Variation of NO₂ and NOₓ concentrations between and within 36 European study areas: Results from the ESCAPE study

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HIGHLIGHTS

► We measured NO_2 and NO_x in 36 European study areas using standardized method.
► Significant contrast in NO_2 and NO_x levels between and within areas were found.
► Concentrations were generally lower in Northern than in Southern Europe.
► Street/urban background contrast was higher than for the particle metrics.
► Epidemiological studies should characterize intra-urban contrasts.

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ABSTRACT

The ESCAPE study (European Study of Cohorts for Air Pollution Effects) investigates long-term effects of exposure to air pollution on human health in Europe. This paper documents the spatial variation of measured NO_2 and NO_x concentrations between and within 36 ESCAPE study areas across Europe.

In all study areas NO_2 and NO_x were measured using standardized methods between October 2008 and April 2011. On average, 41 sites were selected per study area, including regional and urban background as well as street sites. The measurements were conducted in three different seasons, using Ogawa badges. Average concentrations for each site were calculated after adjustment for temporal variation using data obtained from a routine monitor background site.

Substantial spatial variability was found in NO_2 and NO_x concentrations between and within study areas; 40% of the overall NO_2 variance was attributable to the variability between study areas and 60% to variability within study areas. The corresponding values for NO_x were 30% and 70%. The within-area spatial variability was mostly determined by differences between street and urban background concentrations. The street/urban background concentration ratio for NO_2 varied between 1.09 and 3.16 across areas. The highest median concentrations were observed in Southern Europe, the lowest in Northern Europe.

In conclusion, we found significant contrasts in annual average NO_2 and NO_x concentrations between and especially within 36 study areas across Europe. Epidemiological long-term studies should therefore consider different approaches for better characterization of the intra-urban contrasts, either by increasing of the number of monitors or by modelling.

1. Introduction

There is now increasing evidence from epidemiological studies that exposure to ambient air pollution is associated with adverse health effects (Brunekreef and Holgate, 2002; Heinrich and Wichmann, 2004; Pope and Dockery, 2006; WHO, 2006; Rückerl et al., 2011). Adverse effects include pre-mature mortality and morbidity from cardiovascular and respiratory causes. Based upon experimental studies, plausible mechanisms for these associations have been proposed, including oxidative stress in particular (Brunekreef and Holgate, 2002; Brook et al., 2010; Rückerl et al., 2011). Most studies from the USA have focussed on PM_{10} and $PM_{2.5}$ (Brook et al., 2010; Pope and Dockery, 2006). In several European studies, significant associations between adverse health effects and NO_2 or NO_x concentrations (Brunekreef, 2007) have been reported. In these studies, air pollution exposure was assessed at the residential address using dispersion models, land use regression models and traffic indicator variables. Other epidemiological studies on long-term exposure to NO_2 and other air pollutants compared the health status of populations using the contrast in city-average air pollution levels between different areas (e.g. Pope et al., 2002; Laden et al., 2006; Sunyer et al., 2006; Götschi et al., 2008). These studies generally assigned one overall average concentration to all subjects living in each city. For NO_2 this likely results in significant misclassification as high spatial variability within urban areas has been documented previously for nitrogen dioxide (NO_2) in specifically designed studies (Lebret et al., 2000; Monn, 2001;

Lewne et al., 2004). There are a substantial number of studies that have used traffic indicators as exposure variables, including distance to a major road, and traffic intensity on the nearest road (HEI, 2010). A major limitation of these traffic indicators is that their value in characterizing actual air pollution exposure contrasts may differ between study areas (Jerrett et al., 2005). Some studies have made use of the spatial variation of air pollution within metropolitan areas (Gauderman et al., 2005; Gehring et al., 2006; Morgenstern et al., 2008; Jacquemin et al., 2009; Modig et al., 2009). These within-city studies often characterized air pollution with the concentration of NO_2 and NO_x obtained from either spatially dense monitoring networks, land use regression models based upon such networks, or dispersion models (Jerrett et al., 2005; Hoek et al., 2008; Modig et al., 2009; Levy et al., 2010).

In 1999, the European Commission established limit values for NO_2, NO_x, PM_{10} and some other air pollutants in the Air Quality Daughter Directive 1999/30/EC (EC, 1996), which was replaced in 2008 by the new Directive 2008/50/EC on ambient air quality and cleaner air for Europe (EC, 2008). The existing air quality guidelines for NO_2 and PM_{10} are currently being exceeded at many locations throughout Europe (Giannouli et al., 2011; European Environment Agency, 2006; Airbase, 2007; Velders and Diederen, 2009). There is therefore substantial interest at the EU policy level in the health effects of current air pollution levels including NO_2, focussing especially on European studies.

A comparison of NO_2 concentrations measured either in study specific monitoring programmes (Hazenkamp-von Arxa et al.,

2004) or in routine monitoring networks across Europe (e.g. Airbase data used in Beelen et al., 2009) showed significant contrast across Europe. The concentrations were generally lowest in Northern Europe and highest in the major cities and Southern Europe.

NO_2 is often used as an indicator of the complex mixture of traffic-related air pollution containing also fine and ultrafine particles. The ratio of NO_2 to other components e.g. soot in emissions of motorized road traffic has changed in the last decade (Williams and Carslaw, 2011). Specifically, the fraction of primary NO_2 emissions has increased.

In 2008 we embarked upon a European-wide study of long-term air pollution exposure health effects. The ESCAPE study – **E**uropean **S**tudy of **C**ohorts for **A**ir **P**ollution **E**ffects – assesses exposure–response relationships between long-term exposures to ambient air pollution using prospective cohort studies in 15 different European countries (http://www.escapeproject.eu). As the estimation of the within-urban variation of air pollution is a key interest of epidemiological long-term studies and the most routine monitoring networks are not sufficiently dense to characterize intra-urban concentration gradients, we decided to carry out study specific monitoring, which was independent of routine monitoring networks.

In the framework of this study we conducted measurements of NO_2 and NO_x concentrations in 36 study areas during a one year long measurement period. The aim of this paper is to assess the variation of measured NO_2 and NO_x concentrations between and within 36 European study areas. We further assessed the difference of NO_2 and NO_x concentrations at traffic stations versus urban background stations as a source of within study area spatial variability. The third aim was to study the variability of the NO_2/NO_x ratio. The companion paper focuses on the PM measurement (Eeftens et al., submitted for publication).

2. Methods

2.1. ESCAPE exposure assessment

In all study areas, NO_2 and NO_x were measured with passive samplers (see Section 2.4). The measured average concentrations were combined with geographic predictors to develop land use regression (LUR) models (Jerrett et al., 2005; Hoek et al., 2008). In all study centres a common protocol was used to ensure high standardization of all procedures across the 36 European study areas. The standardization of the measurements and the selection of the locations using a common protocol across a wide range of European settings (i.e. >1400 monitoring sites across Europe) is one of the major strengths of this study.

2.2. Study areas

ESCAPE included 36 study areas across Europe (Fig. 1). An overview of the characteristics of study areas is presented in Table 1 and in more detail in Online supplement A. The study areas were of substantially different size. Please note that the study areas in Table 1 are sorted from the North to the South. We kept this order in all following tables and figures.

The size of the areas is given by the distribution of the included cohorts. Some areas were restricted to a single city (e.g. Rome, Grenoble, Erfurt) or a metropolitan area including more rural/suburban areas (Stockholm country, Athens). The study also included large study areas such as the Ruhr area, Catalunya and the Netherlands/Belgium. The included main cities were also of very different size, ranging from Umeå (∼100,000 inhabitants) to the largest European metropolitan areas such as Paris and London with several million inhabitants.

As indicated in Fig. 1, in 20 study areas both particulate matter ($PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10}, and PM_{coarse}) as well as NO_2 and NO_x were measured (in the following they are labelled as "NO_x + PM" study areas), whereas in the remaining 16 study areas NO_2 and NO_x was measured only (labelled as "NO_x only" study areas). In total, NO_2 and NO_x measurements were performed at 1483 monitoring sites in 36 study areas (see Table 1). This means that, on average, 41 sites per study area were selected. However, due to the differences between the study areas, the number of regional, urban background and street sites in each study area is also different. In the largest study areas (Netherlands/Belgium and Catalunya) 80 monitoring sites were selected, whereas in the two rather small study areas Granada and Hulvea (Spain) less locations were selected (14 and 24, respectively).

2.3. Site selection

The spatial distribution of the cohort subject addresses determined the borders of the study area. Within each study area the measurement sites were selected to represent the anticipated spatial variation of air pollution at home addresses of subjects in the cohort studies. The long-term average ambient air concentration is a function of the regional background, additional pollution from all (sub)urban sources (resulting in an urban background) and pollution from local sources (e.g. traffic on nearby busy streets). In all areas street sites were overrepresented compared to the fraction of addresses on major roads, as the goal was to describe spatial variation in the area of which traffic is a main source. The requirement was to select a range of realistic traffic intensities, not only the busiest streets in the area. Other sources were also considered, e.g. specific industries, major ports. In some areas, altitude was also a factor in selecting sampling sites.

The measurement sites were classified as regional background, urban background and street sites. A street site was considered a site in a major road carrying at least 10,000 vehicles per day. Measurements were typically made at the façade of the homes, as we were interested in characterizing the exposure at home addresses. Therefore, we measured closer to the buildings, and not at the kerbside. An urban background site was defined as a site with fewer than 3000 vehicles per day passing within a 50 m radius. The distinction between regional and urban background was not strictly defined in the study manual, but typically involved measurements in the smaller towns of the cohort, as we assume that in smaller towns and villages no urban background is detectable.

Based on the ESCAPE guidelines, each local research center made a site selection proposal with a detailed characterization of the sites including Google maps of the study area and sites. The proposal was discussed by the ESCAPE exposure working group to harmonize the site selection across the centers. Each selected site was repeatedly geocoded using a GPS (e.g. at each start of a measurement). Because accuracy is very important – the spatial variability of air pollution concentrations occurs within tens of metres from major roads – the measured geocodes were plotted and checked on GIS maps with high resolution (e.g. road network, building ground map) and, if necessary, the geocodes were corrected to the original spot of measurement.

2.4. Sampling and analysis

In all 36 study areas, NO_2 and NO_x were measured with passive samplers. All local centres used the Ogawa diffusion badge (http://www.ogawausa.com) for those measurements. All badges were prepared and analysed by one central laboratory at the Institute for Risk Assessment Sciences (IRAS), Utrecht, using the ESCAPE SOP.
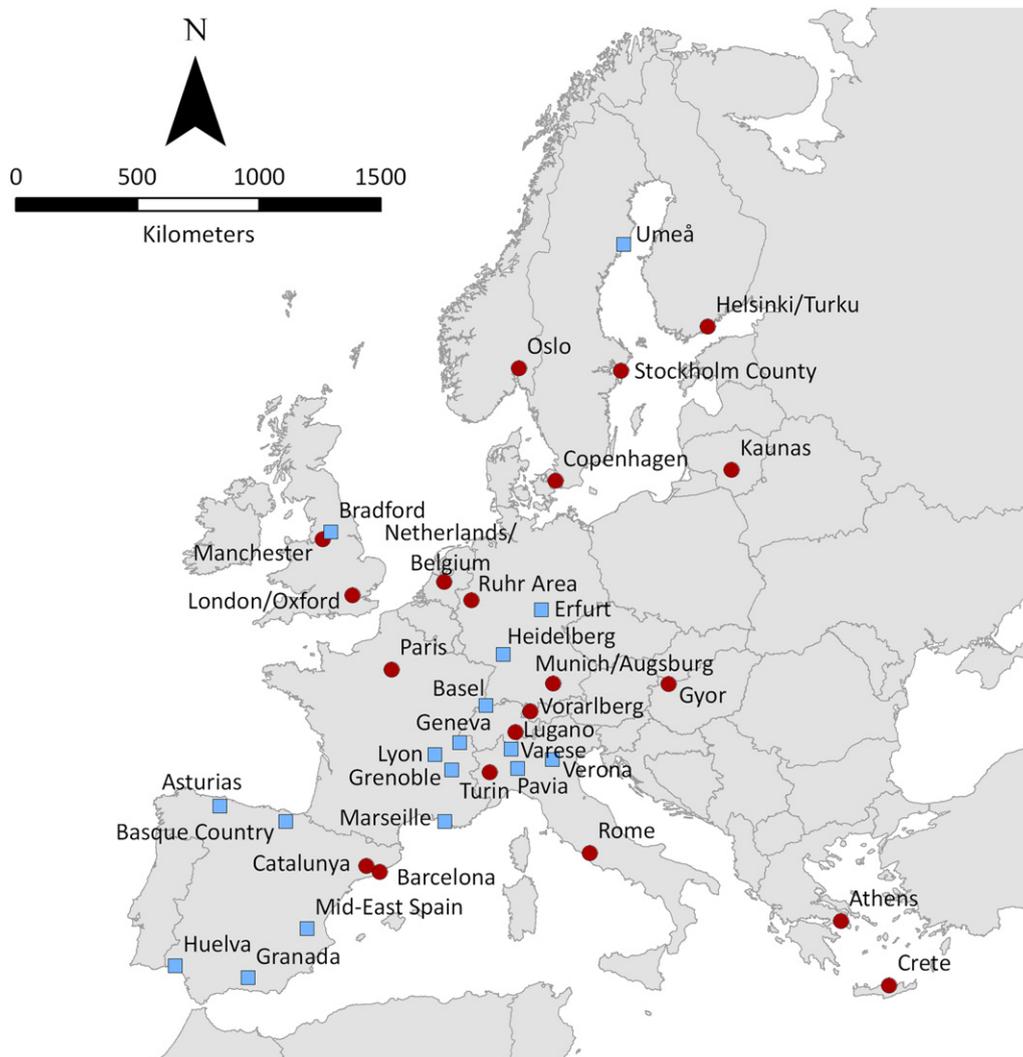
Fig. 1. ESCAPE study areas; dark circles mark the study areas where both PM and nitrogen oxides (NO_2 and NO_x) were measured. Blue squares indicate the study areas where NO_2 and NO_x only were measured.

A detailed description of sampling and analysis has been published previously (Roosbroeck van et al., 2006). In short, the sampler contains two collection filters that are coated with a reactive chemical, one for sampling NO_2 and one for sampling NO_x (NO_2 plus NO). NO is calculated by subtraction. Ogawa badges were transported from the central laboratory in individual plastic bags and cooled during transport and storage. The analysis is spectro-photometrically based upon the Saltzman method (Roosbroeck van et al., 2006). From each batch of 40 filters obtained from the manufacturer, four filters were kept at IRAS laboratory as lab blanks. These four lab blanks were analyzed on the same day as the exposed filters and their results were subtracted from results of filters in the same batch.

To establish the agreement with the chemiluminescence method being the European reference method, we compared the Ogawa diffusion badges during every 2-week sampling period with a chem-iluminescence monitor in most study areas. The Ogawa samplers were located in direct vicinity to the chemiluminescence monitor.

2.5. Sampling campaign

Because of limited equipment availability, especially for PM measurements, the study areas were divided into a first and second year group (Table 1). The sampling period for the first year group was between October 2008 and February 2010, and for the second year group it was between November 2009 and April 2011. At each monitoring site three measurement periods of two weeks were conducted; in the cold, warm and one intermediate temperature season. The two week sampling periods were performed in weeks with no unusual events such as bonfires and major holidays (i.e. school holidays of a week or longer).

In the NO_x only study areas the two week measurements were done almost simultaneously at all sites — the start day of all measurements was within a maximum time period of three days. In contrast, due to equipment limitations for PM sampling, the measurements in the NO_x + PM study areas were performed simultaneously at only 5 sites and one continuous reference site. Thus, for completion of one measurement period four rounds of two week measurements were necessary. Each group of 5 sites included different site types, e.g. regional, urban background and traffic. NO_2 and NO_x in the NO_x + PM study areas was measured exactly at the same time as the PM measurements were performed.

The reference site was used for sampling during all sampling periods, covering a full year. This reference site was located at an urban or rural background location and measured over the whole year. The site was used to adjust for temporal variation. If a PM and/

Table 1
Descriptive characteristics of all ESCAPE study areas.

Country	Study area	Study area description; major cities	PM + NO$_x$?	Year	Measurement period	# of sites	Distribution over site types: [b] RB/UB/ST
Norway	Oslo	Oslo city	PM + NO$_x$	1	05-02-2009–29-01-2010	40	4/18/18
Sweden	Umeå region	Vasterbotten county; Umeå, Skelleftea and Lycksele	NO$_x$	1	01-12-2008–11-07-2009	42	4/32/6
	Stockholm county	Stockholm county; Stockholm	PM + NO$_x$	1	03-12-2008–01-12-2009	40	5/20/15
Finland	Helsinki/Turku	Two areas: Helsinki/Vantaa and Turku/Loimaa	PM + NO$_x$	2	27-01-2010–26-01-2011	40	2/18/20
Denmark	Copenhagen	Copenhagen city and Hillerød	PM + NO$_x$	2	19-11-2009–17-11-2010	41	6/13/22
Lithuania	Kaunas	Kaunas city	PM + NO$_x$	2	20-01-2010–19-01-2011	40	5/13/22
United Kingdom	Bradford	Metropolitan borough of Bradford	NO$_x$	1	01-06-2009–15-12-2009	41	2/24/15
	Manchester	Greater Manchester urban area	PM + NO$_x$	1	27-01-2009–20-01-2010	39	0/15/24
	London/Oxford	Thames valley; London, Oxford and smaller towns	PM + NO$_x$	2	26-01-2010–18-01-2011	40	1/23/16
Netherlands/ Belgium	Netherlands/ Belgium	Entire country: Amsterdam, Rotterdam, Antwerp	PM + NO$_x$	1	17-02-2009–19-02-2010	80	20/24/36
Germany	Ruhr area	Dortmund, Duisburg, Essen and smaller towns	PM + NO$_x$	1	15-10-2008–12-10-2009	40	8/14/18
	Heidelberg	Heidelberg city and smaller surrounding towns	NO$_x$	1	07-04-2009–11-11-2009	43	3/16/24
	Erfurt	Erfurt city	NO$_x$	1	11-08-2009–16-12-2009	40	3/18/19
	Munich/Augsburg	Munich, Augsburg and smaller surrounding towns	PM + NO$_x$	1	27-10-2008–05-11-2009	40	10/12/18
Austria	Vorarlberg	Cities and areas along the main valley of Vorarlberg	PM + NO$_x$	2	03-03-2010–16-02-2011	40	5/11/24
France	Paris	Paris city and suburban areas	PM + NO$_x$	2	04-01-2010–04-01-2011	40	4/20/16
	Grenoble	Grenoble city and suburban areas	NO$_x$	2	20-01-2010–07-07-2010	40	3/17/20
	Lyon	Lyon city and suburban areas	NO$_x$	2	20-01-2010–07-07-2010	40	3/17/20
	Marseille	Marseille city	NO$_x$	2	14-01-2010–24-06-2010	39	2/17/20
Hungary	Gyor	Gyor city and neighbouring villages	PM + NO$_x$	2	22-02-2010–24-02-2011	40	1/19/20
Switzerland	Basel	Basel city and some surrounding smaller towns	NO$_x$	1	20-11-2008–10-06-2009	40	3/13/24
	Geneva	Geneva city and some surrounding smaller towns	NO$_x$	1	07-01-2009–03-07-2009	41	3/13/25
	Lugano	Lugano city and its neighbouring communities	PM + NO$_x$	1	02-03-2009–10-03-2010	42	4/16/22
Italy	Turin	Turin city and five smaller municipalities	PM + NO$_x$	2	01-02-2010–25-01-2011	40	1/13/26
	Pavia/Varese	Cities of Pavia, Varese and surrounding areas	NO$_x$	2	08-02-2010–14-06-2010	40	3/14/23
	Verona	City of Verona and surrounding areas	NO$_x$	2	20-01-2010–22-06-2010	40	3/14/23
	Rome	Rome city	PM + NO$_x$	2	27-01-2010–26-01-2011	40	2/19/19
Spain	Asturias	North part of Asturias and Oviedo region: Oviedo and Avilés	NO$_x$	1	17-02-2009–22-06-2009	40	2/13/25
	Basque country	Galdakao, Gipuzkoa and San Sebastián areas; many small towns	NO$_x$	1	03-02-2009–15-07-2009	39	2/12/25
	Barcelona	Barcelona city	PM + NO$_x$	1	14-01-2009–14-01-2010	40	1/13/26
	Catalunya	Three areas around Barcelona, Girona, Sabadell	PM + NO$_x$	1	14-01-2009–14-01-2010	80	5/23/52
	Mid East Spain	Valencia region and Albacete city	NO$_x$	1	17-02-2009–23-07-2009	38	2/13/23
	Granada	Granada city and smaller towns around Granada and Loja	NO$_x$	1	17-03-2009–15-09-2009	14	0/7/7
	Huelva	Hulvea city	NO$_x$	1	17-03-2009–15-09-2009	24	0/8/16
Greece	Athens[a]	Greater Athens area, 16 municipalities; Athens	PM + NO$_x$	2	21-04-2010–27-04-2011	40	1/22/17
	Heraklion[a]	Heraklion prefecture; Heraklion	PM + NO$_x$	1	18-02-2009–16-02-2010	40	0/21/19

[a] PM + NO$_x$ areas: dates refer to the period when the reference site was operated. NO$_x$ only areas: dates refer to the start of first and end of third measurement period.
[b] RB = regional background/UB = urban background/ST = street site.

or NO$_x$ measurement failed, the measurement was repeated, preferably in the same season.

2.6. Adjustment for temporal variation

Air pollutants have a substantial temporal variation. Therefore adjustment for temporal variability is essential if annual means of the measured NO$_2$ and NO$_x$ concentrations are calculated, particularly for the NO$_x$ + PM areas with non-simultaneous measurements. For details we refer to the PM companion paper (Eeftens et al., submitted for publication). Briefly, the difference between the concentration for a specific two week sampling period and the annual average at the reference site was subtracted from each measurement. This procedure is based upon procedures developed previously within the TRAPCA (Traffic-related Air Pollution and Childhood Asthma) study (Hoek et al., 2002, 2008). The annual average of NO$_2$ and NO$_x$ is calculated from the ESCAPE reference site for the NO$_x$ + PM areas. For the NO$_x$ only sites, an urban or rural background station routine monitoring data was used. We did not require a year-long reference campaign, because measurements had already been made simultaneously and only occasional missing data could lead to bias in the spatial comparison of calculated averages. In Umeå, where no appropriate routine monitoring site was available, adjustment was made using

the average of the three sampling campaigns of all sites with complete data. This approach only takes care of bias due to missing data and does not scale the data to an annual average.

In the following this paper focuses on adjusted annual average NO_2 and NO_x concentration. We report concentrations in $\mu g \, m^{-3}$. This has implications e.g. for reported NO_2/NO_x ratios. In case of equal NO_2 and NO concentrations in ppb, the NO_2/NO_x ratio is 0.50 expressed in ppb and 0.60 in $\mu g \, m^{-3}$. If the NO_2 is twice the NO concentration in ppb, the NO_2/NO_x ratio is 0.67 expressed in ppb and 0.75 in $\mu g \, m^{-3}$.

2.7. Quality control

Four field blanks and four duplicates were collected in each of the three measurement periods for the NO_x only study areas, to achieve a total of 12 field blanks and 12 duplicates ($4 \times 3 = 12$). For the $NO_x + PM$ areas in each sampling period one field blank and one duplicate were taken, which resulted in at least 12 field blanks and duplicates. Typically, more blanks and field duplicates were taken as these samples were taken in each sampling period of the year-round reference site. The plastic bags of the field blanks were opened at the measurement spot for a short time. Duplicates were installed at the same location. The limit of detection was calculated as three times the standard deviation of the blanks. Precision was calculated from field duplicates according to Eeftens et al. (submitted for publication).

2.8. Data analysis

Locally calculated adjusted annual averages were gathered centrally and their range and distribution were calculated, and stratified by site type. To quantify the amount of spatial variation relative to the background level, the interquartile range and total range (maximum−minimum) were calculated as a percentage of the mean. For each study area, we used ANOVA (SAS 9.2, PROC GLM) to test for significant differences between urban background and street sites and (where applicable) between urban and regional background sites. We also tested if urban background levels differed significantly between study areas. The same analyses were performed for street sites and regional background sites (where applicable), again using SAS 9.2, PROC GLM. Correlation R^2-values between components were determined using SAS 9.2, PROC REG. Percentages of between and within-area variance were determined using analysis of variance with PROC VARCOMP, METHOD = reml.

3. Results

3.1. Quality control

Detection limits were low for NO_2 for all study areas and very few samples were below the limit of detection (Table 2). Detection limits were slightly higher for NO_x but in nearly all study areas very few samples were below the limit of detection. Only in Turin, Catalunya and Albacete was a sizable fraction below the detection limit. This was partly due to unexplained outliers which were all included in the detection limit calculations presented in Table 2. Samples below the detection limit were retained at their original value.

Precision was expressed as the Coefficient of Variation (CV was lower than 10% in 31 areas for NO_2 and in 34 areas for NO_x (out of 36)). The average CV was 9.2% for NO_2 and 6.1 for NO_x, respectively.

In most study areas NO_2 concentrations measured by the Ogawa badge were lower than measured by chemiluminescence monitors (Fig. 2a). With the exception of Athens, average measurements agreed within 30% of the monitor. The ratio differed between study areas with no consistent pattern across Europe or concentration level. For NO_x the ratio was closer to unity for most areas (Fig. 2b). In study areas with a large number of co-located comparisons NO_2 measured by both methods were highly correlated (Online supplement B). Considering the difference in measurement principle, differences are acceptable.

3.2. Calculation of average NO_2 and NO_x concentrations

Unadjusted average concentrations and average concentrations adjusted for temporal variation were very highly correlated (Online supplement C, Table C1). Squared correlations (R^2) were generally above 0.95, indicating that temporal adjustment resulted only in small changes to the calculated average concentrations. For the NO_x only areas, this reflects that sampling was performed simultaneously and that there were few measurements that failed due to stolen badges or sampling and analysis errors. For sites with three valid observations, the adjustment involves the same scaling to an annual average. For the sites with less than three, a different scaling may occur, e.g. if the winter measurement had failed, the adjustment to the average is typically larger than for the sites with valid measurements in the three seasons. For the $NO_x + PM$ study areas, where measurements were not conducted simultaneously, measurements of two-week samples in three seasons were apparently sufficient to obtain a fairly stable estimate of spatial contrasts, in agreement with findings of the TRAPCA study using the same approach (Lewne et al., 2004).

3.3. Spatial variability within study areas

In many study areas the contrast of individual averages within the study area was as large as the contrast in median concentration across the study areas (Fig. 3a and b, Table 3.). Substantial spatial variability was found for NO_2 and NO_x concentrations within study areas. The average range for NO_2 (difference between the highest and the lowest annual average in one specific study area) was 54 $\mu g \, m^{-3}$. The range was on average 38.2 $\mu g \, m^{-3}$, 50.1 $\mu g \, m^{-3}$ and 67.1 $\mu g \, m^{-3}$ in the Northern-European, Central-Western and Southern European areas, respectively. The largest spatial variation for NO_2 was found in the largest European cities, such as London, Paris, Barcelona and Marseille. The results of variance component applied to all monitoring sites show that the variance of NO_2 within study areas (60.1% of the total variance) is considerably larger than the variance attributable to the differences between the study areas (39.9% of the total variance). The difference for NO_x is even more pronounced; within study area variability is 70% and the between study area variability is 30% of the total variance.

The type of the measurement sites (regional background, urban background and street sites) was a major determinant of the overall variability of NO_2 and NO_x. After including the variable "type of the measurement site" to the variance analysis, the results show that 30.4% of the overall NO_2 variance is attributable to the variability between the study areas, 37.4% of the variability is caused by different site type and 32.3% could be traced to the variability within the site types. The corresponding values for NO_x are 23.4% (variability between the study areas), 36.5% (variability because of different site type) and 40.1% (variability within the site types).

The distribution of annual averages of NO_2 and NO_x concentrations by site type, for each study area is shown in Figs. 4 and 5, respectively. The NO_2 and NO_x concentrations at urban background sites are in general lower than at street locations. Table 4

Table 2
Detection limits and precision for NO_2 and NO_x measurements.

Study area	Field blanks								Field duplicates		
	Number field blanks	Average field blank ($\mu g\ m^{-3}$)		Detection limit ($\mu g\ m^{-3}$)		N samples below the detection limit (total number of valid samples)			Number duplicates	CV (%)	
	N	NO_2	NO_x	NO_2	NO_x	NO_2	NO_x		N	NO_2	NO_x
Oslo	18	0.0	0.4	2.5	2.8	1 (123)	0 (123)		28	23.1	11.5
Umeå Region	6	0.1	0.7	0.6	1.2	0 (124)	7 (124)		6	4.5	4.8
Stockholm County	22	0.1	0.5	1.1	2.4	2 (143)	0 (143)		22	9.5	7.5
Helsinki/Turku	23	0.1	0.2	0.6	1.7	0 (154)	0 (154)		24	4.9	6.2[b]
Copenhagen	20	0.3	−0.2	1.2	3.1	0 (143)	0 (143)		20	5.6	7.6
Kaunas[a]	24	0.1	0.8	0.6[a]	5.7[a]	0 (146)	3 (146)		24	8.5	10.4[b]
Bradford	5	0.5	0.9	1.1	2.4	0 (112)	0 (112)		6	12.0	1.9
Manchester	12	0.5	1.5	1.1	5.3	1 (116)	0 (116)		12	4.9	4.4
London/Oxford	23	0.3	1.3	1.3	5.5	0 (131)	0 (131)		23	5.4	5.7
Netherlands/Belgium	20	0.2	0.5	1.3	3.6	0 (263)	0 (263)		20	7.1	4.7
Ruhr area	15	0.2	0.4	0.8	2.5	0 (120)	0 (120)		23	4.5	4.5
Heidelberg	8	0.3	1.1	0.7	2.3	0 (127)	0 (127)		10	9.7	3.2
Erfurt	12	0.2	0.0	0.8	2.6	0 (118)	0 (118)		12	8.8	3.8
Munich	15	0.3	0.9	1.0[a]	4.6[a]	0 (142)	0 (142)		16	3.0	2.9
Vorarlberg	25	0.6	2.4	1.2	6.7	1 (144)	1 (144)		25	9.3	6.6
Paris	22	0.3	2.1	1.2	6.6	0 (141)	0 (141)		22	8.9	7.1
Grenoble	12	0.1	0.5	0.8	3.1	0 (120)	0 (120)		12	19.5	7.5
Lyon	12	0.2	0.9	0.5	3.6	0 (117)	0 (117)		12	7.9	6.3
Marseille	12	0.1	0.1	0.6	1.6	0 (114)	0 (114)		17	4.5	4.3
Gyor	25	0.3	0.6	1.3	2.5	0 (145)	0 (145)		24	10.7	13.2
Basel	8	0.4	0.8	0.7	5.0	0 (120)	0 (120)		8	4.9	5.6
Geneva	10	0.1	1.1	1.0	8.3	0 (121)	0 (121)		8	5.6	4.1
Lugano	16	0.5	1.5	1.2[a]	9.5[a]	0 (137)	4 (137)		23	7.7	5.1
Turin	24	0.3	1.7	1.4[a]	10.0[a]	0 (144)	36 (144)		24	9.6	10.0[b]
Pavia/Varese	12	0.8	1.2	2.2	3.2	0 (120)	0 (120)		12	9.3	5.2
Verona	11	0.3	0.3	0.5	2.2	0 (120)	0 (120)		13	7.2	2.5
Rome	24	0.2	0.4	0.5	2.0	0 (144)	0 (144)		25	9.0	4.3
Asturias:[c]											
Asturias region	12	0.0	0.0	0.9	1.9	0 (45)	0 (45)		12	5.1	4.5
Oviedo	12	0.4	3.8	1.6	17.2	0 (68)	8 (68)		12	5.6	2.3
Basque Country:[c]											
Bilbao	12	0.2	0.2	1.0	3.0	0 (40)	0 (40)		12	6.4	3.3
San Sebastian	11	0.0	0.4	0.8	1.3	0 (71)	0 (71)		9	4.6	2.9
Barcelona	23	0.5	3.7	3.3[a]	28.1[a]	1 (142)	6 (142)		27	7.7	7.8
Catalunya	23	0.5	3.7	3.3[a]	28.1[a]	1 (309)	39 (309)		27	7.7	7.8
Mid-East Spain:[c]											
Albacete	12	0.4	3.1	1.9	9.4	1 (51)	12 (51)		10	9.9	14.2
Valencia	10	0.1	0.1	0.4	1.0	1 (51)	1 (51)		11	11.9[b]	3.9
Granada	3	0.5	0.7	1.1	1.5	0 (29)	1 (29)		3	35.6	5.5
Huelva	2	−0.2	0.0	0.3	2.7	0 (67)	0 (67)		3	19.5	13.4
Athens	23	0.1	0.3	0.8	1.3	0 (143)	1 (143)		23	10.9	5.6
Heraklion	21	0.5	0.7	2.7	3.5	2 (140)	1 (140)		13	7.8	5.6

Detection limit calculated as three times the standard deviation of field blanks. CV = Coefficient of Variation calculated from field duplicates.

[a] Detection limit (DL) affected by one blank for which no explanation was found. Without this one blank, NO_x DL and number of samples below DL become 2.0 $\mu g\ m^{-3}$ and 0 (Munich): 6.7 $\mu g\ m^{-3}$ and 27 (Turin); 2.7 $\mu g\ m^{-3}$ and 0 (Kaunas); 7.2 $\mu g\ m^{-3}$ and 0 (Barcelona); 7.2 $\mu g\ m^{-3}$ and 0 (Catalunya); 2.8 $\mu g\ m^{-3}$ and 0 (Lugano).

[b] CV affected by one poor duplicate, for which no explanation was found. Without this duplicate CV becomes 3.8% for NO_x (Helsinki); 5.4% for NO_x (Kaunas); 3.1% for NO_x (Turin); 4.7% for NO_2 (Valencia).

[c] In three subareas different teams performed fieldwork, field duplicates/blanks treated analysed separately.
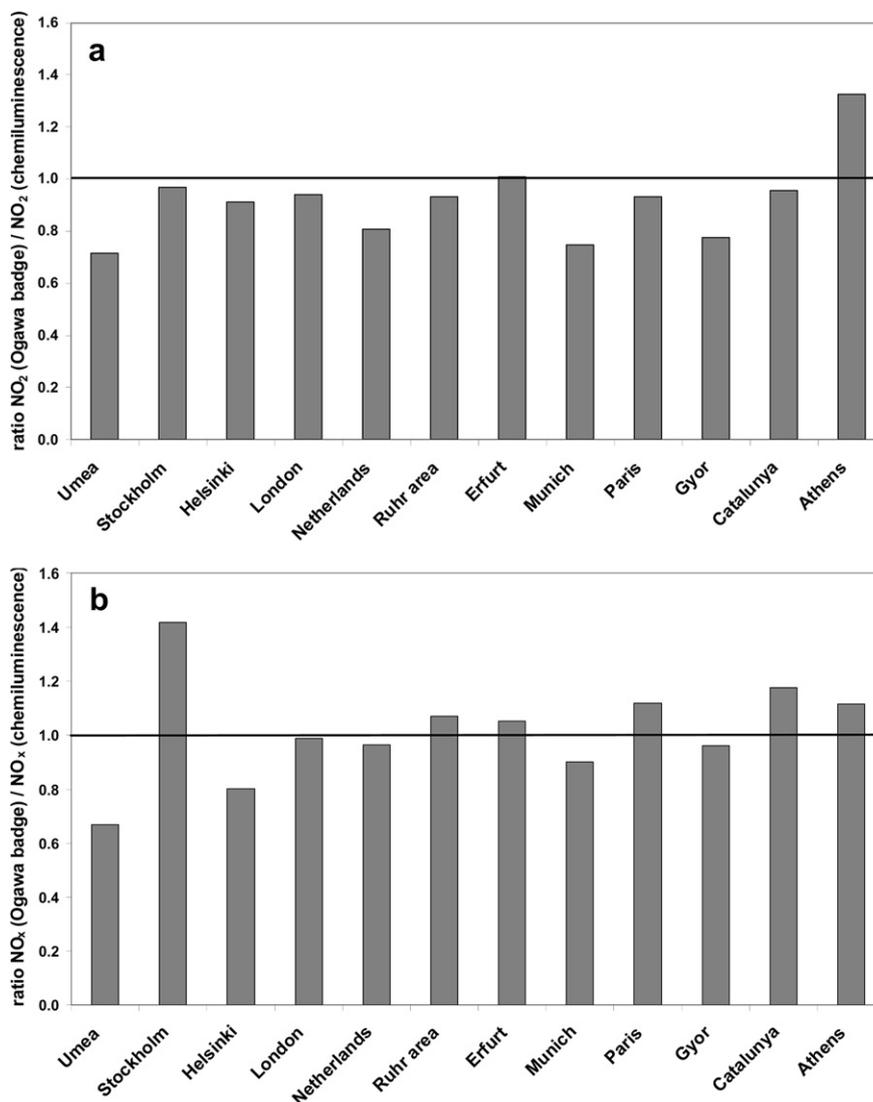
presents the ratio of concentrations measured at regional versus urban background and the ratio of street to urban background sites for NO_2 and NO_x, respectively. In almost all study areas the differences between the site types were statistically significant ($p < 0.05$ level).

In all study areas, the concentrations at street stations were higher than at urban background stations; the street site vs. urban background site ratio was 1.63 for NO_2 and 1.93 for NO_x on average for all sites over all study areas. The ST/UB ratio ranged substantially from 1.09 to 3.16 for NO_2. NO_2 concentrations were lower at regional background sites compared to urban background concentrations; the mean ratio for all sites over all study areas was 0.63 for NO_2 and 0.60 for NO_x, respectively. However, the RB/UB concentration ratio varied substantially across study areas, from 0.24 in London to 0.91 in Gyor (Table 4).

3.4. Spatial variability across Europe

Because the overall mean concentration may be affected by differences in the fraction of different site types, the direct comparison of the results between the study areas (across Europe) is based mainly on urban background locations.

Substantial differences in annual average NO_2 and NO_x concentration were found across Europe (Table 3). The lowest annual average NO_2 concentration was found in Umeå, a medium sized city in Northern Sweden. Concentrations in the large North-European cities (Helsinki, Stockholm, Oslo, and Copenhagen) were relatively low, but similar to the smaller cities in southern (Heraklion, Crete) and central Europe (Erfurt, Gyor, Kaunas). The highest concentrations were measured in the Mediterranean area, especially Barcelona and Turin.

Fig. 2. a: Ratio of concentrations measured by Ogawa badge and chemiluminescence monitor co-located at routine monitoring sites for NO_2. b: Ratio of concentrations measured by Ogawa badge and chemiluminescence monitor co-located at routine monitoring sites for NO_x.

3.5. NO_2–NO_x relationship

In all study areas a high correlation between NO_2 and NO_x was found (Table 5). The average NO_2 to NO_x ratio ranged from 0.51 to 0.72 for urban background sites in the different study areas (Table 5). At street stations the NO_2 to NO_x ratio ranged from 0.42 to 0.73. Overall there was no clear North-South gradient in the ratio. Mean ratios at urban background sites were 0.62, 0.63 and 0.61 for the Northern-European, central-western and southern European areas, respectively.

In most areas, the NO_2 to NO_x ratio was smaller at street sites than at urban background sites. On average, the ratio was 0.69, 0.62 and 0.54 for regional background, urban background, and street sites respectively. Without Mid-East Spain the ratio was 0.65 for regional background. The difference in ratio varied significantly across Europe, e.g. in Copenhagen, Paris, and the Netherlands the ratio was much lower at street sites than at urban background sites, whereas in e.g. Basel, Geneva there was little difference, possibly reflecting differences in primary NO_2 emissions across Europe.

4. Discussion

Overall, we found significant contrasts in annual average NO_2 and NO_x concentrations between and especially within 36 study areas across Europe. NO_2 concentrations at street locations were on average between 1.22 and 3.6 times higher than at urban background stations. The NO_2/NO_x ratio varied between 0.47 and 0.72 across study areas. Concentrations were generally lower in Northern than in Southern Europe.

4.1. Variability within study areas

The observation of high within study area variability of NO_2 concentrations compared to between study area variability agrees with previous observations in urban areas of four cities in western and central Europe (Lebret et al., 2000), 16 cities of the European Community Respiratory Health Survey (ECRHS) (Götschi et al., 2008) and 8 areas of the longitudinal Swiss cohort study on air pollution and lung and heart disease in adults in Switzerland (SAPALDIA) (Liu et al., 2012).
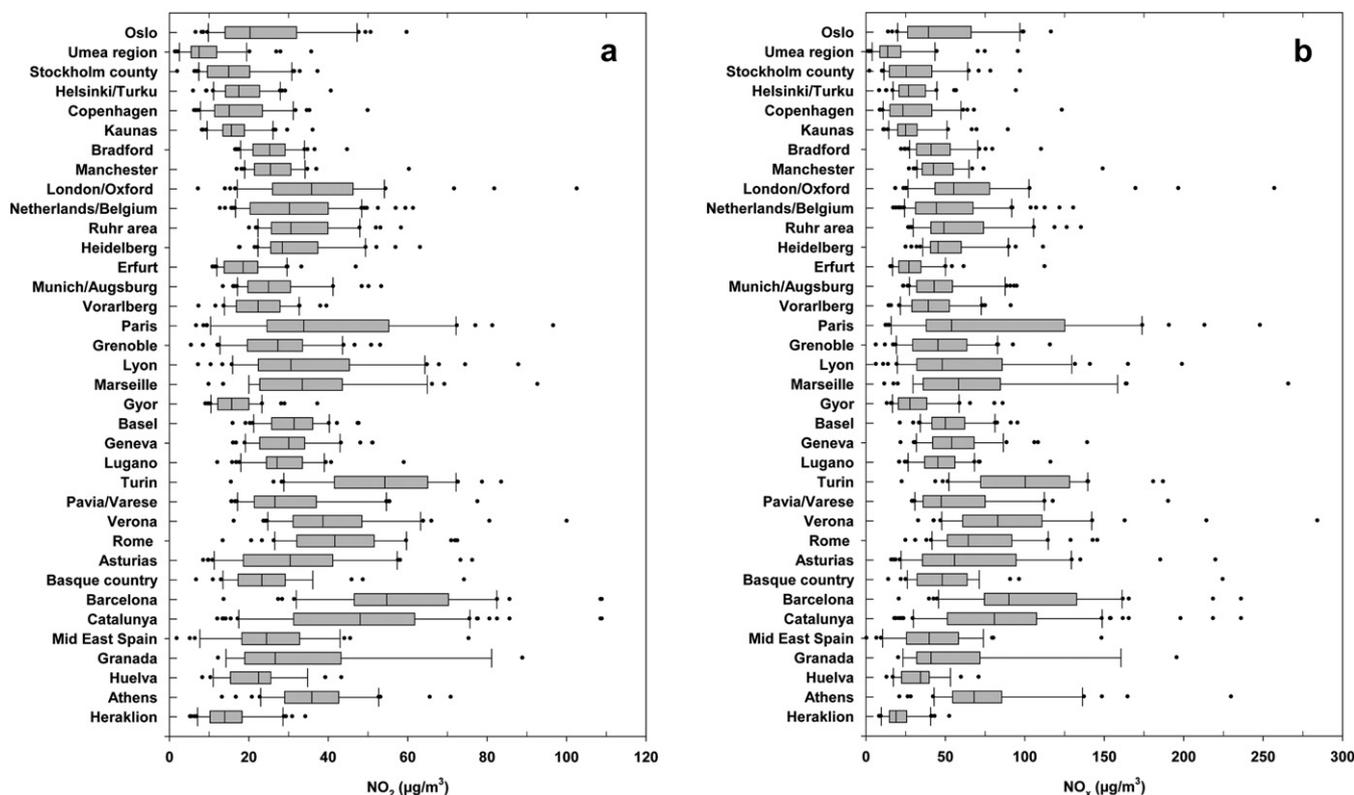
Fig. 3. a: Distribution of annual average concentration of NO$_2$ for each study area separately. Median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points. b: Distribution of annual average concentration of NO$_x$ for each study area separately. Median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points.

This raises one of the central questions of air pollution epidemiology. In epidemiological studies evaluating the health effects of long-term exposure to air pollutants, the usual place of residence explains much of the exposure contrasts between persons. The vast majority of long-term epidemiological studies compare the health status of populations living in different cities (Pope and Dockery, 2006; Götschi et al., 2008). These studies generally assigned one overall average concentration to all subjects living in each city and use the contrast in city-average air pollution levels between different areas. This approach assumes that a limited number of monitors per area (or even only one monitor per city) could provide an unbiased estimate of the average community exposure to background pollution. The observation of large within study area contrasts which exceed between study areas contrasts raises considerable doubts about this type of analysis. For air pollutants showing pronounced spatial variability and affected by local sources (such as traffic), assessing exposure at the community level could lead to substantial misclassification and, together with other factors, may introduce considerable random errors in the estimation of the true individual exposure or even contribute to the observed null-findings (Götschi et al., 2008). The large spatial variation of the concentration levels of traffic related air pollutants across cities suggests that it is virtually impossible to characterize the city-average concentration with one monitoring site. Because of this, modelling of concentrations for traffic related air pollutants could be a reasonable option for exposure assessment. For the ESCAPE study we will develop study-area specific LUR models.

Our analysis of variance showed that about 37% of the overall variability was due to site type, 32% to remaining within-area variability and 30% to between-area variability. Differences between street and urban background sites are discussed in Section 4.2. Further variability within study areas was especially explained

by population density (or household density), and type of land use (natural land, industrial land). In a few areas altitude and presence of a port nearby also contributed to the explanation of spatial variation (Beelen et al. submitted for publication).

At 323 sites (out of 1485) in 31 study areas (out of 36), the annual average NO$_2$ concentration measured at a site exceeded 40 μg m^{-3}, the EU limit value for the annual average of NO$_2$ (in Catalunya at 50 sites, Barcelona at 33 sites, and Turin at 31 sites). The comparison with the air quality standard for annual average is limited because we sampled only during three 2-week periods. However, a reference site was used to adjust all measured concentrations to an annual average concentration.

4.2. ST/UB ratio

The lower ST/UB ratio observed for for NO$_2$ compared to NO$_x$ is consistent with the relatively small fraction of NO$_2$ to NO$_x$ in motor vehicle exhaust emissions and the conversion of primary emitted NO to NO$_2$ by ozone (Keuken et al., 2009). The latter reaction leads to increasing NO$_2$/NO ratios with increasing distance to major roads.

The ST/UB ration observed in our study for NO$_2$ and NO$_x$ concentrations was higher and more consistent than for the particle metrics across all study areas (Eeftens et al., submitted for publication). For PM$_{2.5}$, PM$_{2.5}$ absorbance (soot), PM$_{10}$ and PM$_{coarse}$ we found average ratios of 1.14, 1.38, 1.23 and 1.42 respectively. This was not due to differences in the included study areas, as the NO$_2$ and NO$_x$ ST/UB ratio in the areas where PM was measured were 1.54 and 1.80 respectively. The higher ST/UB ratios for NO$_2$ compared to PM$_{10}$ and PM$_{2.5}$ are consistent with previous studies (HEI, 2010). PM$_{10}$ and especially PM$_{2.5}$ have a high regional background concentration and local sources increase concentrations only modestly. The slightly higher ratio compared to absorbance is more

Table 3
Distribution of annual average concentrations of NO_2 and NO_x by study area (note that concentration distribution are presented here for the all sites; differences between different areas might be affected also by different numbers of regional background, urban background and street sites selected for the different study areas).

Study area	NO_2					NO_x			
	n	Median	25%–75% percentile	Min–max	Range/ mean (%)	Median	25%–75% percentile	Min–max	Range/ mean (%)
Oslo	40	20.3	14.1–31.8	6.7–59.8	220%	39.4	26.5–65.7	14.2–116.8	215%
Umeå Region	42	7.5	5.5–11.8	1.5–35.8	349%	13.7	8.8–21.3	1.7–95.9	468%
Stockholm County	40	14.9	9.6–19.7	2.1–37.4	220%	25.3	14.9–40.9	2.5–97.2	311%
Helsinki/Turku	40	17.5	14.1–22.7	6.1–40.8	184%	26.8	20.8–37.4	8.6–94.7	281%
Copenhagen	41	15.1	11.8–23.3	6.3–50.1	245%	23.3	15.4–39.3	9.0–123.6	378%
Kaunas	40	15.6	13.6–18.8	8.3–36.2	167%	25.0	20.2–31.6	11.3–89.6	272%
Bradford	41	25.3	21.1–29.1	16.7–44.9	109%	41.0	32.3–53.0	22.4–110.5	195%
Manchester	39	25.4	21.4–30.6	17.0–60.4	164%	42.5	35.4–54.9	27.4–149.4	256%
London/Oxford	40	35.8	26.6–46.2	7.3–102.7	251%	55.3	43.8–77.5	18.8–257.4	344%
Netherlands/Belgium	80	30.2	20.5–39.9	12.8–61.5	157%	44.3	31.3–67.3	17.5–130.8	219%
Ruhr area	40	30.6	25.9–39.4	20.2–58.4	115%	49.1	41.1–73.8	26.9–135.7	181%
Heidelberg	43	28.4	25.5–37.4	17.7–63.2	141%	45.5	40.6–59.9	25.3–111.8	164%
Erfurt	40	18.6	14.0–22.2	11.0–47.0	187%	27.1	21.0–33.8	15.6–112.8	314%
Munich/Augsburg	40	24.9	19.8–30.3	13.6–53.5	149%	42.9	32.6–54.4	23.7–95.2	152%
Vorarlberg	40	22.3	17.2–27.5	7.4–39.7	143%	39.2	30.5–52.4	14.6–91.4	180%
Paris	40	33.8	24.8–54.8	6.8–96.8	226%	53.9	39.6–121.1	12.7–248.3	293%
Grenoble	40	27.3	20.4–33.2	5.5–53.2	175%	45.3	30.2–63.1	6.5–116.2	227%
Lyon	40	30.6	22.5–45.2	7.3–88.0	231%	48.1	32.4–85.0	6.5–199.2	312%
Marseille	39	33.4	22.8–43.5	10.0–92.8	229%	58.3	36.0–84.6	11.9–266.1	363%
Gyor	40	15.7	12.2–19.9	9.2–37.4	169%	27.6	20.5–38.1	13.4–86.3	224%
Basel	40	31.4	25.7–36.0	16.0–47.8	102%	50.0	41.7–61.4	21.6–95.7	140%
Geneva	41	30.1	22.8–33.3	16.1–51.3	118%	54.0	42.1–67.6	22.1–139.6	203%
Lugano	42	27.1	24.5–33.4	12.2–59.1	164%	45.2	37.4–55.6	21.2–116.4	199%
Turin	40	54.3	42–64.8	15.6–83.7	128%	100.3	73.5–127.2	22.8–187.3	162%
Pavia/Varese	40	26.6	21.5–36.3	15.7–77.7	198%	47.4	36.9–74.7	29.3–190.5	265%
Verona	40	38.7	31.1–48.1	16.3–100.1	202%	83.0	61.0–110.6	33.1–284.4	274%
Rome	40	41.7	32.3–51.4	13.6–72.6	138%	64.4	51.2–91.4	25.1–146.0	166%
Asturias	40	30.4	19.0–40.7	8.6–76.4	212%	55.8	37.0–91.6	16.2–220.3	293%
Basque Country	39	23.3	17.3–29.2	6.8–74.3	264%	48.1	32.2–63.7	14.3–224.9	399%
Barcelona	40	54.7	46.6–70.1	13.8–109.0	165%	90.0	74.8–132.4	21.0–236.4	213%
Catalunya	80	48.0	31.4–61.5	12.2–109.0	202%	80.9	51.6–107.3	18.2–236.4	255%
Mid-East Spain	38	24.4	18.8–31.9	1.9–75.5	282%	39.8	25.6–58.0	0.6–148.6	346%
Granada	14	26.6	19.7–43.1	12.4–89.0	220%	40.9	32.7–70.8	20.6–195.9	294%
Huelva	24	22.4	15.6–25.5	8.4–43.4	160%	34.4	22.4–39.6	13.3–71.3	172%
Athens	40	35.9	29.2–42.6	13.3–71.0	158%	68.1	54.9–85.3	21.4–230.1	268%
Heraklion	40	13.9	10.3–18.2	5.3–34.3	190%	19.0	14.9–25.3	8.6–52.8	206%

striking, as absorbance is strongly affected by local sources including traffic (Janssen et al., 2011). The higher contrast for NO_2 compared to soot could be related to the differences of atmospheric lifetime for NO_x and soot. Soot is enriched in submicrometer particle fraction, thus it have a longer atmospheric lifetime allowing for accumulation at background sites. This interpretation is supported by the observation that the urban background − regional background ratio was also larger and more consistent for NO_2 than for absorbance. Higher emissions of NO_x from road traffic compared to the background could also play a role. The increasing use of after treatment technology that traps soot from diesel vehicles but increases NO_2 because of catalytic oxidation of collected soot may have contributed to this (Grice et al., 2009; Williams and Carslaw, 2011).

The ratio of the ST/UB concentrations varied widely across the study areas. The implication of this variability for epidemiological studies is that the use of traffic indicator variables such as living close to a major road presents a different contrast in actual air pollution exposure in different cities. This likely contributes to the observed heterogeneity of estimated health effects in studies using traffic indicators (Vardoulakis et al., 2003; Heinrich and Wichmann, 2004).

The relatively high ST/UB ratios in Northern European cities, particularly in Umeå, are likely due to the low urban background concentrations in these countries. The variability in ratios is probably explained by a combination of differences in the following factors: traffic intensity, fraction of heavy duty vehicles, emission factor of the car fleet related to e.g. age and fuel type of cars and possibly street configuration. In a recent study in the Netherlands,

higher ST/UB ratios were found for NO_2 and other pollutants in 2 street canyons and 2 streets with homes on one side of the street compared to wider streets (Boogaard et al., 2011). This was explained by poorer dispersion in the narrower streets. We did not find higher ratios in the typically narrower streets in Southern Europe. However, this was partly due to the higher urban background as the average absolute concentration difference between ST and UB was highest in Southern Europe (17.2 μg m^{-3}). Further differences in dispersion may be due to differences in wind speed and the potential for occurrence of inversions.

In interpreting the (often modestly increased) magnitude of the ratio of concentrations measured at street sites and urban background sites, it has to be taken into account that measurements were not made at kerbsides, as is often the case in routine monitoring network street sites. The average ratio further reflects an average street, not the busiest street in the area. Figs. 4 and 5 illustrate that there is significant variability across street sites in one area.

Pronounced contrast between street and background environments was observed also for other traffic related pollutants, including ultrafine particles (Cyrys et al., 2003a; Tuch et al., 2006; Puustinen et al., 2007; Cyrys et al., 2008; Hoek et al., 2011). As shown in the companion paper for $PM_{2.5}$, PM_{10} and $PM_{2.5}$ absorbance (Eeftens et al., submitted for publication) the R^2 between NO_2 and $PM_{2.5}$ absorbance (as indicator for traffic related soot) is generally high: 0.80 (range 0.55–0.91), while the correlation between NO_2 and $PM_{2.5}$ is much lower: $R^2 = 0.50$ (range 0.02–0.90). Also, the temporal variation of NO and NO_2 concentrations
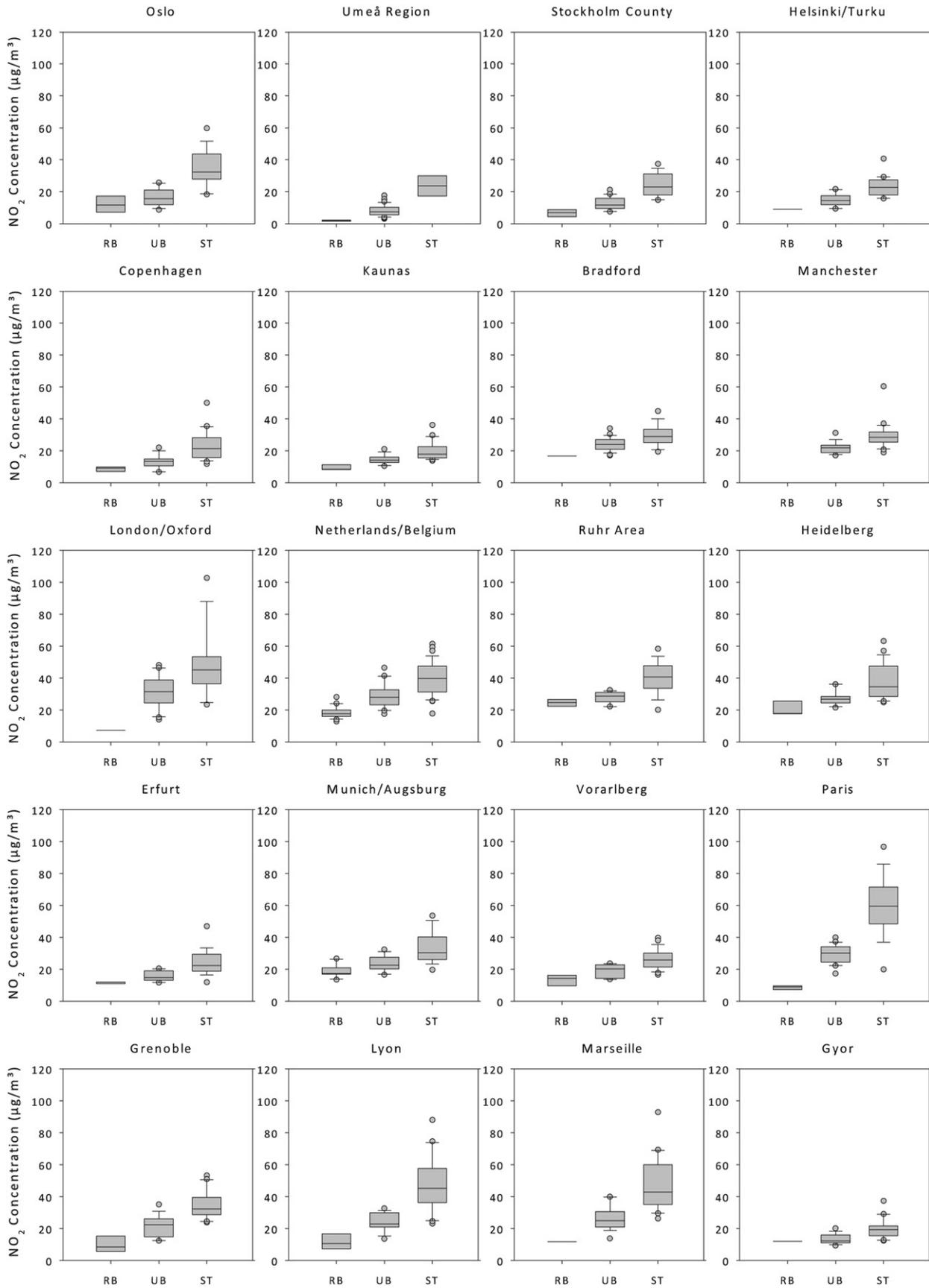
Fig. 4. Annual averages of NO$_2$ concentrations by site type, for each study area (for measurement period please refer to Table 1). Median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points. RB = regional background, UB = urban background and ST = street locations.
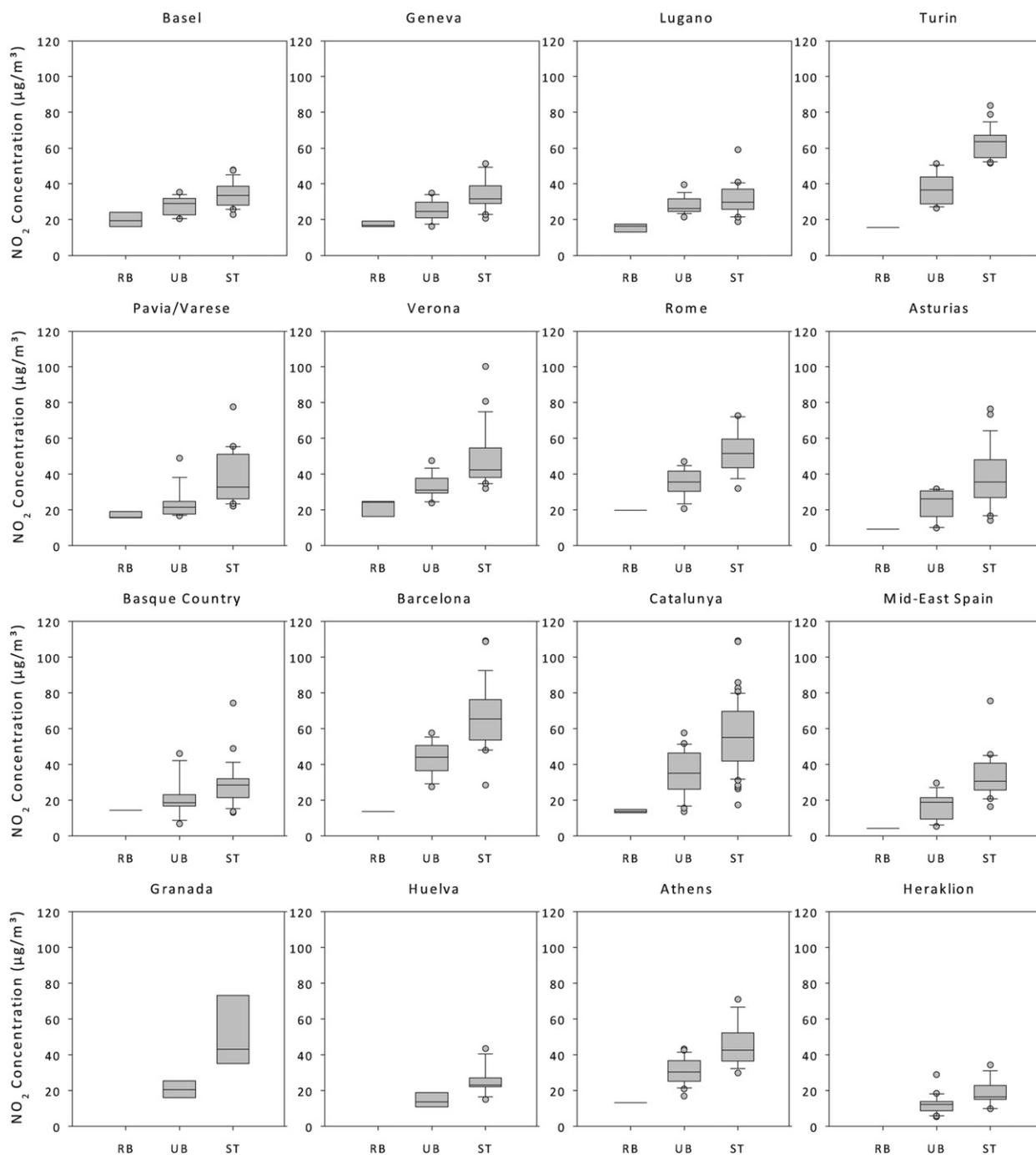
Fig. 4. (continued).

is often strongly correlated with those of other traffic related air pollutants, such as CO, Black Carbon and ultrafine particles (Cyrys et al., 2003b; Hagler et al., 2009; Sabaliauskas et al., in press). Moreover, nitrogen oxides have been found to be the most important predictor variables for ultrafine particles in the urban air (Paatero et al., 2005). It reveals the role of nitrogen oxides as marker for traffic related air pollutants. Due to the close temporal correlation with other combustion related pollutants, NO_2 (in some cases also NO) has been often used in epidemiological studies as a marker for traffic exhaust (Krzyzanowski et al., 2005). Its concentration is measured easily and relatively cheaply, but one should keep in mind that it serves only as a surrogate for a set of sources and resulting mixture of air pollutants.

4.3. Contrasts across Europe

The general pattern of low concentrations across Northern Europe agrees with previous studies based upon study specific monitoring programmes (Hazenkamp-von Arxa et al., 2004) or upon routine monitoring stations (Beelen et al., 2009). The high NO_2 concentrations in Southern European countries could be due to high traffic intensity, a large fraction of diesel-powered vehicles and a higher conversion of NO to NO_2 because of relatively high temperatures and ozone concentrations. Alternatively, the more densely built Mediterranean cities could lead to higher concentrations. Analyses of the high concentrations in Turin and Athens have been reported in previous studies
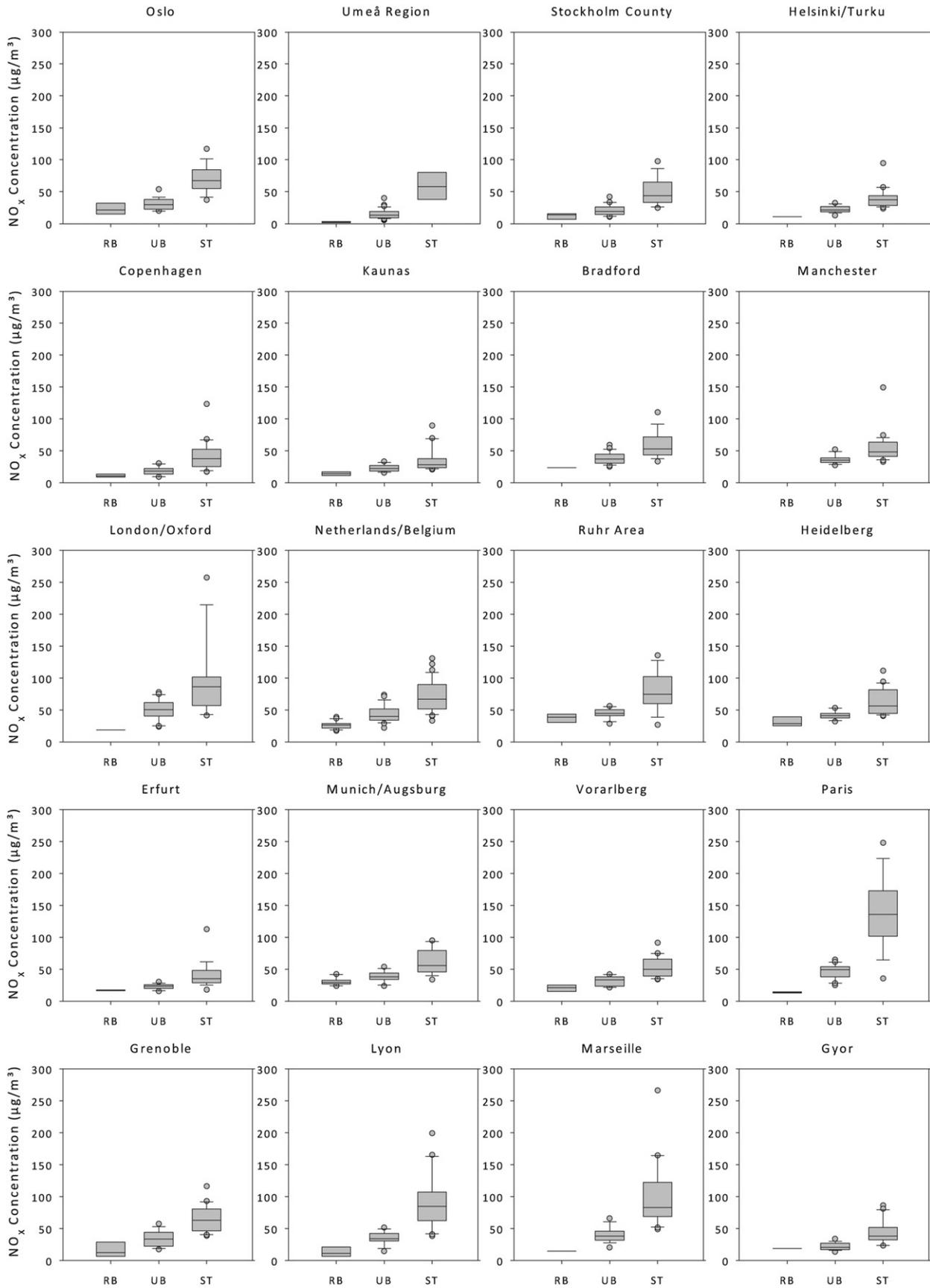
Fig. 5. Annual averages of NO$_x$ concentrations by site type, for each study area (for measurement period please refer to Table 1). Median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points. RB = regional background, UB = urban background and ST = street locations.
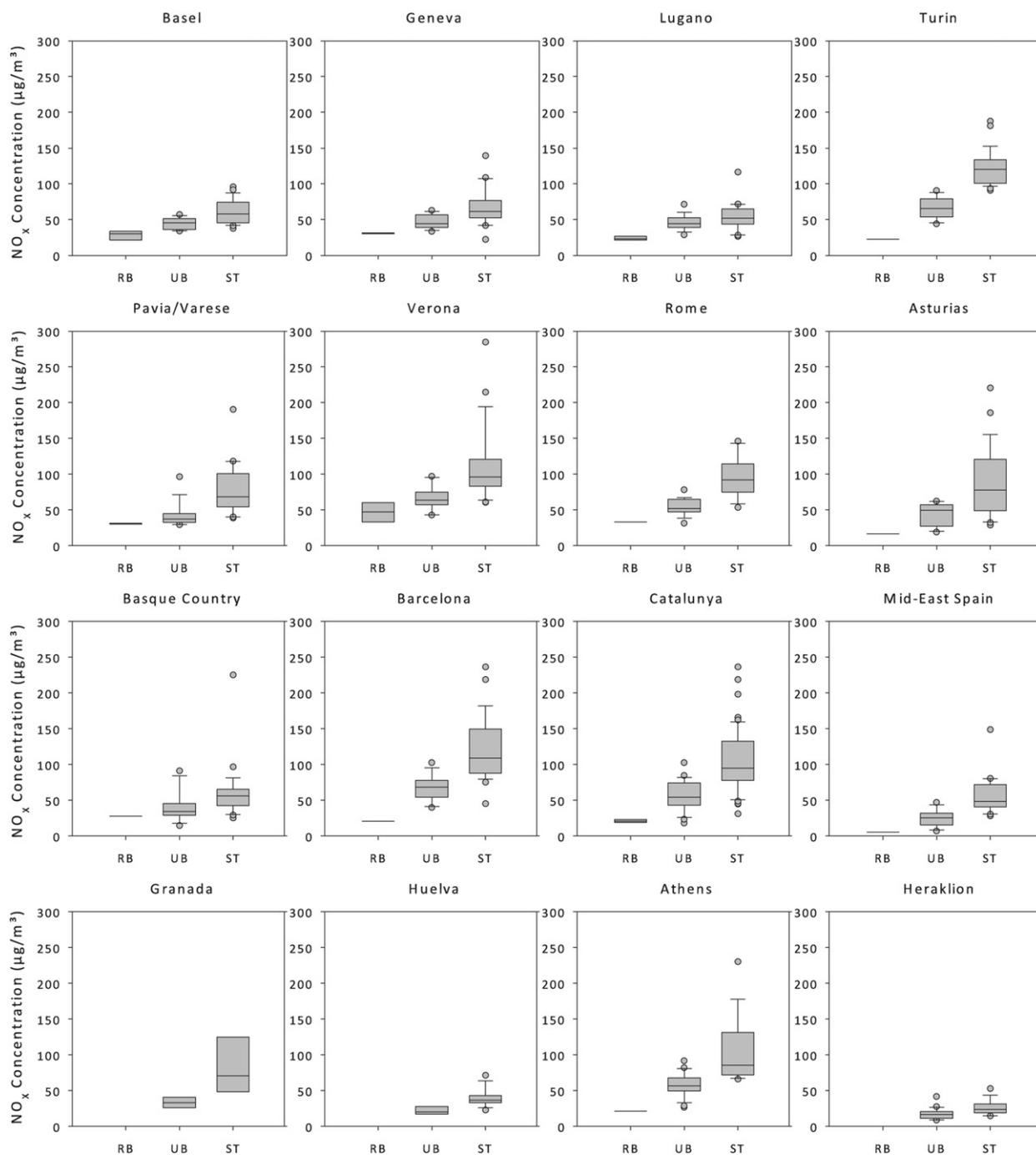
Fig. 5. (continued).

(Hazenkamp-von Arxa et al., 2004; Minguzzi et al., 2005; Lonati et al., 2010; Fattore et al., 2011). In Turin, the combination of stagnant air conditions with high emissions due to heavy traffic and high population density is the cause of very strong pollution episodes. Turin is located mainly on the left bank of the Po River and is surrounded by the Alpine arch. It favours thermal inversions, characterized by low surface wind speeds (stagnation of the air) and trapping air pollutants in the lower layers of the atmosphere. In Athens, high emission rates and the location in a valley surrounded by high mountains contribute to high pollution levels.

Some of the variability across the study areas may be due to weather variability between the two years. Overall, there was no difference in average concentration measured at routine monitoring stations between 2009 and 2010 (downloaded from AIRBASE, the European air pollution database). Mean NO_2 concentration of all sites with more than 75% data capture in both years was 26.9 and 26.6 μg m^{-3} in 2009 and 2010. When assessed by country, the NO_2 concentration was about 3 μg m^{-3} higher in 2010 in the three Northern European countries, with small differences for the other countries. The overall patterns measured in ESCAPE are not affected by these modest differences. With the Ogawa badge, lower concentrations were measured than with the chemiluminescence monitors, the reference method for NO_2. The modest variability of the ratio between study areas could be due to variation in sampling

Table 4
Ratios between regional background and urban background concentrations, and between street and urban background concentrations, for all study areas.

Study area	NO$_2$		NO$_x$	
	Ratio regional/urban background	Ratio street/urban background	Ratio regional/urban background	Ratio street/urban background
Oslo	0.71*	2.09**	0.73*	2.28**
Umeå Region	0.26**	3.16**	0.18**	4.24**
Stockholm County	0.49**	1.86**	0.50**	2.35**
Helsinki/Turku	0.59**	1.55**	0.48**	1.71**
Copenhagen	0.69**	1.72**	0.64**	2.14**
Kaunas	0.66**	1.33**	0.62**	1.43**
Bradford	0.71**	1.22**	0.63**	1.52**
Manchester	–	1.32**	–	1.42**
London/Oxford	0.24**	1.50**	0.39**	1.81**
Netherlands/ Belgium	0.64**	1.36**	0.62**	1.63**
Ruhr area	0.90	1.41**	0.88	1.70**
Heidelberg	0.75*	1.34**	0.74*	1.44**
Erfurt	0.73*	1.48**	0.75	1.65**
Munich/Augsburg	0.79**	1.38**	0.81*	1.56**
Vorarlberg	0.68**	1.38**	0.65**	1.63**
Paris	0.30**	1.98**	0.30**	2.77**
Grenoble	0.44**	1.66**	0.41**	1.94**
Lyon	0.47**	1.93**	0.34**	2.51**
Marseille	0.46**	1.78**	0.38**	2.39**
Gyor	0.91	1.46**	0.89	1.87**
Basel	0.72**	1.24**	0.64**	1.35**
Geneva	0.71**	1.35**	0.67*	1.36**
Lugano	0.56**	1.09	0.53**	1.14
Turin	0.43**	1.71**	0.35**	1.85**
Pavia/Varese	0.75	1.59**	0.78	1.79**
Verona	0.65**	1.44**	0.70	1.63**
Rome	0.55**	1.48**	0.60**	1.71**
Asturias	0.42**	1.58**	0.41**	1.84**
Basque Country	0.72	1.38**	0.75	1.49**
Barcelona	0.33**	1.52**	0.32**	1.76**
Catalunya	0.42**	1.62**	0.41**	1.89**
Mid-East Spain	0.24**	2.12**	0.11**	2.43**
Granada	–	2.30**	–	2.34**
Huelva	–	1.75**	–	1.81**
Athens	0.44**	1.45**	0.38**	1.80**
Heraklion	–	1.60**	–	1.56**

*Significant difference between the site types on $p < 0.10$ level.
**Significant on $p < 0.05$ level.

Table 5
Correlation and ratio between NO$_2$ and NO$_x$ (R-squared).

	Study area	Correlation between NO$_2$ and NO$_x$ (R^2)[a]	Ratio NO$_2$/NO$_x$			
			All sites	Regional background	Urban background	Street
1	Oslo	0.96	0.51	0.53	0.54	0.49
2	Umeå Region	0.97	0.57	0.81	0.56	0.42
3	Stockholm County	0.93	0.58	0.62	0.63	0.50
4	Helsinki/Turku	0.91	0.64	0.82	0.66	0.61
5	Copenhagen	0.94	0.65	0.78	0.72	0.58
6	Kaunas	0.91	0.62	0.68	0.64	0.60
7	Bradford	0.88	0.60	0.72	0.64	0.52
8	Manchester	0.91	0.58	–	0.60	0.56
9	London/Oxford	0.93	0.58	0.39	0.63	0.53
10	Netherlands/Belgium	0.94	0.63	0.70	0.68	0.56
11	Ruhr area	0.97	0.59	0.66	0.64	0.53
12	Heidelberg	0.97	0.62	0.66	0.65	0.60
13	Erfurt	0.92	0.65	0.66	0.68	0.62
14	Munich/Augsburg	0.95	0.59	0.60	0.62	0.55
15	Vorarlberg	0.83	0.55	0.64	0.60	0.51
16	Paris	0.93	0.57	0.63	0.64	0.46
17	Grenoble	0.93	0.60	0.70	0.64	0.55
18	Lyon	0.96	0.64	0.95	0.71	0.54
19	Marseille	0.94	0.58	0.80	0.66	0.49
20	Gyor	0.90	0.55	0.63	0.62	0.48
21	Basel	0.94	0.60	0.70	0.62	0.57
22	Geneva	0.88	0.53	0.56	0.53	0.53
23	Lugano	0.83	0.62	0.66	0.62	0.61
24	Turin	0.94	0.54	0.69	0.56	0.52
25	Pavia/Varese	0.96	0.53	0.55	0.57	0.51
26	Verona	0.95	0.47	0.47	0.51	0.45
27	Rome	0.94	0.60	0.59	0.65	0.56
28	Asturias	0.95	0.49	0.55	0.54	0.46
29	Basque Country	0.92	0.50	0.51	0.53	0.49
30	Barcelona	0.93	0.60	0.66	0.65	0.57
31	Catalunya	0.92	0.59	0.66	0.64	0.56
32	Mid-East Spain	0.98	0.71	0.67[b]	0.70	0.61
33	Granada	0.95	0.61	–	0.61	0.61
34	Huelva	0.97	0.65	–	0.67	0.64
35	Athens	0.81	0.51	0.62	0.55	0.44
36	Heraklion	0.92	0.72	–	0.71	0.73

[a] All p-values were below <0.0001.
[b] In Mid-East Spain two regional background sites were selected. However, at one site the NO$_2$ and NO$_x$ concentrations were close to the detection limit (NO$_2$ = 1.9 µg m^{-3}, NO$_x$ = 0.6 µg m^{-3}) and therefore not used for the calculation of the ratio.

performance of the Ogawa badge, though the lack of a consistent pattern across Europe or concentration level argues against this. Ogawa measurements were highly standardized across areas. The difference in Ogawa/monitor ratios between areas may explain a small part of the patterns of concentrations between study areas as these are substantially larger.

A similar North-South gradient across Europe was also observed for PM$_{2.5}$ and PM$_{10}$ (Hazenkamp-von Arxa et al., 2004; Eeftens et al., submitted for publication). However, city size is also clearly visible in the NO$_2$ pattern. NO$_2$ concentrations in the moderately sized cities of Heraklion, Crete, Gyor and Kaunas were similar to those in the Northern large cities of Helsinki, Stockholm and Oslo, whereas the PM$_{10}$ and PM$_{2.5}$ concentrations were substantially lower in the Northern cities. This reflects the larger impact of long range transported secondary aerosol on the PM$_{2.5}$/PM$_{10}$ levels and, in contrast, the predominant role of local source emissions on the NO$_x$ concentrations. Another example is that NO$_2$ concentrations in London exceeded concentrations in the Netherlands and the Ruhr area whereas the PM$_{2.5}$ and PM$_{10}$ concentrations showed the opposite pattern.

With regard to the comparison of the NO$_2$ and NO$_x$ concentrations across the study areas one should keep in mind that the study areas are of substantially different size and type (which is given by the different distribution of the existing study cohorts). Some study areas consist of one single city, some include more rural and/or suburban locations and some others cover whole regions (Catalunya) or even countries (the Netherlands/Belgium). Also the included cities differ regarding their size from small cities like Umeå to the largest European metropolitan areas such as Paris or London. Because of those differences also the number of regional, urban background and street sites in each study area is different.

4.4. NO$_2$ to NO$_x$ ratio

The major sources of NO$_2$ and NO$_x$ are motorized road traffic, industry, shipping and heating. Nitrogen oxides are emitted as NO and NO$_2$. In the atmosphere NO reacts with ozone to form secondary NO$_2$. The observed NO$_2$ to NO$_x$ ratio varied between study areas and site types within study areas. The typically higher ratios observed at urban background sites reflect enhanced transformation of NO to NO$_2$ through equilibrium reactions with ozone at those sites. The relatively modest difference between traffic and urban background sites may reflect the increase in primary NO$_2$ emissions close to street sites. Recent studies reported evidence of increasing NO$_2$ to NO$_x$ ratios from road traffic emissions due to an increase in primary NO$_2$ emissions observed in several urban areas in Europe. Primary NO$_2$ emissions have therefore gained importance compared to the ozone/NO$_x$ equilibrium (Keuken et al., 2009; Mavroidis and

Chaloulakou, 2011). This increase have been attributed to the more common use of diesel-fuelled vehicles, since they emit a higher fraction of NO_2 compared to gasoline-fuelled vehicles (Grice et al., 2009; Anttila et al., 2011; Carslaw et al., 2011). In addition, the after-treatment devices (such as an oxidation catalyst) implemented for reducing particulate matter emissions by diesel vehicles contribute to increasing fraction of primary NO_2 in NO_x (Williams and Carslaw, 2011; Mavroidis and Chaloulakou, 2011). For diesel-fuelled vehicles equipped with catalytic diesel particulate filters, primary NO_2 fractions of around 40–50% are reported (Carslaw et al., 2007).

Significant variability of the fraction of primary NO_2 in traffic emissions across Europe was reported in the studies, but there is less information on NO_2/NO_x concentration ratios.

Due to higher ozone levels and a higher percentage of diesel vehicles in the car fleet, we expected higher NO_2 to NO_x ratio in southern Europe. We did not find this, possibly because of more street canyons within Southern European cities, in which poor dispersion of ozone may become a limiting factor (Vardoulakis et al., 2003). For Athens, it was suggested that the fraction of primary NO_2 is not increasing (as is the case for other urban areas in Europe) as diesel passenger cars are not allowed there and particle after-treatment technologies are not applied in Greece (Mavroidis and Chaloulakou, 2011).

5. Conclusion

We found significant contrasts in annual average NO_2 and NO_x concentrations between and especially within 36 study areas across Europe. The within-area spatial variability contributed significantly to the overall variance of NO_2 and NO_x concentrations (60% and 70%, respectively). It was mostly determined by differences between street and urban background concentrations. The street/urban background concentration ratio varied between 1.09 and 3.16 for NO_2 and between 1.14 and 4.24 for NO_x across all study areas. Concentrations were generally lower in Northern than in Southern Europe, but a clear impact of city size was also found. The NO_2/NO_x ratio varied between 0.47 and 0.72 across study areas.

The substantial spatial variability of NO_2 and NO_x within one given study area observed in this study indicates that future epidemiological long-term studies might consider different approaches of exposure assessment for better characterization of the intra-urban contrasts, e.g. increasing of the number of monitoring sites or modelling of annual average NO_2 and NO_x concentrations across the study area. The use of traffic indicators such as "living close to major road" as an exposure variable in epidemiological studies results in different actual NO_2 contrasts for different study areas and makes the comparison of the studies difficult.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2012.07.080.

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