two year period a small improvement of about 1% in FVC and a 9% reduction in airway resistance was found, the latter somewhat less stable. No significant associations were seen for other spirometric measures and exhaled NO. Results were largely driven by the small group of residents living at the one urban street where traffic flows as well as air pollution were drastically reduced. FEV_1 and FVC improved with about 3 and 5% in those residents compared to suburban background residents. This was accompanied with a suggestive reduction in airway resistance.

Changes in respiratory function at one urban street

The improvement of respiratory function observed in a small group of residents living at the one street where both traffic and air pollution was drastically reduced, was robust when compared to all suburban background locations, to their matching suburban control, as well as compared to the other streets in this study. Associations were found both for spirometry (FVC and FEV_1) and airway resistance. This study adds to the small number of studies suggesting that a reduction in air pollution concentrations have measurable respiratory health improvements within a relatively short time period. MacNeill et al.[17] reported small improvements in childhood PEF after the introduction of traffic management policies in Oxford, UK. Avol et al.[5] showed an improvement in lung function growth among children who moved to cleaner areas within one to three years after movement. Downs et al.[3] documented improvement in lung function in adults after relatively small reductions in air pollution exposure.

Our study has several strengths and limitations. One limitation is the small study population that experienced a large reduction in air pollution exposure. We nevertheless observed significant associations because our design was based upon comparing changes in health within individuals. Within-subject variability is small compared to between-subject variability, especially for spirometry indices like FVC and FEV_1[15]. Lower within-subject variability of FVC and FEV_1 compared to airway resistance and exhaled NO, probably explains the smaller standard errors of the effect estimates for FVC, and possibly the more consistent associations observed for FVC.

We further included a heterogeneous study population (all residents of the streets to which the policy applied). Because of the small study population we could not focus exclusively on sensitive subgroups like children, asthmatics or the elderly. Response rates were low and therefore we can not claim that the study population was a representative sample of the general population in the selected streets. The low response has likely not caused bias in our effect

estimates as our study design is based upon evaluating changes within individuals in time. Population characteristics did not change much within the short study period of only two years. When we excluded people that changed smoking status, results were similar. Some residual confounding by smoking may be present since people who smoked at baseline, may smoke more or less cigarettes at follow-up. Results were similar after excluding current smokers at the Stille Veerkade.

Participants and technicians were aware of the purpose of the study. At the time of examination, the participants and also the technicians performing the tests did not know the air pollution concentration changes at the street under investigation. In addition, we felt that participants and technicians would be unlikely to recognise often subtle changes in air pollution concentrations, apart from the participants at the Stille Veerkade in The Hague, where traffic flows were notably drastically reduced. This may have influenced the self-reported respiratory symptoms results, but less likely the lung function and airway resistance measurements.

A strength of our study is that we measured exposure to air pollution at the same street as where the residents lived, and therefore, exposure misclassification will be limited. Most studies assessed air pollution exposures on a much larger spatial scale. Avol et al.[5] modelled community-wide exposure based on a few fixed site monitors. Down et al.[3] modelled annual average air pollution concentrations for each residence on a 200 m grid, thus quite extensively, although for PM_{10} solely, and the study was not designed to look at traffic effects. We measured air pollution exposure for six weeks spread over half a year in 2008 and 2010 at each location. The average based upon those six weeks was representative for that half a year as was demonstrated by a comparison with fixed site monitors running throughout the country that had continuous measurements. We measured exposure to air pollution half a year before actual health measurements took place. Changes in air pollution concentration may happen in the time before that half a year as well. For example in The Hague, traffic was already drastically reduced since November 2009, whereas the follow-up of the health measurements were performed in February 2011, more than a year later. Avol et al.[5] showed an improvement in lung function growth among children who moved to cleaner areas already within one year. That improvement in respiratory function can be expected within a relatively short time period after a drastic change in air pollution levels, has been seen in studies in bar workers before and after implementation of smoking bans in their place of work. Those studies showed lung function improvements up to 8% within a few months after the smoking ban[18,19].

In our study we included a control population to account for autonomous trends in air pollution and/or respiratory function, which is a major advantage. We selected suburban background populations in the neighbourhood of the cities under study. The participating population characteristics were somewhat different at the control locations, although we tried to include participants with similar age and socioeconomic status structure. We further adjusted for individual level confounders. Moreover,

the spirometry associations were not largely affected by different levels of adjustment for confounders.
The observed improvements in respiratory function were small, and likely clinically insignificant in healthy subjects[20]. However, small shifts in the population mean may result in a more substantial shift in the fraction of subjects with low lung function. In the 24-city study, decrements of population mean FVC of 3.5% in children associated with particle acidity were accompanied by an odds ratio of 2.5 for FVC below 85% of predicted[21]. Similar observations were made when comparing population mean PEF decrements associated with short-term PM_{10} exposure and the percentage of (clinically more relevant) large decrements in PEF[22]. Furthermore, a respiratory function improvement may serve as an important indicator of future health and life expectancy[23].
The larger effect size for FVC compared to FEV_1 and the lack of a PEF response suggest an improvement of a restrictive rather than an obstructive response. We did not study the mechanisms underlying these changes, but PM air pollution has been associated with increased airway, systematic inflammation and oxidative stress[24]. We observed decreases in transition metals and soot (likely associated with organic components) that may have contributed to a reduced oxidative stress response[12]. We did not find any association with exhaled NO, measured as a marker of airway inflammation. We can not exclude a small inflammatory response in our study, as the standard error of our effect estimates was substantially larger for exhaled NO than for FVC and FEV_1. There are also studies which suggest that exhaled NO reflects inflammation particularly in allergic subjects[25].

Full study population
In the full study population we used the detailed information on changes in air pollution for each street and control location to study the associations between changes in air pollution and changes in respiratory function directly, rather than restrict ourselves to comparisons of changes in respiratory function between intervention and control locations. These associations should therefore not be interpreted as an effect of the intervention.
FVC effects of traffic-related components and airway resistance effects of both PM_{10} and $PM_{2.5}$ remained in two-pollutant models. In this study we were not able to disentangle effects of the different traffic-related components like soot, NO_2, NO_x, Cu and Fe.
After exclusion of the street residents in The Hague and the matching suburban background residents, effects on respiratory function largely disappeared. The main reason was probably the lack of sufficient exposure contrast, since at the Stille Veerkade, traffic-related components like soot and NO_x almost halved, whereas for most other locations decreases were only modest.

Conclusion

Small improvements in respiratory function were found after a modest decrease in outdoor air pollution concentrations within two years time. Results were largely driven by the small group of residents living at the one urban street where traffic flows as well as air pollution were drastically reduced. Our results suggest that a decline in air pollution may lead to positive consequences for public health.

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Supplement

Supplement Table 1 Association between changes in average air pollution concentrations at street level and changes in forced expiratory flow in the first second (FEV_1) of residents[a]

		Unadjusted			Adjusted - Basis			Adjusted - Extensive		
	IQR	**% change**	**SE**	**P-value**	**% change**	**SE**	**P-value**	**% change**	**SE**	**P-value**
Forced expiratory flow in the first second (FEV_1) (N=585)										
PM_{10}	5.5	0.40	0.57	0.49	0.20	0.46	0.67	0.27	0.47	0.57
$PM_{2.5}$	2.6	-0.71	0.53	0.18	-0.63	0.43	0.14	-0.20	0.49	0.68
Soot	0.55	-0.34	0.37	0.36	-0.36	0.30	0.23	-0.41	0.31	0.18
NO_2	5.4	-0.46	0.45	0.30	-0.14	0.36	0.70	-0.75	0.40	0.06
NO_x	13.5	-0.41	0.48	0.39	-0.34	0.39	0.38	-0.31	0.39	0.44
Cr	2.4	-0.61	0.65	0.34	-0.62	0.52	0.23	-0.91	0.53	0.08
Cu	12.6	-0.45	0.40	0.26	-0.17	0.32	0.60	-0.33	0.33	0.32
Fe	385.2	-0.44	0.55	0.43	-0.19	0.45	0.67	-0.46	0.46	0.32

[a]Estimates are calculated from linear regression with change (2010-2008) in health outcome as dependent variable and change in air pollution concentration as independent variable in one-pollutant models. Slope and standard error (SE) were multiplied by the interquartile range (IQR) and divided by mean baseline value of the health outcome (See Table 1 main article) and then multiplied by 100 to obtain a percentage change. Expected direction is a negative percentage.

Supplement Table 2 Distribution of important individual covariates per location

Cities	Urban streets	N people[a]	Age (range)	N current smokers (%)	N cigarettes per day (range)	N passive smokers (%)	N people highly educated (%)
Amsterdam	Haarlemmerweg	58 (2)	39 (6-71)	12 (21)	11 (1-20)	2 (3)	48 (83)
Amsterdam	Hoofdweg	28 (4)	50 (5-84)	5 (18)	4 (1-8)	3 (11)	15 (54)
The Hague	Stille Veerkade	31 (4)	45 (11-75)	7 (23)	11 (6-15)	6 (19)	14 (45)
Den Bosch	Brugstraat	29 (0)	40 (5-57)	7 (24)	9 (1-25)	7 (24)	17 (59)
Den Bosch	Koningsweg	52 (11)	44 (4-76)	7 (13)	10 (1-20)	8 (15)	41 (79)
Tilburg	HVB	78 (6)	64 (18-87)	17 (22)	14 (1-25)	9 (12)	11 (14)
Utrecht	Vleutenseweg	62 (1)	39 (5-74)	9 (15)	10 (2-20)	8 (13)	48 (77)
Utrecht	Weerdsingel Wz	12 (0)	39 (8-75)	2 (17)	8 (4-12)	4 (33)	8 (67)
Suburban control							
Amsterdam		45 (8)	49 (5-77)	7 (16)	10 (3-20)	2 (4)	17 (38)
The Hague		48 (3)	54 (12-86)	7 (15)	13 (3-25)	4 (8)	11 (23)
Den Bosch/Tilburg		89 (4)	53 (4-82)	9 (10)	9 (1-20)	5 (6)	45 (51)
Utrecht		108 (8)	49 (4-85)	5 (5)	13 (2-20)	1 (1)	73 (68)

[a]in brackets the number of people visiting at home for the follow-up health measurements.

Supplement Table 3 Association between changes in average air pollution concentrations at street level and changes in respiratory health of residents – subgroup analysis excluding residents from the street with the largest air pollution reduction and their matching suburban background[a]

		Unadjusted			Adjusted - Basis			Adjusted - Extensive		
	IQR	% change	SE	P-value	% change	SE	P-value	% change	SE	P-value
Forced vital capacity (FVC) (N=515)										
PM_{10}	5.5	0.68	0.60	0.26	0.30	0.52	0.56	0.42	0.54	0.43
$PM_{2.5}$	2.6	0.51	0.71	0.47	0.45	0.60	0.46	0.30	0.70	0.66
Soot	0.55	-0.28	0.58	0.63	-0.04	0.49	0.94	0.02	0.52	0.97
NO_2	5.4	-0.98	0.56	0.08	-0.30	0.49	0.55	0.16	0.61	0.79
NO_x	13.5	-0.16	0.72	0.82	0.37	0.63	0.56	0.63	0.64	0.33
Cr	2.4	-1,34	0,77	0,08	-0,81	0,67	0,23	-0,78	0,69	0,26
Cu	12.6	-0.98	0.51	0.05	-0.29	0.44	0.52	-0.10	0.46	0.82
Fe	385.2	-1.28	0.71	0.07	-0.43	0.62	0.49	-0.17	0.64	0.79
Airway resistance (N=427)										
PM_{10}	5.5	10.87	2.94	0.00	7.09	3.27	0.03	5.82	3.33	0.08
$PM_{2.5}$	2.6	6.60	3.60	0.07	6.39	3.67	0.08	3.28	4.22	0.44
Soot	0.55	2.55	3.32	0.44	-0.83	3.62	0.82	-2.09	3.73	0.58
NO_2	5.4	-4.82	3.31	0.15	-5.83	3.99	0.14	-3.50	4.81	0.47
NO_x	13.5	-7.01	3.66	0.06	-5.66	3.75	0.13	-6.08	3.75	0.11
Cr	2.4	15.07	4.31	0.00	14.82	4.37	0.00	13.81	4.49	0.00
Cu	12.6	-6.46	2.82	0.02	-6.15	2.98	0.04	-6.29	3.11	0.04
Fe	385.2	-9.22	3.90	0.02	-8.94	4.08	0.03	-9.42	4.42	0.03

[a]Estimates are calculated from linear regression with change (2010-2008) in health outcome as dependent variable and change in air pollution concentration as independent variable in one-pollutant models. Slope and standard error (SE) were multiplied by the interquartile range (IQR) and divided by mean baseline value of the health outcome (3956 ml for FVC; 0.35 kPa/l/s for airway resistance) and then multiplied by 100 to obtain a percentage change. Expected direction is a negative (FVC) and positive percentage (airway resistance).

Chapter 7

General discussion

Main findings

In the measurement campaign before implementation of the local traffic policies, high contrasts between busy streets and background locations in the same city were found for Cr, Cu and Fe (factor 2-3). These elements were especially present in the PM coarse fraction. In addition, high contrasts were found for soot and NO_x (factor 1.8-1.9), typically indicators of combustion emissions. Contrast was lower for the regulated component PM_{10}, $PM_{2.5}$ and NO_2 (factor 1.2-1.5). The largest contrast was found for two street canyons and two streets with buildings at one side of the street only (Chapter 2).
The contrast for PNC was similar to soot in a subset of streets and corresponding background locations. At the street location, high temporal variability of PNC concentrations occurred within each sampling day, probably related to variations in traffic volumes, high-emission of individual vehicles and wind direction. For the background locations, smaller and more gradual temporal variability was found probably reflecting similar area-wide source contributions and meteorology (Chapter 3).
Before implementation of local traffic policies, the oxidative potential of PM_{10} collected at major streets and measured with the EPR assay was 3.6 times higher than that of simultaneously measured PM_{10} from urban background locations, and 6.5 times higher than PM_{10} from suburban locations. The contrast exceeded corresponding contrasts in other PM characteristics, including concentrations of transition metals such as Cu and Fe (Chapter 4).
After the LEZ introduction, these contrasts were very similar. Overall, the LEZ policies directed at old heavy duty vehicles (trucks) did not substantially change concentrations of traffic-related pollutants (e.g. soot, NO_2, NO_x, Cu, Fe) at street sites more than at suburban background sites outside of the LEZ, though there were substantial differences across locations. Street and urban background $PM_{2.5}$ concentrations did decrease more (30 and 27%) than suburban background $PM_{2.5}$ (20%), which is probably related to other changes over time. In one street - where in addition to the LEZ - traffic intensity was reduced, soot, NO_x and NO_2 concentrations decreased significantly more (41, 36 and 25%) than at the corresponding suburban background location (22, 14 and 7%). With the exception of that street, the local traffic policies including LEZ were probably too modest to produce statistically significant decreases in traffic-related air pollution concentrations (Chapter 5).
Especially in the one street where traffic was drastically reduced, lung function (FVC, FEV_1) was improved with 3-5% in the residents compared to their corresponding control population within two years time. This was accompanied with a suggestive reduction in airway resistance. No association was seen with exhaled NO. In the total population, small improvements in lung function were found up to 1% after a decrease in traffic-related air pollution within two years time (Chapter 6).

Comparison with other studies

In the comparison with other studies we focused first on the air pollution contrast between streets and background locations. After that we described the results of other studies that have evaluated the air pollution concentration effects of LEZ based on modelled as well as measured data. In the next section we reported the results of other (traffic) policies/actions on measured air pollution concentrations. In the last section results from other studies were described that have measured the health effect of (traffic) policies/actions. The methodologically challenges identified in those intervention studies are discussed in a new paragraph.

Air pollution contrast between streets and background locations

Our study confirmed the increasing evidence that proximity to traffic is much better reflected in contrasts in particle number concentrations (PNC) or soot than in PM_{10} and $PM_{2.5}$ levels[1]. PNC and soot are both combustion-related indicators mostly directly emitted from the engine. Soot has been recently recommended as a valuable additional air quality indicator for regulation, also because of its health relevance[2]. Ultrafine particles characterised by PNC rather than particle mass is of interest as well given the emerging epidemiological and toxicological evidence[3,4]. However until now, the high costs and complexity of monitoring equipment and concerns about the validity of central-site to estimate exposure limits the feasibility of ultrafine particles as an additional metric for regulation and hampers wide-scale use[2].

The contrast between busy streets and background locations measured in the same city was even somewhat larger for non-tailpipe traffic indicators (e.g. Cu and Fe) than for tailpipe indicators (e.g. soot). These elements were especially present in the coarse fraction of PM (particles larger than 2.5 µm and smaller than 10 µm). Re-suspended road dust, tyre- and brake wear are sources of non-tailpipe traffic emissions. Around half of the PM_{10} increase near major roads has been estimated to be attributed to non-tailpipe traffic emissions[5]. Non-tailpipe emissions may even dominate PM levels at urban roads in Northern countries like Norway and Sweden during winter as a result of mainly studded tyres[6]. To date, traffic policies have focused almost exclusively on reducing tailpipe emissions of vehicles. With the reduction of tailpipe emissions, emissions from non-tailpipe sources will make up a larger proportion of traffic emissions[1]. Therefore it can be expected that more traffic policies will be targeted to specifically reduce non-tailpipe emissions in the near future, as discussed in a recent European expert workshop as well[7]. However until now, large uncertainties exist surrounding the exact contribution of non-tailpipe emissions in total traffic emissions, and what this effectively means in terms of health. This needs further investigation. Additional health gains can be anticipated, since non-tailpipe traffic components like transition metals and organics (e.g. polycyclic hydrocarbons) may play a role in the pro-inflammatory effects of PM[8,9]. Epidemiological

studies have found associations between daily variation in coarse PM concentrations and a range of adverse respiratory and cardiovascular health morbidity effects, independent from $PM_{2.5}$ effects[10]. The independent effect of coarse PM on mortality has been questioned as in most time-series studies coarse PM effects diminished after controlling for $PM_{2.5}$[10]. However some recent studies showed quite stable mortality effects of coarse PM also with adjustments for $PM_{2.5}$[11,12].

Contrast in oxidative potential of PM_{10} between busy streets and background locations was large, and exceeded corresponding contrasts in other PM characteristics, including concentrations of transition metals such as Cu and Fe. Oxidative stress has been suggested as an important underlying mechanism of action by which exposure to PM may lead to adverse health effects[13]. To measure the ability of PM to cause oxidative stress (=oxidative potential of PM) is a novel metric, and may provide a more health-based exposure measure by integrating various biologically relevant properties of traffic-related air pollution particles into a single predictor of biological activity[14]. There are various assays to measure the oxidative potential, and until now there is no consensus regarding the most appropriate assay[15]. In our study, oxidative potential was measured as the ability to generate hydroxyl radicals in the presence of hydrogen peroxide[14,16]. To our knowledge no other studies have included a systematic comparison of oxidative potential between PM collected near major urban streets and background locations. Künzli et al.[17] compared oxidative potential with other $PM_{2.5}$ characteristics measured at twenty different European locations and reported that oxidative potential varied by one order of magnitude both in time and in space, and that it was not well correlated with other PM characteristics. The study of Künzli et al.[17] was not designed to look at traffic pollution effects specifically, because most of the sites in that study were urban background locations. In our study oxidative potential was highly correlated with traffic-related PM components including soot, Cu and Fe.

The health relevance of oxidative potential of PM is at the moment largely unknown. Almost no epidemiological studies have linked oxidative potential of particles to adverse health effects such as respiratory or cardiovascular morbidity and mortality, apart from two studies. In a panel study Delfino et al.[18] related oxidative potential using the alveolar macrophage ROS assay to airway and systematic inflammation in 60 elderly people. The oxidative potential effects on IL-6 and exhaled NO were comparable with the effects of traffic pollutants such as black carbon and PNC[18]. To disentangle the independent health effects of individual PM characteristics in epidemiological studies is often limited by high correlations between air pollution components. Recently, Strak et al.[19] investigated which PM characteristics including oxidative potential have the most consistent associations with acute changes in respiratory function in healthy volunteers. They measured exposure at five real-world locations with well-established different PM characteristics to reduce correlation between PM characteristics. Volunteers were exposed for five hours, while exercising intermittently. Respiratory effects were measured before and at three time points after exposure.

Especially PNC, NO_2 and NO_x were associated with acute airway inflammation and impaired lung function, not oxidative potential of PM or PM_{10} and $PM_{2.5}$ mass concentrations[19]. Oxidative potential was measured using the antioxidant depletion assay[20], a different assay than used in our study.
Our hypothesis was that traffic policies would affect components with a large concentration contrast between streets and urban background locations like the oxidative potential of PM more than PM_{10} and $PM_{2.5}$, since the latter showed much smaller gradients. Unfortunately, oxidative potential of PM from the post measurement campaign was assessed in another laboratory, because the former laboratory closed down. Though the lab procedures were sent to the new laboratory that had prior experience with the assay, results of the post analysis varied more than an order of magnitude between the two campaigns, which was unlikely to be real. Moreover, the correlation with transition metals like Cu and Fe – the main driving forces in this assay - appeared to be much lower than in the baseline campaign. In addition, there was almost no spatial and temporal variation in the post analysis visible, which is again highly questionable. At this moment, the use of different oxidative potential assays, a lack of standardised operational procedures within one assay, difficulties to assess exposure because the oxidative potential of PM varies substantially in time and space, uncertainties in terms of health-relevance and the high costs of the different assays currently limits wide-scale use in e.g. air quality management.

Effect of LEZ on air pollution concentrations
In our study the LEZ policies directed at heavy duty vehicles (trucks) did not substantially change concentrations of traffic-related pollutants at street sites more than at suburban background sites outside of the LEZ. LEZ were seen as one of the most promising policies in the efforts to comply with EU air quality standards in the Netherlands as well as in other European countries[21]. Before implementation of the LEZ in the Netherlands effects on air quality have been modelled. Models predicted widely varying estimates for the concentration reductions due to the LEZ in specific streets, up to 4 $\mu g/m^3$ for NO_2 and 1.5 for PM_{10}[22]. A recent evaluation study in 2010 showed already far lower concentration reductions due to the LEZ, below 0.3 $\mu g/m^3$ for both pollutants[23]. With our study design, we were not able to significantly detect changes within that order of magnitude. Three main explanations were given why the modelled LEZ effects were smaller than anticipated when the policy was formulated. Firstly, NO_x emissions of new trucks under typical real-world urban driving conditions were not that different from old truck emissions as was estimated in standard type approval tests under laboratory conditions[24]. How this will work out for new trucks (EURO-VI) entering the market in 2013 remains to be seen. The second reason is the lower amount of trucks driving at the streets than originally thought[23]. The absolute amount of old trucks affected by the policy will be even lower. Some Dutch cities are now considering a possible extension of the LEZ targeting old vans as well as all old passenger cars. The air pollution effects of this extension will probably be

larger, as this would affect a much larger number of vehicles. The third major reason for the smaller modelled effects than reported before is that model predictions did not take into account non-compliance, and the fact that most old trucks were largely tolerated in practice in the first year after implementation. Enforcement was tightened with cameras and special staff since 2010, with high fines for drivers entering illegally.
Many other cities in Europe as well as in cities in other parts of the world have implemented long-term traffic management schemes like the LEZ including London, Stockholm and Rome, as well as in Tokyo and Singapore. Those zones are directed to trucks, buses, all old vehicles, or all vehicles entering the inner-city. There are a few evaluation studies on LEZ or similar zones, often based on model predictions instead of measurements. Johansson et al.[25] modelled rooftop concentration reductions in the zone up to 7 and 12% for PM_{10} and NO_x along four densely trafficked streets in Stockholm compared to a baseline scenario without the zone. In Rome, traffic is restricted in the inner-city. Cesaroni et al.[26] modelled reductions between 10 to 23% in PM_{10} and NO_2 concentrations in the most affected areas of Rome. Tonne et al.[27] estimated the impacts of the congestion charging zone introduced in the inner-city of London, UK, which charges for travel into central London. After implementation of the congestion charging zone, traffic volumes reduced considerably, and air pollution concentration reductions in the zone were estimated to be on average 0.2 $\mu g/m^3$ for PM_{10} and 0.7 $\mu g/m^3$ for NO_2[27]. In a more recent extensive evaluation of the congestion charging zone, including measured data, effects were smaller or even not existing[28,29]. In addition to the congestion charging zone in London, a LEZ directed at middle and heavy traffic was recently introduced. The LEZ covers a much wider area than the congestion charging zone, but results on the effects on air pollution concentrations were not published yet[30]. In Kopenhagen, Denmark, the LEZ directed to old trucks and buses was evaluated with the use of measurements along one busy street and one urban background site. An extra reduction was seen in $PM_{2.5}$ of about 0.7 $\mu g/m^3$ (5%), compared to background trend. Effects on other pollutants including NO_x and EC were smaller[31]. In Germany 41 cities implemented LEZ banning old vehicles (both cars and trucks) from city centers[21], but results on the effectiveness was not published yet. See Table 1 for a summary of the LEZ studies. Most evaluation studies on the LEZ reported small average concentration reductions below 1 $\mu g/m^3$ $PM_{10}/PM_{2.5}$, with generally larger concentration reductions at busy urban streets within the LEZ.

Effect of other (traffic) policies / actions on air pollution concentrations
There have been a small number of studies of the impact of short-term reversible traffic management schemes on measured air pollution concentrations (Table 1). During the Atlanta and Beijing Olympic Games many traffic policies were taken to reduce traffic congestion and to improve air quality[32-35]. Especially during the Olympic Games in Beijing radical policies were implemented including tightening of fuel standards, banning old vehicles

and incorporating odd and even number plate driving days for cars[36]. Substantial reduction in air pollution concentrations was found up to 64-70% for BC and $PM_{2.5}$ (Beijing) during the Olympics compared to before[35]. Also during the Asian Games in Busan, Korea, traffic policies were taken and air pollution (CO, NO_2, SO_2) was reduced up to 10%[37]. A study in Boston investigated effects of temporary road closures aimed to reduce traffic volumes due to a major event of a few days[38]. Another study investigated air pollution concentrations in Haifa, Israel, during a military conflict of about a month[39]. Traffic was reduced quite substantially (40%) and NO_2 and PM_{10} dropped considerably with about 20-55% as well during the conflict compared to periods before and after, as basically the conflict interrupted all 'normal' commercial and personal activities[39]. Other temporary not-traffic related policy examples are published including the unique opportunity to assess the air pollution effects during an eleven-month strike in a steel factory in Utah Valley, USA, and a copper smelter strike in the Southwest of USA, which resulted in large reductions of especially sulphate particles and transition metals[40-42].

In addition, there are a few studies that evaluated air pollution changes of more permanent (traffic) policies/actions. After the coal ban in Dublin, Ireland in 1990, air pollution concentrations (soot) dropped almost immediately with about 70%[43]. Goodman et al.[44] assessed coal bans in multiple Irish cities implemented in later years till 2000, and generally showed often much smaller air pollution reductions than after the Dublin coal ban. In Hong Kong, China, the government restricted the use of sulphur in gasoline in 1990, and after that SO_2 concentrations dropped significantly (45%)[45]. Peters et al.[46] assessed slower, less well defined changes in air pollution in Erfurt, Germany in the years 1990-2002, a period associated with changes in domestic heating, industry and vehicle fleet. Strongest reduction was found in SO_2. PM_{10}, $PM_{2.5}$ and CO decreased by more than 50%. Other pollutants like NO_2, O_3 and ultrafine particles decreased to a lesser extent[46]. Noonan et al.[47] investigated outdoor as well as indoor air pollution effects of a three years woodstove exchange programme in Montana, USA. Outdoor $PM_{2.5}$ concentrations were significantly reduced up to 30% after the programme, but indoor air pollution changes varied significantly more. Outdoor $PM_{2.5}$ concentrations were not different in the first winter after the exchange because only ~13% of the woodstoves had been exchanged at that time. A few other studies evaluated the switch to compressed natural gas in public transport in New-Delhi, India[48,49]. They showed rather mixed results, including a substantial increase in benzene of about 60%, attributed to an increase in traffic at the same time[48].

The studies above investigated policies or natural experiments implemented city-wide or in large areas within cities. A few other examples of measured evaluations exist which are on a more local scale, like the effects of a new-build tunnel in Sydney[50], a construction of a bypass in North Wales, UK and Monasterevin, Ireland[51,52], or effects of a speed limit reduction in Amsterdam, the Netherlands[53].

Table 1 Summary of intervention studies on air pollution effects of the LEZ and other (traffic) policies and actions

Study	Main policy / action	Design	Main results
Studies on the LEZ			
Johansson et al.[25]	Congestion zone for all vehicles in the inner-city of Stockholm, Sweden, 2006.	Modelling approach. Annual (rooftop) concentration averages modelled with a dispersion model. Meteorology and other emission sources kept constant.	15% reduction in road use in the zone. Largest reductions within the zone, up to 2 $\mu g/m^3$ in PM_{10} and NO_x. Average decrease in population weighted concentration within the zone is ~10%.
Cesaroni et al.[26]	LEZ for mainly (old) vehicles in the inner-city of Rome, Italy, 2001-2003.	Modelling approach. Annual concentration averages for traffic roads were modelled with a dispersion model. Meteorology and other emission sources kept constant.	4% reduction in traffic Rome-wide. Concentration reduction of 10-23% in most affected area (PM_{10}, NO_2). Average decrease in population concentrations city-wide <1 $\mu g/m^3$ (1-2%).
Tonne et al.[27]	Congestion charging zone for all vehicles in the inner-city of London, UK, 2003.	Modelling approach. Annual concentration averages modelled with an emission-dispersion model. Meteorology and vehicle fleet kept constant.	7-26% reduction in traffic in the zone. Average decrease in the zone is 0.2 and 0.7 $\mu g/m^3$ (PM_{10}, NO_2), which is <1%.
Atkinson et al.[28]	Congestion charging zone for all vehicles in the inner-city of London, UK, 2003.	Measuring approach. Comparison: two year before and two years after the policy. With multiple fixed site urban monitors in and outside the zone.	No concentration effects were found on the one roadside monitor in the zone. Some evidence for a small reduction at three urban background sites in the zone for NO, and an increase in NO_2 and O_3.
Jensen et al.[31]	LEZ for old trucks and buses in the inner-city of Kopenhagen, Denmark, 2008.	Measuring approach. Comparison: over a three year period. With one traffic site, an urban background site, and a regional site.	An extra reduction was seen in $PM_{2.5}$ of 0.7 $\mu g/m^3$ (5%) compared to background trend. NO_x and EC effects were smaller.
Studies on short-term traffic policies / actions / unplanned events			
Friedman et al.[32]	Temporary traffic policies taken during the Olympic Games in Atlanta, USA, 1996.	Measuring approach. Comparison: during Games with 1 month before and after Games. With 1-3 fixed site urban monitors.	3% reduction in 24-h total traffic. 23% reduction in rush hour traffic. 28% concentration reduction in O_3.
Peel et al. [33]	Temporary traffic policies taken during the Olympic Games in Atlanta, USA, 1996.	Measuring approach. Comparison: same time frame as Friedman et al. With multiple fixed site urban monitors. Inclusion of control locations and years.	No reduction in 24-h total traffic. 2-20% reduction in rush hours traffic. O_3 concentration reduction also visible in neighbouring USA cities used as controls.

Table 1 Continued

Study	Main policy / action	Design	Main results
Studies on short-term traffic policies / actions / unplanned events (continued)			
Li et al.[34]	Temporary traffic policies taken during the Olympic Games in Beijing, China, 2008.	Measuring approach. Comparison: during Games with pre-Games and one month before. With three fixed site urban monitors.	Traffic reductions not reported in the paper. 60% concentration reduction in $PM_{2.5}$. 7% concentration reduction in O_3.
Lin et al.[35]	Temporary traffic policies taken during the Olympic Games in Beijing, China, 2008.	Measuring approach. Comparison: during Games with 4 different time periods of 10 sampling days before the Games within 2 years time. With one fixed site urban monitor.	64-70% concentration reduction in BC and $PM_{2.5}$.
Lee et al.[37]	Temporary traffic policies taken during the Asian Games in Busan, Korea, 2002.	Measuring approach. Comparison: 3 weeks after Games with baseline (3 weeks before and during Games). Inclusion of control years. With 8 fixed site urban monitors.	1 (PM_{10}) to 10% (CO, NO_2, SO_2) concentration reduction. O_3 was also reduced in other years (~25%).
Levy et al.[38]	Temporary road closures due to a 4-day major event in Boston, USA, 2004.	Measuring approach. Comparison: during event with one week before and after event. With 40 sites (NO_2).	~30% reduction in traffic at the most affected highway, not city-wide. Weak evidence for an average concentration reduction.
Yuval et al.[39]	A military conflict of 1 month in Haifa, Israel, 2006.	Measuring approach. Comparison: during conflict with 1.5 month before and 2 weeks after conflict. Inclusion of control years. With 7 fixed site urban monitors.	40% reduction in traffic. 20-55% concentration reduction in NO_2 and PM_{10}.
Pope et al.[40]	An 11-month steel factory strike in Utah Valley, USA. 1986-1987.	Measuring approach. Comparison: during strike with 1 year before and after strike. With unknown amount of fixed site monitors in Utah valley.	11-43% reduction in PM_{10} depending on the season.
Pope et al.[41]	A 9-months copper smelter strike in 4 USA states, 1967-1968.	Measuring approach. Comparison: during strike with the months before and after strike. With unknown amount of fixed site monitors in 4 Southwest states.	60% concentration reduction in sulphate particles.

Table 1 Continued

Study	Main policy / action	Design	Main results
Studies on other more permanent (traffic) policies / actions – city level			
Clancy et al.[43]	Coal ban in Dublin, Ireland, 1990.	Measuring approach. Comparison: 6 years after ban with 6 years before ban. With 6 fixed site urban monitors.	70% concentration reduction in BS. 34% concentration reduction in SO_2.
Goodman et al.[44]	Coal ban in multiple Irish cities from 1990-2000.	Measuring approach. Comparison: 5 years after ban with 5 years before the ban. With varying fixed site monitors (1-6) per city.	45 to 76% concentration reduction in BS in the different cities. SO_2 concentration reduction not consistent.
Hedley et al.[45]	A sulphur restriction in fuel, Hong Kong, China, 1990.	Measuring approach. Comparison: 5 years after policy with 2 year before the policy. With 5-8 fixed site urban monitors.	45% concentration reduction in SO_2. PM_{10} and NO_2 and O_3 concentration reduction not consistent.
Noonan et al. [47]	A woodstove exchange in Montana, USA, 2005-2009.	Measuring approach. Comparison: 4 winters after exchange with 2 winters before exchange. With 1 outdoor fixed site monitor and an indoor 24-hr measurement campaign at ~26 homes.	Outdoor $PM_{2.5}$ concentration was 22-30% lower in 3 winters, not the first winter after exchange. Indoor $PM_{2.5}$ reduction was much smaller.
Khillare et al.[48]	Change to compressed natural gas in pubic transport in New-Delhi, India, 2001-2002.	Measuring approach. Comparison: 2 winter months in 2007 with 2 winter months in 2002. With 3 fixed site monitors.	60% concentration increase in benzene.
Ravindra et al.[49]	Change to compressed natural gas in pubic transport in New-Delhi, India, 2001-2002.	Measuring approach. Comparison: 2 years after with 4 years before the change. With one fixed site street monitor.	Reduction in CO, SO_2 and PAHs. NO_x increased after policies. Percentages unknown.

Table 1 Continued

Study	Main policy / action	Design	Main results
Studies on other more permanent (traffic) policies / actions - more local level			
Cowie et al.[50]	A new-build tunnel in Sydney, Australia, 2007 which leads to traffic flow changes.	Measuring approach. Comparison: 1 year after tunnel with 1 year before tunnel. With 4 urban background and 3 regional fixed site monitors.	No consistent concentration reduction of NO_2, PM_{10} and $PM_{2.5}$.
Burr et al.[51]	A construction of a bypass in North Wales, UK, 1998.	Measuring approach. Comparison: 4 measurements in 1 year after bypass with 4 measurements in 1 year before bypass. With 2 sampling sites.	Heavy traffic decreased with 50%. 23-29% concentration reduction in PM_{10} and $PM_{2.5}$ at both sites.
O'Donoghue et al.[52]	A construction of a bypass in Monasterevin, Ireland, 2004.	Measuring approach. Comparison: 2 months after bypass with 3 weeks before bypass. With 1 traffic sampling site.	60% reduction in traffic in the city. Concentration reduction was found for NO_x (62%), CO (35%), PM_{10} (28%), and benzene (18%).
Dijkema et al.[53]	A speed limit reduction at an urban highway in Amsterdam, the Netherlands, 2005.	Measuring approach. Comparison: 1 year after policy with 1 year before the policy. With 2 fixed site road monitors.	2% reduction in traffic. 4% extra concentration reduction in PM_{10}. No concentration reduction of NO_x and BS.

Effect of policies on health

Especially at the one street where traffic was drastically reduced, lung function was improved with 3-5% in the residents compared to their matching control population within two years time. In our total study population, small improvements in lung function were found up to 1% after a decrease in traffic-related air pollution within two years time, not related to LEZ policies in particular. There are no other studies which estimated the respiratory health effects of residents associated with implementation of the LEZ specifically. Two modelling studies predicted small health gains in terms of life expectancy for residents living in the zone[26,27].

Numerous studies have documented adverse health effects of traffic-related air pollution[1], therefore it is plausible that air pollution reductions resulted in health improvements. Most evidence is based on cross-sectional comparisons and cohort studies which investigated association between (changes in) health status within individuals and baseline air pollution[54]. Until now, there is only little direct evidence of the effectiveness of local policies in reducing air pollution and ultimately improving health. Below a short overview is given of evaluation studies that have actually measured the health effects of (traffic) policies and actions (Table 2).

There are some studies which evaluated health effects associated with short-term reversible traffic management schemes to improve air quality including the Olympic Games in Atlanta, USA and Beijing, China. Friedman et al.[32] showed reductions in respiratory hospital admissions in children during the Games compared to four weeks before and after the Games. In an extensive reanalysis of this study, Peel et al.[33] did not find consistent evidence of an improvement in health during the Games: estimates were sensitive for the choice of analytical model, and to the method of adjusting for temporal trends. Lee et al.[37] investigated the Asian Games in Busan, Korea and did not report convincing evidence of an improvement in respiratory health associated with the Games either. Especially during the Olympics in Beijing more radical policies were taken to improve air quality. This was associated with an immediate improvement in airway inflammation in children[35], and a reduction in asthma hospital admissions in adults during the Games[34].

A few other not traffic-related policy examples exist including a temporary steel mill and copper smelter strike which resulted in substantial air pollution reductions[40,41]. During an eleven-month strike in a steel factory in Utah Valley, USA, especially children's respiratory hospital admissions were 3 times lower during the winters when the mill was closed compared to when it was open[40]. Overall mortality was reduced between 1.5 to 4% during a copper smelter strike in four Southwest USA states[41].

There are a few studies evaluating the health effects of other more permanent (traffic) policies/actions. Mortality rates declined as well with about 5.7%, whereas respiratory mortality rates decreased with about 15.5% after the coal ban in Dublin, Ireland[43]. In Hong Kong, a 3.9% decline in respiratory mortality was found after the government restricted the use of sulphur in gasoline[45]. Also after the sulphur restriction an improvement was reported in respiratory health in children in Hong Kong[55,56]. Although no

formal intervention study, Peters et al.[46] evaluated how the association between air pollution and risk of death changed in Erfurt, Germany within the years 1990-2002, a period where sources of air pollution changed from mainly coal combustion to more traffic emissions. Mortality was in particular associated with ultrafine particles, not with PM_{10}, $PM_{2.5}$ and SO_2 in the whole study period. Stronger responses were visible in the second, transitional sub-period, from 1995 to 1998 not entirely consistent with the more pronounced air pollution falls in earlier years. Likewise more prolonged mortality studies have been done, addressing questions like whether the health risk of exposure to air pollution changed over a period of substantial alterations in the sources and of declines in air pollution concentrations[57-59]. Pope et al.[59] directly evaluated changes in life expectancy with differential changes in air pollution in US cities during the 1980s and 1990s. A decrease of 10 $\mu g/m^3$ in $PM_{2.5}$ was associated with an increase in mean life expectancy of 0.6 year (7 months). Although in these more prolonged mortality studies reductions in mortality can not be associated to specific policies, and the large spatial scale may not be perfect for a policy evaluation, these studies provided valuable information about general air quality reductions and health patterns. Apart from mortality, there are a few studies evaluating the health effects of other more permanent (traffic) policies/actions on other health endpoints including respiratory hospital admissions and respiratory symptoms[47,60,61]. Noonan et al.[47] evaluated respiratory symptoms and school absences in children after the introduction of a woodstove exchange programme in Montana, USA. They reported reductions in some respiratory symptoms (e.g. wheezing) when $PM_{2.5}$ decreases at the one central site monitor. However, there was only weak evidence that the change in respiratory symptoms was due to the exchange programme. El-Zein et al.[60] reported some evidence for a reduction in respiratory symptoms and upper respiratory tract infections after a diesel ban in Beirut, Lebanon. Unfortunately no data was available on reinforcement of the policy and air pollution concentrations were not actually measured. MacNeill et al.[61] reported small improvements in childhood peak expiratory flow after the introduction of traffic management policies in Oxford, UK.

Some intervention studies are on a more local scale, like the respiratory effects due to implementation of a bypass in North Wales, UK[51], and the effects on respiratory hospital admissions in residents close to the Peace Border crossing which experienced a large drop in traffic in the weeks after September 11 2001[62].

In our study we reported small respiratory effects of a reduction in air pollution. Most intervention studies described above have investigated other health endpoints, with other time frames, in other places, as a result of different policies, making a direct comparison limited. There are few other studies specifically investigated the effects of changes in air pollution over time (not related to policies) and changes in respiratory health within individuals. Downs et al.[63] studied whether an improvement in air quality over the years is followed by an improvement in lung function in adults. A reduction of 10 $\mu g/m^3$ PM_{10} over 11 years of follow-up slowed the annual rate of FEV_1 decline by 9%. In a subsample of the large Children's Health Study[e.g.64]

of children who moved away, Avol et al.[65] showed an improvement in lung function growth among those who moved to cleaner areas within a few years. Some additional evidence comes from cross-sectional studies over longer time periods. In cross-sectional analysis, a decline of coal combustion related air pollution in East Germany after reunification was associated with improvements of respiratory health in children[66,67]. This study was replicated in Swiss children, with much lower baseline and declines in air pollution[68].

Table 2 Summary of intervention studies on health effects of the LEZ and other (traffic) policies and actions

Study	Main policy / action	Design	Population	Health	Main results
Studies on the LEZ					
Cesaroni et al.[26]	LEZ for mainly (old) vehicles in the inner-city of Rome, Italy, 2001-2003.	With the use of concentration-response functions from the literature.	Children and adults	Mortality	Small mortality reductions in the zone.
Tonne et al.[27]	Congestion charging zone for all vehicles in the inner-city of London, UK, 2003.	With the use of concentration-response functions from the literature.	Children and adults	Mortality	Small mortality reductions in the zone.
Studies on short-term traffic policies / actions / unplanned events					
Friedman et al.[32]	Temporary traffic policies taken during the Olympic Games in Atlanta, USA, 1996.	Hospital counts during Games with 1 month before and after Games.	Children	Respiratory HA	Large reduction for respiratory HA in especially one registry (RR=0.48, 95 CI:0.44-0.86).
Peel et al.[33]	Temporary traffic policies taken during the Olympic Games in Atlanta, USA, 1996.	Same time frame as Friedman et al. Inclusion of larger area in Atlanta, control years and other cities.	Children and adults	Respiratory and cardio-vascular HA	No consistent significant reductions in respiratory and cardiovascular HA.
Li et al.[34]	Temporary traffic policies taken during the Olympic Games in Beijing, China, 2008.	Hospital counts during Games with pre-Games and 1 month before Games.	Adults	Asthma HA	A significant reduction in asthma HA (RR=0.54, 95 CI: 0.39-0.75).
Lin et al.[35]	Temporary traffic policies taken during the Olympic Games in Beijing, China, 2008.	Airway inflammation was measured during and 4 times measured before Games in a cohort within 2 years time.	Children	Airway inflammation	Substantial lower airway inflammation. 17-19% (95 CI:14-23) improvement per IQR decreases in BC and $PM_{2.5}$.
Lee et al.[37]	Temporary traffic policies taken during the Asian Games in Busan, Korea, 2002.	Hospital counts 3 weeks after Games with baseline (3 weeks before and during Games). Inclusion of control year.	Children	Respiratory HA	No consistent significant reductions in respiratory HA.

Table 2 Continued

Study	Main policy / action	Design	Population	Health	Main results
Studies on short-term traffic policies / actions / unplanned events (continued)					
Pope et al.[40]	An 11-month steel factory strike in Utah Valley, USA. 1986-1987.	Hospital counts during strike with 1 year before and after strike.	Children and adults	Respiratory HA	A 3 times strike-related decrease in childrens' admissions during winter closure.
Pope et al.[41]	A 9-months copper smelter strike in 4 USA states, 1967-1968.	Total monthly mortality counts for 4 states between 1960 and 1975.	Children and adults	Mortality	A significant strike-related decrease in mortality of 1.5-4.0%.
Studies on other more permanent (traffic) policies / actions – city level					
Clancy et al.[43]	Coal ban in Dublin, Ireland, 1990.	Total and cause-specific daily mortality counts 6 years after with 6 years before ban.	Children and adults	Mortality	Total, cardiovascular and respiratory mortality rates declined with 6-16% (95 CI: 4-19).
Hedley et al.[45]	A sulphur restriction in fuel, Hong Kong, China, 1990.	Total and cause-specific monthly mortality counts 1 year after with 1 year before the policy.	Children and adults	Mortality	Total, cardiovascular and respiratory mortality rates declined with 2-4% (95 CI: 0.3-6).
Peters et al.[55]	A sulphur restriction in fuel, Hong Kong, China, 1990.	Respiratory symptoms were assessed in spring 1991 and in 2 previous years in a cohort in 2 areas.	Children	Respiratory symptoms	A greater decline in the polluted compared with the less polluted area for some respiratory symptoms (e.g. wheezing).
Wong et al.[56]	A sulphur restriction in fuel, Hong Kong, China, 1990.	Bronchial hyper reactivity was assessed in 1 and 2 years after policy with 1 year before the policy in a cohort in 2 areas.	Children	Bronchial hyper reactivity	A greater decline in the polluted compared with the less polluted area for bronchial hyper reactivity in the 2nd year after policy, not in 1st year.
Noonan et al.[47]	A woodstove exchange in Montana, USA, 2005-2009.	Respiratory symptoms were assessed in 4 winters after exchange in a dynamic cohort.	Children	Respiratory symptoms	Weak evidence that respiratory symptoms were decreased due to the exchange.

Table 2 Continued

Study	Main policy / action	Design	Population	Health	Main results
Studies on other more permanent (traffic) policies / actions – city level (continued)					
El-Zein et al.[60]	Diesel-fuel ban in Beirut, Lebanon, 2002.	Hospital counts during 2 winters after with 2 winters before ban.	Children	Respiratory HA	Some evidence for a reduction in respiratory symptoms and upper respiratory tract infection 1 year after ban. Not in second year.
Macneill et al.[61]	Different traffic restrictions in the inner-city of Oxford, UK, 1999.	Respiratory effects were assessed at multiple time points before and after the policies in a dynamic cohort in 1998-2000.	Children	Lung function (PEF)	Improvements of PEF after implementation, beta = 5.52 l/min (96 CI: 3.08-7.97).
Studies on other more permanent (traffic) policies / actions - more local level					
Burr et al.[51]	A construction of a bypass in North Wales, UK, 1998.	Respiratory effects were assessed after bypass and 1 year before bypass in a cohort in two areas.	Children and adults	Respiratory symptoms, lung function (PEF)	Little evidence on most respiratory symptoms but rhinitis and rhinoconjunctivitis after the bypass. No evidence on PEF after bypass.
Lwebuga-Mukasa et al.[62]	A drop in traffic at the Peace Bridge border crossing USA – Canada after September 11 2001.	Hospital counts during a 3 months period before and after September 11 2001. Inclusion of a control year.	Children and adults	Respiratory HA	Some evidence of a reduction in respiratory hospital admissions in the weeks after September 11 2001.

HA= hospital admissions. CI= 95% confidence limits.

Methodological challenges in intervention studies

Most intervention studies have relied on modelling approaches and make use of existing concentration-response functions from the epidemiological literature[26,27,69] to estimate the health effects. In these studies the implicit assumption is made that causal effects will apply 'in reverse' when exposure is reduced. Intervention studies have the potential to give more direct evidence than observational studies, and therefore may contribute substantially to the causality debate. This does not imply that intervention studies are less prone to confounder variables biasing results. By design, in intervention studies the key challenge is to distinguish between policy related changes in air pollution and health, and other time-varying factors which can both influence air pollution concentrations as well as health. Below we discuss a few key issues and challenges identified in intervention studies based upon actual measurements.

Following a full chain approach

To evaluate policies a full chain approach has been proposed. Policies can act at different stages, starting from changes in pressures/driving forces, emissions, concentrations, exposures, doses and health effects[70] (See also Figure 1 in Chapter 1). Policies can fail at every stage of the full chain, starting with actual compliance. If a certain policy has been implemented, with poor enforcement, then the policy will be less effective. In our study, enforcement was tightened with cameras and special staff since 2010, with high fines for drivers entering illegally. At baseline enforcement was much lower, and a lot of official exceptions were given to old truck owners[23]. If a traffic policy is evaluated aimed at reducing traffic at a certain hotspot, and traffic is not influenced by the policy, it makes no sense to evaluate the other stages further down the chain. The traffic restriction measures taken in Atlanta, USA, during the 1996 Olympics resulted in a 2 to 20% traffic reduction at rush hours only, and almost none when assessing 24-hour total traffic counts[33]. Any policy-related change further down the chain would be extremely difficult to measure with such small differences in driving forces. Kelly et al.[29] evaluated the congestion charging zone in London, and decided to not estimate health effects after assessing changes in emissions and concentrations, because of only relatively small air pollution reductions. It is generally believed that in measured policy evaluations, air pollution needs to be reduced by at least a factor 1.5 to 2 in order to reliably detect the health effects of a change in air pollution[30], though the exact exposure contrast needed depends on population characteristics as well as the size and type of health effect under study. Almost all intervention studies dealt with much smaller (if any) air pollution reductions, and have difficulties finding a significant change in health status. The difficulty in detectability of health effects is often a reason to specifically study sensitive subgroups only (e.g. children, elderly, asthmatics), which may show stronger responses than the

general population. In our study we included all residents thus children, adults as well as elderly on the selected streets. Our response rate was already quite low (10%). If we would have selected a subgroup of the population in those streets, the number of people participating in the study would become even lower. Moreover, as we approached mostly families to participate in this study, we considered it inappropriate (and not easy to explain) to make a distinction between family members.

Background air pollution levels and weather

A major issue is the difficulty of detecting significant air quality improvements related to an intervention when air pollution concentrations change due to weather or other trends[30]. Studies investigating policies implemented within a short time frame (e.g. over one weekend), which are followed immediately by a drastically fall in air pollution, can show significant air quality improvements 'easier' than studies evaluating policies implemented more gradually, or policies that are only temporarily in place, with often smaller air pollution effects. In addition, these drastic falls in air pollution are more likely to occur when the policies are specifically targeted to reduce air pollution. They are less likely when the pollution changes are only a side effect, like the congestion charging zone in London where the primary aim was to reduce congestion[70].

Intervention studies showing an immediately drastic air pollution reduction are rare. Until now only the coal ban study in Dublin[43] and the studies that evaluate traffic measures taken at the Beijing Olympics[e.g.35] resulted in those substantial air pollution drops.

In our study we examined the effects of the LEZ, with increasingly stringent standards of which vehicles may enter the LEZ over the years. A more prolonged effect will only be visible when comparing longer time periods. Especially when assessing longer time trends careful adjustment for background and weather patterns is crucial[70].

An improvement in air quality was anticipated after the overnight implementation of the congestion charging zone in London, UK, since this led to immediate reductions in traffic volumes entering the zone. However, at the same time unusual meteorological conditions (an inversion) had led to periods of elevated air pollution regional-wide, that may have obscured observation of any air quality improvement due to the zone[29]. When they evaluated longer time frames (e.g. two years before and after implementation) results were more stable. The opposite was the case in the evaluation of several traffic measures taken at the summer Olympics in Atlanta, USA. The substantial reductions in ozone of about 30% during the Olympics that has been attributed to the policies before[32] were also occurring in other cities throughout the Southeast of the USA where the policies were not in place, indicating that large scale (favourable) weather conditions may have played a large role in the ozone reductions[33]. In the study of coal bans in Ireland, the later bans in other Irish cities showed often much smaller

reductions than the initial coal ban in Dublin[43], probably due to due to lower baseline pollution, among other factors[44].
Thus apart from traffic, other sources of air pollution on both the local and regional scale, coupled with varying meteorological conditions could all bias air pollution trends attributed to the policy. As background and weather patterns can not be predicted beforehand, intervention studies should at least evaluate multiple time windows surrounding the intervention and compare changes at control areas not affected by the policy under investigation[30]. An additional method is to evaluate whether changes will become smaller when the distance to the policy-affected-area increases[28].

Competing impacts on air quality and health
In intervention studies, the actual air pollution effect of for example the LEZ may be counteracted or coincided by other traffic (policy) changes over time. In our study the LEZ went in parallel with the autonomous trend in replacement of old vehicles between 2008 and 2010. Other competing impacts on air quality in our study may be the relative increase of new diesel cars and diesel particle traps in the average car park in recent years, causing rising emissions of especially soot and NO_2. In our study, another change in the average car park between 2008 and 2010 may be due to a relatively increase in old polluting passenger cars in the Netherlands from abroad (mainly from Germany where they are not allowed any more since the implementation of LEZ directed to all old vehicles). Another potential explanation for not finding a substantial LEZ effect can be that because of the economic crisis that affected the Netherlands in 2008, pollution emissions may have decreased everywhere, making it even harder to detect a small LEZ related change.
Other intervention studies struggled with competing impacts as well. For example due to the implementation of the congestion charging zone in London, the number of public buses and taxis entering the zone increased, as well as the traffic just outside the zone[29].
Population health trends – unrelated to the policy under investigation - may bias the observed health response of the policy. Age is by far the strongest predictor of health effects, and changes in age distribution of the general population can produce substantial shifts in health. In addition, population health may change over time due to changes in health care, medication use, lifestyle factors including diet as well as other factors. Especially in policy evaluation studies with longer time frames correction for autonomous health trends are crucial, like in the study of Pope et al.[41], which estimated mortality effects of a copper smelter strike in four Southwest USA states over a prolonged period of fifteen years. An example of a concurrent process that went hand in hand with a decline of coal combustion related air pollution in East Germany after reunification was the 'westernisation' process experienced in East German communities. This change in living conditions towards a Western lifestyle impacted health considerably as well. Thus the associations reported[e.g.66,67] between decreases in air pollution and improvements in health status could be due to other factors than air

pollution. However, results were confirmed in Swiss children, which also experienced a decline in air pollution over the years, but without the 'westernisation' process that could have confounded the reported effects[68].
A traffic policy never occurs in isolation, and there will always be more driving forces behind changes in air pollution and health at the same time as this is inherently associated with intervention type of research in the real-world. In our study, autonomous trends in health will be limited since the study covers a period of two years, a relatively short time period.

Inclusion of control areas / populations
Temporal changes in air pollution as well as health can be influenced by other changing risk factors. Inclusion of control areas not affected by the policy is one method to capture this partly. Many intervention studies have not included a control population. Almost all smoking ban studies have not included a control population group often because of the simple fact that they were not available as the smoking ban in public places was applied nationally[71-73]. A comparison with a city in for example another country would lead to comparability issues because of marked differences in smoking habits and diverging time trends. One smoking ban study in the Republic of Ireland managed to include a control population in the neighbouring Northern Ireland, with a more or less comparable population[74]. As there is no clear-cut answer which control population reflects temporal trends best, some studies showed different options. In a study of Pope et al.[41] they estimated mortality effects of a copper smelter strike in four Southwest US states over a prolonged period of fifteen years. They corrected for mortality trends including either the rest of the US, Eastern US states or neighbouring and bordering states. Reported effects were quite stable apart from a consistent smaller effect when they made use of the bordering states mortality trends[41]. To adjust for temporal mortality trends unrelated to the coal ban implemented in multiple Irish cities, mortality trends from rural populations in Ireland were used[70]. Rural populations are different from city residents in other characteristics like age, socioeconomic and individual factors like smoking, and may have less access to health care. However, often there are no better alternative control populations readily available.
It is not straightforward to select proper control areas. Key issue is that autonomous temporal trends (both in air pollution and health) at the control areas are comparable with the policy-affected locations. In practice, this means that the control population should be at least comparable in terms of age and socioeconomic status to the population under investigation, and that control locations should be as close as possible to the other locations under study. Inclusion of control populations more or less close to the intervened area may be preferable to populations from more distant areas or even from abroad because of a higher chance of comparability. But at the same time, areas closer to the intervened areas may also benefit from the policy - although to a lesser extent - and therefore using those populations as controls can lead to overcontrolling and underestimating the policy-related effects. It

is especially difficult to find appropriate control areas for policies with dramatic changes, and for policies applied (almost) nation-wide like the coal ban in Irish cities, or the smoke ban in pubic places.
In our study we have chosen suburban background locations in the vicinity of the cities as control areas. Population characteristics may be somewhat different at the control locations, although we tried to include participants with similar age and social economic status structure. We reported pronounced improvement of lung function in a small group of residents living at the one street where both traffic and air pollution was drastically reduced. These effects were robust, and not only visible compared to their matching suburban control, but also when compared to all suburban background locations, as well as compared to the other streets in our study. In our study the ideal control locations would have been major streets in other comparable Dutch cities, but where the traffic remains constant over the years, where no local traffic policies took place, and with comparable population characteristics. These locations were however not available.

Choice of time window surrounding policies

Timing of air pollution effects
Another difficulty often associated with more prolonged studies is the timing that a policy is believed to be fully into place. The study of Noonan et al.[47] investigated a woodstove exchange programme in Montana, USA. The study was complicated by the fact that the exchange occurred over three winters. In the first year after the policy only about 13% of the woodstoves were exchanged, and no effects were visible on outdoor $PM_{2.5}$. In the congestion charging zone study in London, they explored extensively at what time scale (weeks, months, years after the implementation) the policy should be analysed[29]. In our study, we believed that effects of the LEZ would be fully visible within two years time after implementation. We may have missed some LEZ effect as the LEZ was already implemented before our baseline measurement campaign. However, this is unlikely a large part, since regulations of which types of trucks were allowed became more stringent and enforcement was tightened after baseline.
Choice of timeline surrounding the implemented policies is far from trivial. Especially in prospective intervention studies, flexibility and rapid planning is needed since policy (plans) often change as a result of different legislation. Initially our study was set up to evaluate more traffic policies than the LEZ, including for example a complete truck ban on certain urban streets. However, local governments decided not to implement those local policies at a very late stadium in the decision-making process, probably a result of the fact that permission was obtained to postpone compliance to EU air quality standards up to 2015 instead of 2010. To study interventions often an opportunistic approach is taken, with multiple time windows surrounding the implementation of the policy.

Timing of health effects

When air pollution concentration was substantially reduced during the 2008 Beijing Olympic Games because of the traffic policies, this was almost immediately followed by an improvement in airway inflammation[35]. Avol et al.[65] showed an improvement in lung function growth among children who moved to cleaner areas within already one year. In a small study where they moved asthmatic children from a high polluted Italian city to a less polluted rural environment to visit a one-week school camp, respiratory benefits were already present after that week[75]. That beneficial respiratory effects can be expected within a relatively short time period after a drastic change in air pollution levels, have been seen in some other research fields as well. Several studies investigating bar workers before and after the smoking ban in public places like bars and restaurants. They showed beneficial effects on the lungs up to 8% already within a few months after the ban[72,73].

Modelling vs. measuring health impacts

Most intervention studies have relied on modelling approaches and make use of existing concentration-response functions from the epidemiological literature[26,27,69] to estimate the health effects. Modelling studies allowed isolating a policy effect by keeping all other influencing factors (e.g. background trends in air pollution and health) constant over time. Furthermore, changes in air pollution concentrations due to the policy under investigation are often relatively small: associated changes in health status would have been difficult to detect using routinely collected measurement data. Moreover, modelling the health responses is by far less demanding than measurement studies, and can be done either retrospectively or in prospect of a certain traffic policy. On the other hand, policy evaluation by measurements reflects real-world conditions, which is an advantage as well. Other advantages of empirical studies include that health effects are directly measured in the population under study. Moreover, a measuring approach allowed studying more subtle health effects as well including lung function, thus effects which are not routinely collected and not easily captured in modelling approaches. Responses of subtle health effects may be more substantial and earlier visible in the population under study.

At the same time both modelling and measuring approaches have their drawbacks as well. Transferability of concentration-response functions to other locations and populations may introduce additional uncertainty in a modelled evaluation. Modelling approaches make assumptions when applying certain concentration-response functions like that there are no important differences in the distribution of factors influencing susceptibility or toxicity of air pollution around different populations and locations, and that the exposure assessment in these studies are comparable to the modelling approach, which may not be particularly true[27]. Studies in which also the effect of the policy on air pollution is modelled have further drawbacks. Assumptions about the effectiveness of the policy in realising changes in driving forces (traffic intensity, shift to cleaner cars) and emissions may not

be correct. In our study there were significant uncertainties about emission factors of old vs. new trucks. Finally, there are uncertainties with respect to modelling concentration changes. Drawbacks of a measuring approach include that a relatively large air pollution reduction is needed to reliable detect the health effects of an air pollution change, and that therefore often a large sample size have to be available. Even relatively large evaluation studies with measured data like Peters et al.[46] assessing the mortality effects in Erfurt, Germany, with approximately 200 000 inhabitants, have run into power problems, also because of slower, less well defined changes in pollution sources. To measure certain longer-term health benefits of air pollution changes such as in cardiovascular and respiratory diseases and eventually mortality, long study periods are needed, which can be a disadvantage as well. As discussed earlier competing risks is another problem in empirical studies. Despite these problems empirical studies should be the method of choice, if feasible.

Implications of this study

We demonstrated that local traffic policies can contribute to air quality improvements and health gains of residents nearby. Although the observed improvements in respiratory function in this study were small, and likely insignificant in healthy subjects[76], a small shift in population mean may result in more substantial shifts in the people with low lung function.

Especially at the street in The Hague where traffic was drastically reduced, a substantial improvement in both air quality and health was measured within two years time. This supports local policies that have included improvement of population health as an additional goal to compliance with air quality standards, which is typically the main driver of policies. Reduction of emissions even when concentrations are already below the air quality standards will likely improve public health as epidemiological studies have not observed a threshold below which no adverse health effect occurred. As part of the local policies implemented in The Hague, traffic was mostly redirected from busy streets in the inner-city to other less busy streets outside the city centre. Therefore the air pollution health burden may have increased in these other streets. From a health point of view, traffic policies aimed to reduce overall traffic load in cities are by far preferable than redistribution of traffic to other residential urban areas.

Until now, most traffic policies are directed to reduce traffic emissions. Examples of emission reduction policies include European-wide policies like emission standards for motor vehicles, regulations for fuel, but also local policies like the LEZ. Although these regulations have clearly led to decreased emissions of traffic, increases in urban populations, number of cars, vehicle km travelled and land-use practises are trends that have partly offset their effectiveness[1]. New strategies to address and mitigate air pollution related health impacts are starting to emerge, which focused more on separating

people from pollution[77]. Recently in the Netherlands, a policy was approved that prohibits new buildings like schools and hospitals close to major roads where air quality standards are exceeded.
Health is an issue of increased interest within the complex decision-making process nowadays in the Netherlands as well as in other Western countries. Our study demonstrated that significant health gains can be achieved within a relatively short time period when traffic is drastically reduced, which is encouraging for policy makers. Earlier cost benefit analyses have clearly shown that the monetised benefits of further pollution reduction in terms of health gains outweigh the costs of further air pollution reductions[78].

Conclusion

Our study showed that the contrast between major streets and urban background locations was somewhat larger for non-tailpipe traffic emission indicators such as Cu and Fe, than for tailpipe emission indicators. PNC and soot are both combustion-related indicators mostly emitted from the engine. The study confirmed the evidence that proximity to traffic is much better reflected in contrasts in particle number concentrations (PNC) or soot than in PM_{10} and $PM_{2.5}$ levels. Contrast between busy streets and background locations for soot and PNC were similar.
This was the first study that showed that oxidative potential of PM - measured with the EPR assay - near major urban roads was highly elevated compared with background locations. The oxidative potential contrast was greater than that for any other measured PM characteristic. When the same assay was applied two years later by another laboratory, results were incomparable with the baseline measurements, preventing an analysis of the effects of the traffic policies including LEZ on oxidative potential.
With the exception of one street, the local traffic policies including LEZ were too modest to produce statistically significant decreases in traffic-related air pollution concentrations. In the one street where traffic was drastically reduced, traffic-related air pollutants decreased significantly as well. Especially in that street, lung function was significantly improved in the residents compared to their matching control population within two years time.
This study adds evidence that local traffic policies contribute to air quality improvements at street level and health gains of residents nearby. Our results contribute to the small number of studies actually measuring the effects of (traffic) policies on air pollution concentrations and health.

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Summary

Associations between air pollution and numerous adverse health effects are now well established. In the past decades substantial efforts have been taken to diminish air pollution emissions from industry as well as motorized traffic. Although these have clearly led to reduced emissions, increases in urban populations, number of cars and vehicle km travelled are a few trends that may partly offset their effectiveness. To protect public health, the European Union (EU) has set air quality standards of which the standards for particulate matter (particles smaller than 10 µm (PM_{10}) and smaller than 2.5 µm ($PM_{2.5}$)) and nitrogen dioxide (NO_2) are the most important. Air quality standards are still exceeded especially near busy roads. Therefore, local policies have been implemented to reduce air pollution concentrations at busy urban roads. Examples of local traffic policy measures include congestion charging directed to all passenger cars entering inner-cities, and low emissions zones (LEZ) designed to limit traffic of old vehicles in city centres. Until now, there is little direct evidence of the effect of local policies in reducing air pollution and ultimately improving health.

The aim of this thesis is to assess the effect of local traffic policy measures on 1) air quality; 2) oxidative potential of PM and 3) respiratory health of residents.

Measurements of air pollution and respiratory health were conducted at eighteen locations in the Netherlands before (2008) and two years after the implementation of the policies (2010). In this study the LEZ was evaluated, which forbids old heavy duty vehicles (trucks) to enter the LEZ, typically the inner-city. LEZ is a popular local policy not only in the Netherlands. To date, LEZ have been implemented in 152 cities in nine EU countries, as well as in other cities in other parts of the world (e.g. Singapore, Tokyo). Additionally, a traffic recirculation plan was evaluated in one city (The Hague), designed to reduce air pollution concentrations at hotspots. Air pollution measurements were performed at eight busy urban streets in five Dutch cities, six urban background locations and four suburban background locations near the selected cities. Suburban background locations were chosen as control areas, likely not affected by the policies under investigation to adjust for difference between 2008 and 2010 in other factors like the weather. Simultaneously at each location, six weekly air samples were collected spread over two six-month periods in 2008 and 2010 respectively, with identical equipment and at exactly the same place. Respiratory function was assessed twice by

spirometry and interrupter airway resistance. In addition, NO in exhaled air was measured as a marker for airway inflammation. Health measurements were conducted at the urban streets and suburban background locations. Baseline and follow-up respiratory function tests were performed in 661 residents in winter 2009 and 2011, following the majority of the air pollution measurements.

Traffic-related air pollution originates from combustion and wear of tyres, brakes and road surface (non-tailpipe emissions) and is a complex mixture of many particulate and gaseous pollutants. Omission of a more traffic-related component in a traffic policy evaluation may lead to an underestimation of the health impacts, because reductions of these components are likely more substantial in response to these policies than reductions in the more general air quality indicators like PM_{10} and $PM_{2.5}$. In chapter 2 and 3 we first compare the difference in concentration between major urban roads and background locations for a large range of regulated and non-regulated air pollutants, prior to the introduction of the policies. Chapter 4 assesses the contrast in oxidative potential of PM between major urban roads and background locations, before the implementation of the local policies. High contrasts between busy streets and background locations in the same city were found for chromium (Cr), copper (Cu) and iron (Fe) (factor 2-3), indicative of mainly non-tailpipe traffic emissions. These elements were especially present in the coarse fraction of PM. In addition, high contrasts were found for soot and NO_x (factor 1.8-1.9), indicators of combustion emissions. Contrasts were lower for the regulated components PM_{10}, $PM_{2.5}$ and NO_2 (factor 1.2-1.5). The contrast for PNC was similar to soot in a subset of streets and corresponding background locations. Oxidative potential of PM_{10} measured with the EPR assay showed the largest contrast between busy streets and urban and suburban background locations (factor 4-7).

Chapter 5 presents an analysis of air pollution concentrations before and after the implementation of local traffic policies including the LEZ. Overall, the 2010-2008 concentration trends of traffic-related pollutants (e.g. soot, NO_2, NO_x, Cu, Fe) did not differ significantly for busy urban streets compared to suburban background locations outside of the LEZ, though there were substantial differences across streets. This suggests that LEZ policies directed at old heavy duty vehicles (trucks) did not substantially affect traffic-related air pollution concentrations. We did find a reduction in $PM_{2.5}$ that was significantly larger at the busy urban streets than at the control locations, but it was unlikely due to the LEZ. In the one street in The Hague - where in addition to the LEZ - traffic intensity was substantially reduced, soot, NO_x and NO_2 concentrations decreased significantly more (41, 36 and 25%) than at the corresponding suburban background location (22, 14 and 7%). With the exception of that street, the local traffic policies including LEZ were probably too modest to produce statistically significant decreases in traffic-related air pollution concentrations. When the same oxidative potential assay was applied two years later by another laboratory, results were incomparable with the baseline measurements, preventing an analysis of the effects of the traffic policies on oxidative potential.

Chapter 6 describes the relation between changes in air quality and changes in the respiratory health status of residents. Especially in the one street in The Hague where traffic and air pollution concentrations was drastically reduced, lung function (FVC, FEV_1) was improved with 3-5% in those residents compared to suburban background residents. This was accompanied with a suggestive reduction in airway resistance. In the total population, small improvements in lung function (FVC) were found up to 1% after a decrease in air pollution within two years time. No significant associations were seen for other spirometric measures and exhaled NO.
This study adds evidence that local traffic policies contribute to air quality improvements at street level, resulting in health gains of residents nearby.

Samenvatting

In de afgelopen jaren hebben veel studies een verband tussen luchtverontreiniging en gezondheid gevonden. Er zijn al veel maatregelen getroffen om emissies van luchtverontreiniging vanuit de industrie en het gemotoriseerd verkeer te laten dalen. Dit heeft zeker tot een afname in emissies geleid, hoewel een toename van de bevolking, voertuigen, aantal kilometers per voertuig de emissieafname deels teniet heeft gedaan. Om de gezondheid van de bevolking te beschermen heeft de Europese Unie (EU) luchtkwaliteitsnormen vastgesteld waarvan de normen voor fijn stof (deeltjes kleiner dan 10 µm (PM_{10}) en kleiner dan 2,5 µm ($PM_{2.5}$)) en stikstofdioxide (NO_2) de meest belangrijke zijn. De luchtkwaliteitsnormen worden nog steeds overschreden, vooral op drukke wegen. Daarom worden lokale verkeers-maatregelen ingevoerd om luchtverontreiniging terug te dringen op drukke binnenstedelijke wegen. Voorbeelden van lokale verkeersmaatregelen zijn een tolheffing voor alle voertuigen die een stadscentrum in willen rijden, en de invoering van milieuzones die gericht zijn op het weren van oude voertuigen uit het stadscentrum. Het effect van lokale maatregelen op de luchtkwaliteit en gezondheid is tot nu toe weinig onderzocht.

Het doel van dit proefschrift is om het effect van lokale verkeersmaatregelen te onderzoeken op 1) luchtkwaliteit; 2) oxidatieve potentieel van fijn stof en 3) de respiratoire gezondheid van omwonenden.

Metingen van luchtverontreiniging en respiratoire gezondheid zijn uitgevoerd op 18 locaties in Nederland voor (2008) en twee jaar na de invoering van de lokale maatregelen (2010). In deze studie is de milieuzone geëvalueerd, die is ingevoerd in het centrum van veel Nederlandse steden. De milieuzone is gericht op het weren van oude vrachtwagens. De milieuzone is een populaire maatregel, niet alleen in Nederland. Tot nu toe zijn milieuzones geïmplementeerd in 152 steden in negen EU landen en in een aantal grote steden buiten Europa (bijvoorbeeld Singapore, Tokyo). Daarnaast is een verkeerscirculatieplan geëvalueerd in één stad (Den Haag). Dit plan is gericht op een vermindering van luchtverontreiniging in drukke straten in Den Haag waar de luchtkwaliteitsnormen worden overschreden. Er zijn luchtmetingen verricht op acht drukke straten in vijf Nederlandse steden, zes rustige achtergrondlocaties in de stad en vier rustige achtergrondlocaties in gemeenten in de omgeving van de steden. De vier rustige achtergrondlocaties waren geselecteerd als controlelocaties, en worden niet beïnvloed door de onderzochte maatregelen. De controlelocaties zijn gekozen om te corrigeren

voor verschillen tussen 2008 en 2010 in andere factoren, zoals het weer. Gelijktijdig zijn er op elke locatie zes weekgemiddelde metingen van de luchtverontreiniging uitgevoerd, verspreid over een half jaar in 2008 en in 2010, met identieke apparatuur en op exact dezelfde plek. De respiratoire gezondheid is twee keer gemeten door middel van standaard longfunctietesten (spirometrie en luchtwegweerstand). Ook is stikstofoxide in uitgeademde lucht bepaald, als een marker voor ontstekingen in de bovenste luchtwegen. Voor- en nameting van de longtesten is uitgevoerd bij 661 omwonenden in de winter van 2009 en 2011, nadat het grootste gedeelte van de luchtmetingen verzameld was.

Verkeersgerelateerde luchtverontreiniging is een complex mengsel van deeltjes en gassen. In een straat draagt het verkeer bij aan emissies via de uitlaat door verbrandingsprocessen, door banden- en remslijpsel, en het opwervelen van straatstof (niet-uitlaatemissies). In een verkeersmaatregelen-evaluatie kan het weglaten van een meer verkeersgerelateerde component tot een onderschatting van de gezondheidseffecten leiden, omdat de verkeersmaatregel waarschijnlijk een groter effect heeft op verkeers-gerelateerde componenten dan op andere componenten die meer een indicator zijn voor generieke luchtkwaliteit zoals fijn stof. In hoofdstuk 2 en 3 vergelijken we eerst het verschil in concentraties op drukke straten en rustige achtergrondlocaties voor een groot aantal gereguleerde en niet-gereguleerde componenten, voorafgaand aan de invoering van de lokale maatregelen. Hoofdstuk 4 beschrijft een analyse van het verschil in het oxidatieve potentieel van het fijn stof tussen drukke straten en rustige achtergrond-locaties, voor de invoering van de lokale maatregelen. Grote verschillen tussen drukke straten en rustige achtergrondlocaties in de stad zijn gemeten voor chroom, koper en ijzer, indicatoren voor hoofdzakelijk niet-uitlaat verkeersemissies (2 tot 3 keer zo hoog op de straten). Deze componenten waren vooral in de grovere fractie van het fijn stof verhoogd. Ook zijn er grote verschillen gemeten voor roet en NO_x (bijna 2 keer zo hoog op de straten), indicatoren voor uitlaatemissies (verbranding). Verschillen waren veel kleiner voor de gereguleerde componenten PM_{10}, $PM_{2.5}$ en NO_2 (1.2-1.5 keer zo hoog op de straten). Het verschil voor ultrafijn stof was net zo groot als voor roet in een subset van drukke straten en rustige achtergrondlocaties. Het grootste verschil tussen drukke straten en rustige achtergrondlocaties in en om de stad is gevonden in de oxidatieve potentieel metingen van het fijn stof (4 tot 7 keer zo hoog op de straten), gemeten met behulp van het EPR-assay.
Hoofdstuk 5 bevat een analyse van luchtverontreiniging voor en na de invoering van de lokale maatregelen waaronder de milieuzone. Gemiddeld was de 2010-2008 concentratie trend van verkeersgerelateerde lucht-verontreiniging (zoals roet, NO_2, NO_x, koper, ijzer) niet verschillend op de drukke straten, van de trend op de rustige controlelocaties buiten de milieuzones, hoewel er verschillen waren tussen de drukke straten. Dit suggereert dat de milieuzone gericht op oude vrachtwagens geen meetbaar effect heeft gehad op concentraties van verkeersgerelateerde lucht-

verontreiniging. Wel was er een grotere afname van $PM_{2.5}$ gemeten in de drukke straten dan op de rustige controlelocaties, maar waarschijnlijk was dit niet toe te schrijven aan de milieuzone. In de enige straat in Den Haag, waar naast de milieuzone ook het verkeer flink was afgenomen, waren vooral roet, NO_x en NO_2 concentraties meer gedaald (41, 36 en 25%) dan op de rustige controlelocatie (22, 14 en 7%). Met uitzondering van deze straat, waren de effecten van de lokale maatregelen waaronder de milieuzone op de verkeersgerelateerde luchtverontreiniging te klein om meetbaar te zijn. Het effect van de lokale maatregelen op het oxidatieve potentieel van het fijn stof kon niet onderzocht worden omdat dezelfde laboratoriumanalyse twee jaar later gedaan is door een ander laboratorium, en de resultaten onvergelijkbaar waren met de voormeting.

Hoofdstuk 6 geeft de resultaten weer van de relatie tussen verschillen in luchtkwaliteit en verschillen in de respiratoire gezondheid van omwonenden. Vooral in de straat in Den Haag waar zowel het verkeer als de luchtverontreiniging flink was afgenomen, was de longfunctie (FVC, FEV_1) van omwonenden verbeterd met 3-5%, vergeleken met de controlepopulatie op rustige achtergrondlocaties buiten de stad. Ook was de luchtwegweerstand afgenomen in deze omwonenden, hoewel minder consistent. Gemiddeld over de hele populatie waren er kleine verbeteringen in longfunctie (~1%) meetbaar na een afname van luchtverontreiniging in twee jaar tijd. Er is geen significante relatie gevonden in andere longfunctiematen en in stikstofoxide in uitgeademde lucht.

De resultaten van dit proefschrift laten zien dat lokale maatregelen bijdragen aan een verbetering van luchtkwaliteit op straatniveau en van gezondheid van omwonenden.

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About the author

Johanna Maria Cornelia (Hanna) Boogaard was born in Utrecht, the Netherlands on December 5 1982. In 2001, she graduated from secondary school at the Driestar College in Gouda, and started her study in Environmental Health Sciences with an additional MSc in Epidemiology at Maastricht University, the Netherlands. As part of her study she investigated the effects of long-term exposure to traffic-related air pollution on incidence and mortality of bladder cancer in a large Netherlands Cohort Study on Diet and Cancer (NLCS) at Maastricht University, in collaboration with the Institute for Risk Assessment Sciences (IRAS), Utrecht University, the Netherlands. After graduation in 2005, she started working as a researcher at IRAS on several (inter)national projects, including the INTARESE project (Integrated Assessment of Health Risks of Environmental Stressors in Europe), and the CAIR4HEALTH project (Clean Air for Health). In addition, from 2008 she worked on a study to evaluate traffic policy measures in the Netherlands as described in this thesis. She will continue working in the air pollution and health field as a staff epidemiologist at the Health Effects Institute (HEI) in Boston, MA, USA.

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'Laat de rollator daar maar staan, ja ik draag u wel'
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