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Spatial variation of $PM_{2.5}$, PM_{10}, $PM_{2.5}$ absorbance and PM_{coarse} concentrations between and within 20 European study areas and the relationship with NO_2 – Results of the ESCAPE project

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Abbreviations: CV, coefficient of variation; ESCAPE, European Study of Cohort for Air Pollution Effects; GIS, Geographic Information Systems; LUR, Land Use Regression; $PM_{2.5}$, mass concentration of particles less than 2.5 μm in size; $PM_{2.5}$ absorbance, measurement of the blackness of $PM_{2.5}$ filters, this is a proxy for elemental carbon, which is the dominant light absorbing substance; PM_{10}, mass concentration of particles less than 10 μm in size; PM_{coarse}, mass concentration of the coarse fraction of particles between 2.5 μm and 10 μm in size; RB, regional background; RH, relative humidity; ST, Street; TRAPCA, Traffic-Related Air Pollution and Childhood Asthma; UB, urban background; US EPA, United States Environmental Protection Agency.

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HIGHLIGHTS

- We used one method to measure $PM_{2.5}$, $PM_{2.5}$ absorbance & PM_{10} in 20 European areas.
- We studied contrasts of these metrics and PM_{coarse}within andbetweenall 20 areas.
- Concentrationswerehigher in Southern than in Western and Northern European areas.
- Within-area contrasts varied by area andwerelarger for $PM_{2.5}$ absorbance & PM_{coarse}.
- Concentration ratios of particle metrics and NO_2varied significantly across areas.

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ABSTRACT

The ESCAPE study (European Study of Cohorts for Air Pollution Effects) investigates relationships between long-term exposure to outdoor air pollution and health using cohort studies across Europe. This paper analyses the spatial variation of $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10} and PM_{coarse} concentrations between and within 20 study areas across Europe.

We measured NO_2, NO_x, $PM_{2.5}$, $PM_{2.5}$ absorbance and PM_{10} between October 2008 and April 2011 using standardized methods. PM_{coarse} was determined as the difference between PM_{10} and $PM_{2.5}$. In each of the twenty study areas, we selected twenty PM monitoring sites to represent the variability in important air quality predictors, including population density, traffic intensity and altitude. Each site was monitored over three 14-day periods spread over a year, using Harvard impactors. Results for each site were averaged after correcting for temporal variation using data obtained from a reference site, which was operated year-round.

Substantial concentration differences were observed between and within study areas. Concentrations for all components were higher in Southern Europe than in Western and Northern Europe, but the pattern differed per component with the highest average $PM_{2.5}$ concentrations found in Turin and the highest PM_{coarse} in Heraklion. Street/urban background concentration ratios for PM_{coarse} (mean ratio 1.42) were as large as for $PM_{2.5}$ absorbance (mean ratio 1.38) and higher than those for $PM_{2.5}$ (1.14) and PM_{10} (1.23), documenting the importance of non-tailpipe emissions. Correlations between components varied between areas, but were generally high between NO_2 and $PM_{2.5}$ absorbance (average $R^2 = 0.80$). Correlations between $PM_{2.5}$ and PM_{coarse} were lower (average $R^2 = 0.39$). Despite high correlations, concentration ratios between components varied, e.g. the $NO_2/PM_{2.5}$ ratio varied between 0.67 and 3.06.

In conclusion, substantial variability was found in spatial patterns of $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10} and PM_{coarse}. The highly standardized measurement of particle concentrations across Europe will contribute to a consistent assessment of health effects across Europe.

1. Introduction

Human exposure to ambient levels of air pollution is a risk to public health (Brunekreef and Holgate, 2002; Pope and Dockery, 2006; WHO, 2006). It has been estimated that, in Europe, exposure to outdoor and traffic-related air pollution has greater adverse effects on public health in the long term, versus the short term (Künzli et al., 2000). Epidemiological studies have suggested associations of long-term exposure to current air pollution levels and particularly cardio-respiratory health (Brunekreef and Holgate, 2002; Rückerl et al., 2011). Most studies have found associations of health with particulate matter characterized as the mass concentration of particles smaller than 2.5 or 10 μm ($PM_{2.5}$ or PM_{10}) and nitrogen dioxide (NO_2) (Brunekreef and Holgate, 2002). In the EU, the air quality limit values for PM_{10} and NO_2 are still exceeded frequently (European Environment Agency (EEA) 2009; Velders and Diederen 2009), raising significant public concern. Less information is available concerning the exceedance of the new $PM_{2.5}$ guideline (Brunekreef and Maynard, 2008).

Early epidemiological studies compared air pollution concentrations and health outcomes between cities, and have mostly ignored within city variability. Long-term city pollution levels were often characterized by a single (averaged) concentration, based on a limited number of monitors per city, e.g. the American Six Cities Study (Dockery et al., 1993) the American Cancer Society (ACS) study (Pope et al., 2002) and the ECRHS study in Europe (Götschi et al., 2005). Multiple recent studies have shown significant intra-urban spatial contrasts (Beelen et al., 2007; Hoek et al., 2002a; Jerrett et al., 2005b). Land Use Regression (LUR) modelling has been used frequently to explain these spatial contrasts, using predictor variables derived from Geographic Information Systems (GIS) (Hoek et al., 2008; Jerrett et al., 2005a). Several epidemiological studies have since made use of estimated within-urban pollution contrasts based on LUR models (e.g. Morgenstern et al., 2007; Beelen et al., 2008) often focussing mainly on motorized traffic as an important source of intra-urban air pollution contrast (HEI, 2010).

Significant variability of PM_{10} concentrations between cities across Europe has been reported based upon routine monitoring data (European Environment Agency (EEA) 2009), a series of research projects (Putaud et al., 2004; Van Dingenen et al., 2004) and a wintertime study in 14 European cities (Hoek et al., 1997). The lowest concentrations were generally found in Northern Europe and the highest in Southern and Eastern Europe. Spatial variation of $PM_{2.5}$ across Europe is less well characterized because it is not routinely measured in most monitoring networks. Nevertheless, significant north–south gradients have been reported for $PM_{2.5}$ based on research projects (Van Dingenen et al., 2004) and a purpose designed network consisting of 21 urban background stations across Europe (Götschi et al., 2005; Hazenkamp-von Arx et al., 2004). Within-urban contrasts have been characterized in various studies (Monn, 2001; Hoek et al., 2002b), but there are few. The interpretation of spatial contrasts of PM concentrations is limited by differences in site selection, and differences of sampling and analysis methods including different correction factors used to compensate for sampling losses of volatile components between

countries and network operators (European Environment Agency (EEA) 2009; Van Dingenen et al., 2004). Furthermore, there is limited information on the spatial patterns of the coarse fraction, except from a few research projects (Van Dingenen et al., 2004; Puustinen et al., 2007). Yet, there is increasing epidemiological evidence of the adverse health effects of coarse particles (Brunekreef and Forsberg, 2005).

Ambient concentrations of $PM_{2.5}$, PM_{10}, particle composition, NO_2 and NO_x were measured in the framework of the ESCAPE project (European Study of Cohorts for Air Pollution Effects; www.escapeproject.eu). The objective of ESCAPE is to investigate the health effects of long-term exposure to ambient air pollution in 36 study areas across Europe. Individual exposure estimates for cohort subjects will be assigned based on predictions of land-use regression (LUR) models (Hoek et al., 2008), which are developed based on the air pollution measurements and geographic predictors. The study areas were selected because of the availability of informative cohort studies in these areas. We decided on performing study-specific sampling as most existing monitoring networks have insufficient density to capture small-scale spatial variation; locations may not be representative for human, residential exposures, or do not measure all components of interest routinely (e.g. $PM_{2.5}$, $PM_{2.5}$ absorbance). While NO_2 and NO_x were measured in all 36 areas, particulate matter was measured in 20 out of 36 areas. In each of the 20 areas, PM measurements were made at 20 sites using a standardized protocol using identical gravimetric samplers.

The aim of this paper is to assess the spatial contrasts of $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10}, and PM_{coarse} within and between areas. A second aim is to assess the variability of differences between regional background, urban background and street locations across Europe, as a likely source of within-area variability. The third aim is to assess the variability in concentrations ratios and correlations between the various particle metrics and NO_2 across Europe. A detailed analysis of the spatial contrast for NO_2 and NO_x will be reported separately (Cyrys et al., 2012).

2. Methods

2.1. Sampling design

Particulate matter was measured in 20 study areas (Fig. 1). In each study area, 20 sites were measured. Sampling campaigns were conducted over an entire year, Measurements took place in all study areas between October 2008 and April 2011. Participating centres used identical sampling protocols and common criteria for the selection of sampling sites. Furthermore, they employed the same equipment and all samples were analysed centrally at one laboratory (IRAS, Utrecht University). Calculation of the concentrations was performed using identical templates, which were extensively checked by the coordinating institute.

2.2. Sampling site selection

Most study areas included a major city and its surroundings, while others comprised several cities or larger areas. In 18 study areas, 20 sampling sites were selected. In the large study areas of Catalunya and The Netherlands/Belgium, two neighbouring areas were combined and treated as one study area to comprise a total of 40 monitoring sites. In Catalunya, 20 sites were located in the city of Barcelona. Separate LUR models were made for Barcelona and Catalunya. Therefore, Barcelona and Catalunya areas are presented separately throughout this paper. An overview of the study area characteristics can be found in Table 1, and more detailed descriptions and maps of the individual study areas in Online Supplement A.

All participating centres selected sampling sites locally, by considering the spatial distribution of the cohorts of interest and the specific characteristics of the study area. Measurement sites were selected to reflect a large diversity of potential sources of air pollution variability (e.g. population density, traffic intensity, industry, proximity to harbours etc). In each area, we selected regional background, urban background and street sites. Regional background sites were located outside of major urban areas, and were not influenced directly by traffic sources. Urban background sites were located inside an urban area, but at least 50 m away from major roads. Street sites were selected at building facades representative for homes, in streets with traffic intensities of around 10,000 vehicles per day or more. All sites were selected so that the influence of other local sources of particulate matter and combustion gases (e.g. construction works, district heating plants, parking lots) within 100 m was minimized. Because of the importance of motorized traffic, we chose to over-represent street sites. We included a variety of streets with different traffic intensity, distance of the sampling site to the road, and different street configurations. Site selection proposals were evaluated centrally, to ensure that all centres applied the same selection criteria.

To adjust for the temporal variability of concentrations, one centrally located reference site was chosen in each study area where measurements were taken over an entire year, following the design of a previous study (Hoek et al., 2002b) and other LUR studies (Hoek et al., 2008). The reference site was chosen at a regional or urban background location, not directly influenced by local sources.

2.3. Sampling schedule

For each site, PM measurements were done 3 times for 14 days, in different seasons. Due to a limited amount of samplers, five sites and the reference site were measured simultaneously representing all different site types (regional background, urban background and street). Due to a limited amount of samplers, it was not feasible to measure at all sites simultaneously within a study area. The 14-day average measurements at the reference site continued between and after measurement rounds for a full year. If a measurement at one of the sites failed, it was repeated in a later round for all components, to ensure that three valid measurements were available for each site.

2.4. Measurement methods

The standard operating procedure is available from the ESCAPE project website (http://www.escapeproject.eu/manuals/). All particulate matter samples were collected using Harvard impactors, designed to collect particles smaller than 2.5 μm ($PM_{2.5}$) or smaller than 10 μm (PM_{10}) at a flow rate of 10 l min^{-1} (Hoek et al., 2002b; Brunekreef et al., 2005). The air flow was measured before and after sampling, using the same type of rotameters. Rotameters were centrally provided and calibrated at Utrecht University before and after the sampling year, using a soap film device. Samples with a start or end flow below 8.0 l min^{-1} were excluded. Total sampling volume was calculated for each filter based on the average flow and total sampling time. A single pump unit was used at each site, operating both the $PM_{2.5}$ and PM_{10} inlets simultaneously. To prevent overload of filters, we employed timers which were set to sample for 15 min every 2 h (12:00–12:15, 2:00–2:15, 4:00–4:15 etc) so that effectively a 42-h sample was collected over 14 days. Total runtime was recorded by elapsed time counters. Samples which had a sampling duration of less than 28 or more than 56 h (±33% of 42 h) were excluded from analysis. Failed measurements were repeated in a later round for all components.

Fig. 1. The ESCAPE study areas.

Table 1
Descriptive characteristics of 20 ESCAPE study areas where airborne particulate matter was measured.

Country	Study area	Study area description; major cities	Year	Dates	Total number of sites	Distribution over site types: (RB/UB/ST)
Norway	Oslo	Oslo city	1	05-02-2009–29-01-2010	19[a]	3/9/8
Sweden	Stockholm County	Stockholm County; Stockholm	1	03-12-2008–01-12-2009	20	3/6/11
Finland	Helsinki/Turku	Two areas: Helsinki/Vantaa and Turku/Loimaa	2	27-01-2010–26-01-2011	20	2/10/8
Denmark	Copenhagen	Copenhagen city and Hillerød	2	19-11-2009–17-11-2010	20	3/6/11
Lithuania	Kaunas	Kaunas city	2	20-01-2010–19-01-2011	20	4/6/10
United Kingdom	Manchester	Greater Manchester urban area	1	27-01-2009–20-01-2010	20	0/8/12
	London/Oxford	Thames valley: London, Oxford and smaller towns	2	26-01-2010–18-01-2011	20	1/12/7
Netherlands/ Belgium	Netherlands/Belgium	Entire Country: Amsterdam, Rotterdam, Antwerp, Utrecht, Groningen, Maastricht, Doetinchem, Amersfoort and smaller towns	1	17-02-2009–19-02-2010	40	10/12/18
Germany	Ruhr area	Dortmund, Duisburg, Essen and smaller towns	1	15-10-2008–12-10-2009	20	4/8/8
	Munich/Augsburg	Munich, Augsburg and smaller surrounding towns	1	27-10-2008–05-11-2009	20	5/6/9
Austria	Vorarlberg	Cities and areas along the main valley of Vorarlberg	2	03-03-2010–16-02-2011	20	3/7/10
France	Paris	Paris city and suburban areas	2	04-01-2010–04-01-2011	20	4/9/7
Hungary	Gyor	Gyor city and neighbouring villages	2	22-02-2010–24-02-2011	20	1/9/10
Switzerland	Lugano	Lugano city and its neighbouring communities	1	02-03-2009–10-03-2010	18[a]	3/6/10
Italy	Turin	Turin city and five smaller municipalities (Collegno, Moncalieri, Grugliasco, Nichelino, Rivoli)	2	01-02-2010–25-01-2011	20	1/8/11
	Rome	Rome city	2	27-01-2010–26-01-2011	20	2/8/10
Spain	Barcelona	Barcelona city	1	14-01-2009–14-01-2010	20	1/8/11
	Catalunya	Three areas around Barcelona, Girona, Sabadell	1	14-01-2009–14-01-2010	40	4/13/23
Greece	Athens	Greater Athens area, 16 municipalities; Athens	2	21-04-2010–27-04-2011	20	1/12/7
	Heraklion	Heraklion prefecture; Heraklion	1	18-02-2009–16-02-2010	20	0/12/8

Dates refer to the period when the reference site was operated, RB = regional background, UB = urban background and ST = street site.
[a] Failed measurements explain fewer than 20 sites for Lugano and Oslo.

The weighing and reflectance protocols are available from the ESCAPE project website (http://www.escapeproject.eu/manuals/). Briefly, we used Andersen 37 mm 2 μm pore size Teflon filters for particulate matter sampling. All filters were pre- and post-weighed at a central laboratory (IRAS, Utrecht University, Utrecht, The Netherlands) on a microbalance, following a standard operating procedure, previously described in Brunekreef et al. (2005). Prior to each weighing session, filters were conditioned for at least 24 h inside the weighing room, at a constant temperature of 23 ± 1 °C and constant relative humidity of 37 ± 2%. A Polonium de-ionizer was used to discharge static electricity from the filters prior to weighing. Reflectance of all filters was also measured in the central laboratory after post-weighing, using the same procedure, also described in Brunekreef et al. (2005). Reflectance was transformed into absorbance (a) according to (ISO (International Standardization Organization) 1993) (Equation (1))

$$a = \frac{A}{2V} * \ln\left(\frac{R_F}{R_S}\right) \quad (1)$$

Equation (1): Calculation of the absorbance from reflectance measurements. Where A is the area of the stain no the filter ($780*10^{-6}$ m^2), V is the volume sampled in m^3, R_F is the average reflectance of the field blank filters as percentage of R_0 (the reflectance of the clean control filter, 100.0 by definition) and R_S is the reflectance of the sample filter as a percentage of R_0.

Absorbance has previously been found to be highly correlated with elemental carbon (Cyrys et al., 2003). We report only the absorbance of the $PM_{2.5}$ filters, as most of the elemental carbon is found in the fine fraction (Putaud et al., 2004). Consistently, in the TRAPCA study (Traffic-Related Air Pollution and Childhood Asthma), $PM_{2.5}$ absorbance was highly correlated with and almost identical to PM_{10} absorbance (Cyrys et al., 2003). PM_{coarse} concentrations were calculated by subtracting $PM_{2.5}$ from PM_{10} concentrations.

2.5. Quality control

Filters were transported to and from the central laboratory in individual petri dishes, and were sealed and cooled during transport. A field blank and duplicate for PM_{10} were collected at the reference site during every sampling period. Field blank filters were placed inside unconnected Harvard impactors, and remained on site for 14 days. PM_{10} field blanks were assumed to be representative of $PM_{2.5}$ measurements as well, as the same type of filters and impactors were used. The limit of detection was calculated as 3 times the standard deviation of the field blanks. The average field blank concentration was subtracted from each sample to correct for the effects of transport and handling. Reproducibility of the measurements was assessed by calculating the coefficient of variation (CV) from the duplicates and their corresponding samples using Equation (2) (Hoek et al., 2002a,b). Similar to the detection limit, the coefficients of variation calculated for PM_{10} were assumed to be representative also for $PM_{2.5}$. Since PM_{coarse} is calculated as the difference between PM_{10} and $PM_{2.5}$ concentrations, its variance is higher ($\sqrt{2}*SD$).

$$CV = \frac{\sqrt{\frac{\sum_{i=1}^{n}(S_i - D_i)^2}{2*n}}}{\frac{\sum_{i=1}^{n}(S_i + D_i)}{2*n}} * 100\% \quad (2)$$

Equation (2): Calculation of coefficient of variation. Where n is the number of duplicates and i is the sampling round (1 to n). S is the concentration of sample i and D is the concentration of corresponding duplicate i.

2.6. Temporal adjustment

For each round, a temporal correction was calculated as the difference of each individual reference site measurement from the annual mean at the reference site. Subsequently, measurements at all other sites were corrected for temporal variation, by subtracting the correction calculated for that particular round.

Based upon better performance in the TRAPCA study (Hoek et al., 2002b), we used the absolute difference between the yearly average and each individual reference site measurement as a correction instead of the ratio between the two. To verify this assumption, we compared results from the ratio method to those obtained from the difference method in three different areas (Stockholm County, the Netherlands/Belgium and Catalunya) and report the results in Online supplement B.

2.7. Data analysis

Locally calculated adjusted annual averages were gathered centrally and their range and distribution were calculated, stratified by site type. To quantify the amount of spatial variation, the total range (maximum−minimum) was 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. Relationships between concentrations of pollutants were expressed as R^2-values determined using SAS 9.2, PROC REG. R^2 is the Pearson correlation squared. Percentages of between and within area variance were determined using analysis of variance with PROC MIXED. We analysed overall patterns and patterns in three groups of study areas: Northern (Oslo, Stockholm County, Helsinki/Turku, Copenhagen), Southern (Turin, Rome, Barcelona, Catalunya, Athens and Heraklion) and West/Central European (other areas).

3. Results

3.1. Quality control

Detection limits were low for all centres for both PM_{10} (0.7–4.0 μg m^{-13}) and PM_{10} absorbance (0.04–0.10 × 10^{-5} m^{-1}). Only 3 samples (2 for $PM_{2.5}$ and 1 for PM_{10}) from Helsinki/Turku were below the detection limit (Online supplement C). We retained the original values. Reproducibility was good in most areas, as coefficients of variation (CV) varied between 2% and 7% for PM_{10}, and between 2% and 5% for PM_{10} absorbance (Online supplement C). CV values for PM_{10} in Helsinki/Turku and Manchester were higher (11% and 37% respectively), caused in both cases by a single pair of incomparable duplicates.

3.2. Adjustment for temporal variation

Unadjusted mean concentrations correlated well with temporally adjusted means for $PM_{2.5}$ absorbance, PM_{10} and PM_{coarse} for most areas (Online Supplement B, Table B.1). For $PM_{2.5}$, adjusted mean concentrations correlated moderately with unadjusted means. The more moderate correlations for $PM_{2.5}$ are probably due to relatively large temporal variation compared to other components (Online Supplement B, Table B.2). A comparison between the

difference and ratio adjustment approaches in Catalunya, Stockholm and the Netherlands revealed that adjusted concentrations from both methods were highly correlated with each other for all three selected areas (Online Supplement B, Table B.3).

3.3. Spatial variability within and between study areas

Spatial variability of the average concentration for all components between and within study areas is shown in Fig. 2A–D and Table 2, respectively. Contrasts between background and street sites are shown in Figs. 3–6.

3.3.1. $PM_{2.5}$

Concentration levels of $PM_{2.5}$ were lowest in the Northern European study areas and highest in the Southern European (particularly Turin) and two Eastern European study areas (Gyor and Kaunas). Clear regional patterns are visible, e.g. with similar concentrations in the Netherlands/Belgium and the Ruhr Area, which exceed those measured in London and Manchester (Fig. 2A). There was substantial variation within most study areas. The lowest contrasts (range $<$ 50% of the mean) were found in Manchester, Ruhr area, Gyor, and Turin (Table 2). Absolute $PM_{2.5}$ contrasts were largest in the Southern study areas: Rome, Barcelona and Catalunya (Fig. 3).

Concentrations at street sites were significantly higher than those at urban background sites for 12 out of the 20 areas (Table 3). Street/urban background ratios varied between 0.96 and 1.30, with no differences in ratios between the Northern, Western/central and southern European study areas. Regional/urban background ratios were lower than 1 for all study areas except Munich/Augsburg and Rome (Table 3).

3.3.2. $PM_{2.5}$ absorbance

For $PM_{2.5}$ absorbance, the lowest median concentrations were found in Northern European study areas and Heraklion. The highest concentrations were found in the Southern European study areas (Rome, Turin and Barcelona) (Fig. 2B). Gyor and Kaunas had high concentrations of $PM_{2.5}$, but only moderately high concentrations of $PM_{2.5}$ absorbance. In general, within study area contrasts for $PM_{2.5}$ absorbance were larger than those for $PM_{2.5}$ (Fig. 4, Table 2).

For all study areas, concentrations were higher at street sites than at urban background sites. Street/urban background ratios varied between 1.02 and 1.77, with statistically significant differences for almost all study areas (Table 3). Mean ratios were 1.51, 1.32 and 1.35 for the Northern, West/Central and Southern

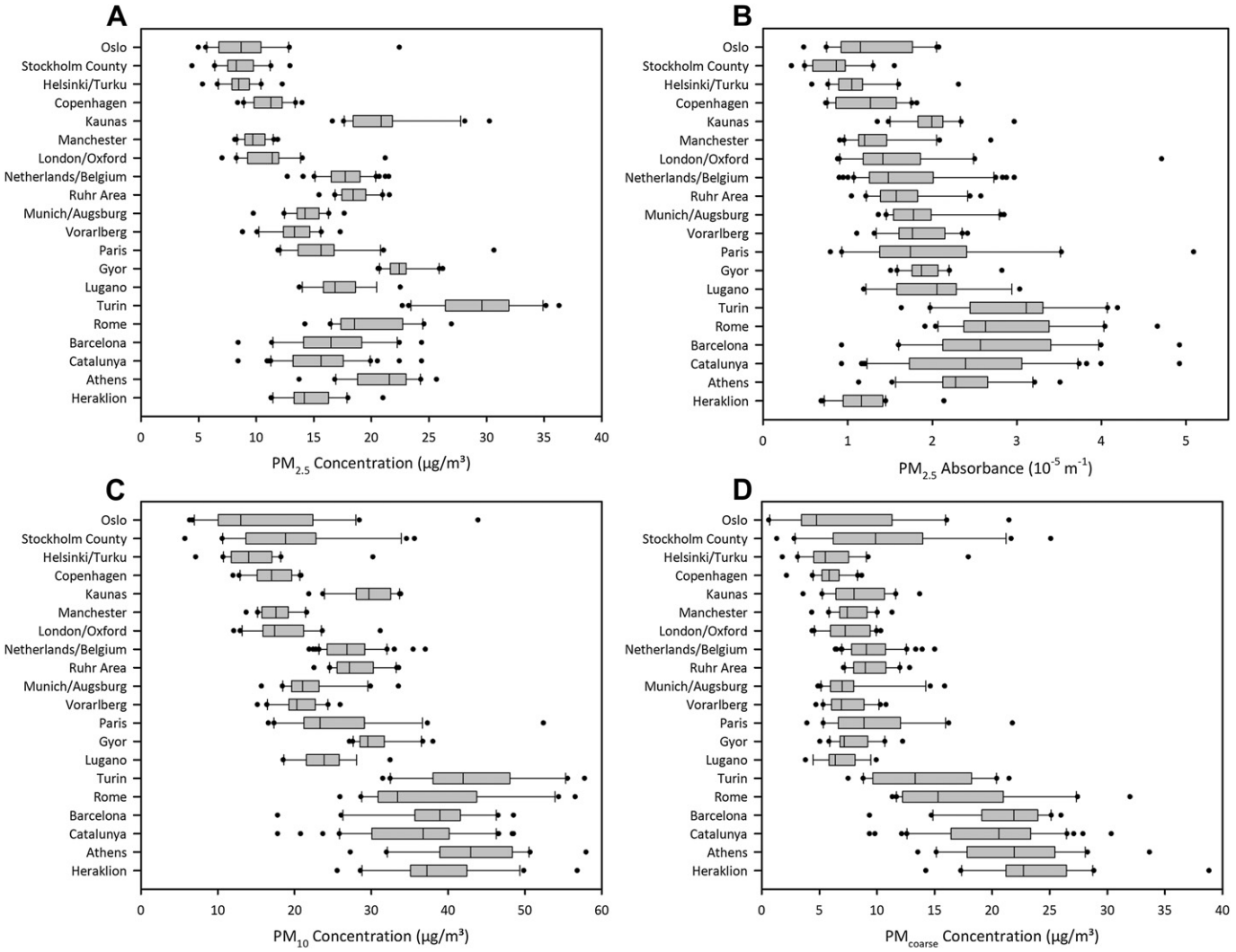

Fig. 2. Distribution of average concentrations within study areas for (A) $PM_{2.5}$, (B) $PM_{2.5}$ absorbance, (C) PM_{10}, and (D) PM_{coarse}. Median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points.

Table 2
Mean and overall contrasts (total range/mean) of adjusted annual average concentrations of $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10} and PM_{coarse} by study area.

Study area	$PM_{2.5}$		$PM_{2.5}$ absorbance		PM_{10}		PM_{coarse}	
	Mean (*n*)	Range/mean (%)	Mean (*n*)	Range/mean (%)	Mean (*n*)	Range/mean (%)	Mean (*n*)	Range/mean (%)
Oslo	8.6 (20)	91%	1.3 (20)	109%	14.8 (19)	149%	6.1 (19)	259%
Stockholm County	8.5 (20)	100%	0.8 (20)	144%	19.1 (20)	156%	10.6 (20)	224%
Helsinki/Turku	8.6 (20)	80%	1.1 (20)	155%	14.8 (20)	156%	6.2 (20)	260%
Copenhagen	11.1 (20)	50%	1.2 (20)	88%	17.1 (20)	52%	6.0 (20)	109%
Kaunas	21.1 (20)	65%	2.0 (20)	81%	29.5 (20)	41%	8.4 (20)	121%
Manchester	9.8 (20)	38%	1.4 (20)	132%	17.6 (20)	45%	7.7 (20)	90%
London/Oxford	11.2 (20)	127%	1.6 (20)	235%	18.6 (20)	103%	7.4 (20)	81%
Netherlands/Belgium	17.7 (40)	50%	1.7 (40)	123%	27.1 (40)	56%	9.3 (40)	92%
Ruhr Area	18.5 (20)	33%	1.6 (20)	93%	27.9 (20)	39%	9.4 (20)	61%
Munich/Augsburg	14.3 (20)	55%	1.9 (20)	79%	22.1 (20)	81%	7.7 (20)	142%
Vorarlberg	13.3 (20)	64%	1.8 (20)	72%	20.6 (20)	52%	7.3 (20)	83%
Paris	16.0 (20)	117%	2.0 (20)	213%	25.6 (20)	140%	9.6 (20)	185%
Gyor	22.6 (20)	25%	1.9 (20)	68%	30.6 (20)	36%	8.0 (20)	90%
Lugano	17.2 (19)	51%	2.0 (19)	91%	23.9 (18)	58%	6.8 (18)	90%
Turin	29.3 (20)	47%	3.0 (20)	85%	43.1 (20)	61%	13.8 (20)	101%
Rome	19.8 (20)	64%	2.9 (20)	96%	37.0 (20)	83%	17.2 (20)	120%
Barcelona	16.3 (20)	97%	2.7 (20)	146%	37.4 (20)	82%	21.0 (20)	79%
Catalunya	15.6 (40)	102%	2.5 (40)	162%	35.6 (40)	86%	20.0 (40)	105%
Athens	20.9 (20)	57%	2.4 (20)	101%	42.8 (20)	72%	21.9 (20)	92%
Heraklion	14.7 (20)	66%	1.2 (20)	121%	38.4 (20)	81%	23.6 (20)	104%

Differences between areas might be affected by different numbers of regional background, urban background and street sites selected.

European areas respectively. Regional background levels were lower than urban background levels for all study areas, with regional/urban ratios varying between 0.45 and 0.92, with no differences between the Northern, West-Central and Southern European areas (Table 3).

3.3.3. PM_{10}

The pattern for PM_{10} was similar to $PM_{2.5}$ with low concentrations in Northern Europe and high concentrations in southern and Eastern Europe (Fig. 2C). In Heraklion, which had moderate concentrations of $PM_{2.5}$ and $PM_{2.5}$ absorbance, high PM_{10} concentrations were found (Fig. 2C), reflecting coarse particles. Lower contrasts were found in both Kaunas and Gyor study areas (Fig. 5, Table 2).

Street/urban background ratios ranged between 1.03 and 1.76 and were significantly higher than 1 for 13 out of 20 areas (Table 3). In nearly all study areas, the street/urban background ratio was higher for PM_{10} than for $PM_{2.5}$. In the Northern European study areas the street/urban background ratio (mean 1.43) was higher than in the western (1.15) and southern (1.17). Regional background concentrations were lower than urban background concentrations in all areas (regional/urban ratios between 0.52 and 0.99) (Table 3).

3.3.4. PM_{coarse}

Coarse particles exhibited a different spatial pattern than $PM_{2.5}$. Concentrations were highest in Southern European areas, but the highest concentrations were found in Heraklion, Athens, and Barcelona, with more modest concentrations in Turin and Rome (Fig. 2D). Concentrations in the Northern European areas were low, but did not differ much from the Western and Central European areas (Fig. 2D). In Stockholm County, PM_{coarse} levels were similar to those measured in Southern Europe. Contrasts in PM_{coarse} were similar to those of PM_{10}, and are similarly high for the Northern European areas (Fig. 6, Table 2).

Street/Urban background ratios were all above 1, ranging from 1.06 to 3.41. This difference was statistically significant in 14 out of 20 study areas (Table 4). Mean street/urban background ratios were 1.97, 1.27 and 1.31 for the Northern, West-Central and Southern European areas respectively. The ratio was typically much larger than for $PM_{2.5}$ and similar to the ratio observed for absorbance and statistically significant in 14 out of 20 study areas (Table 3). Ratios between regional and urban background levels were only significant in 4 areas, and varied widely between 0.36 and 1.10 (Table 3).

3.3.5. Analysis of variance

Between area variance was higher than within area variance for $PM_{2.5}$, PM_{10} and PM_{coarse} (81%, 72% and 71% of the total, respectively). For PM_{coarse} this was mainly caused by the high concentrations in the Southern European study areas (Heraklion, Athens, Barcelona, Catalunya). For $PM_{2.5}$ absorbance, we found that within area variance (52% of the total) was slightly larger than between area variance (48%), indicating that $PM_{2.5}$ absorbance varies predominantly on a local scale.

3.4. Relation between the pollutants

The correlation between the different particle metrics and NO_2 within study areas was highly variable across Europe (Table 4). The correlation (R^2) between $PM_{2.5}$ and PM_{10} was high: 0.74 (range 0.44–0.95), but the correlation between the fine and coarse fractions ($PM_{2.5}$ and PM_{coarse}) was moderate 0.39 (range 0.02–0.81). $PM_{2.5}$ absorbance had a moderately high correlation with the particle mass metrics.

The correlations (R^2) between NO_2 and $PM_{2.5}$ absorbance were generally high: 0.80 (range 0.55–0.91), and were similar between NO_x and $PM_{2.5}$ absorbance: 0.83 (range 0.41–0.98), while the correlation between NO_2 and $PM_{2.5}$ was much lower: 0.50 (range 0.02–0.90). There was no clear spatial pattern of the correlations across Europe (Table 4).

Despite the high *correlation* between several components, the *ratio* of average concentrations varied substantially across study areas (Table 5), thereby indicating the different compositions of these pollution mixtures. The median $NO_2/PM_{2.5}$ ratio varied from 0.67 to 3.06. The median $PM_{2.5}/PM_{2.5}$ absorbance ratio varied between 6.24 and 12.96. The $PM_{2.5}/PM_{10}$ ratio ranged from 0.39 to 0.74, with the lowest values in the south and higher values in Eastern Europe (Kaunas and Gyor).

4. Discussion

We found significant concentration differences for $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10} and PM_{coarse} across 20 European study areas.

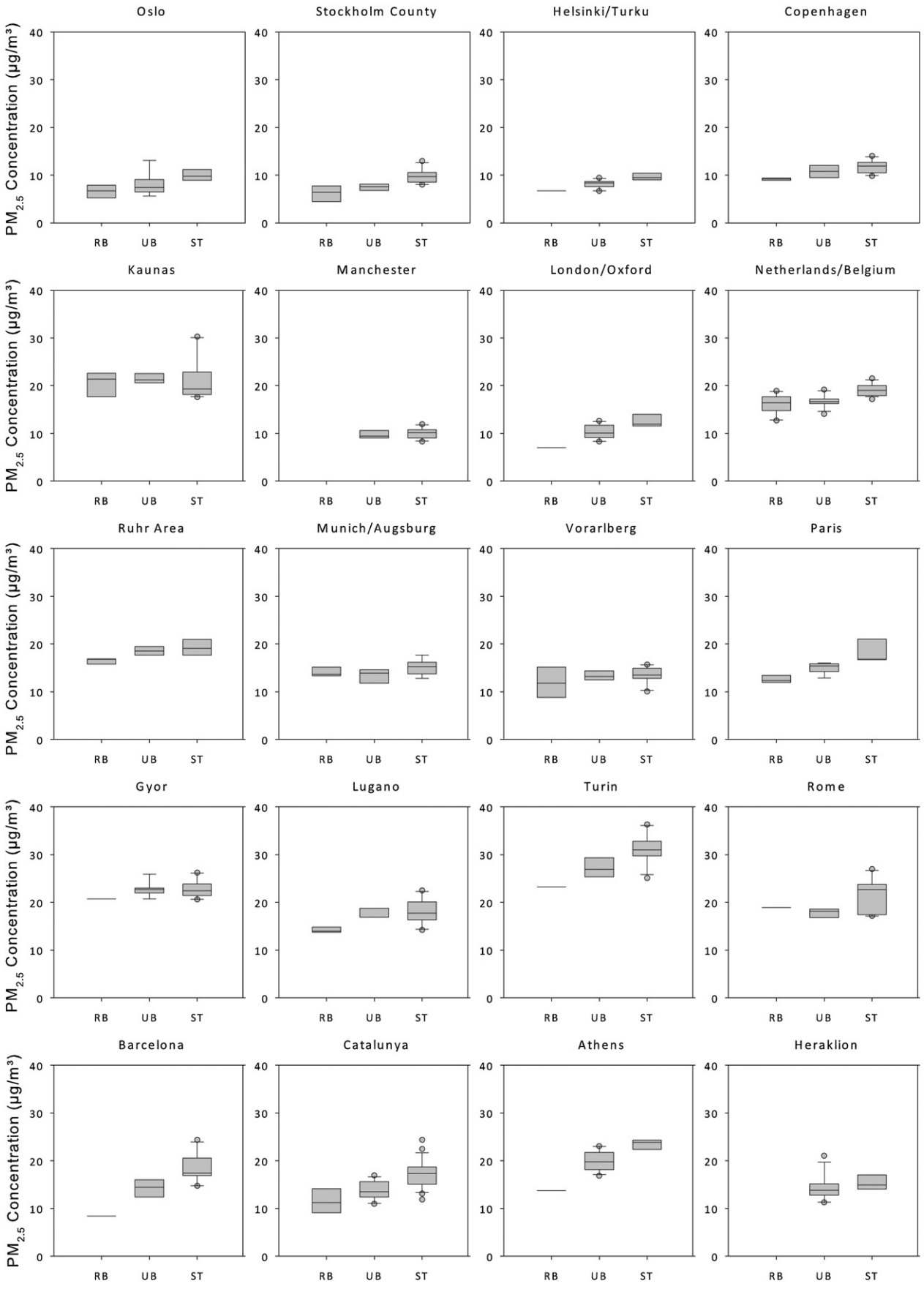

Fig. 3. Adjusted annual average $PM_{2.5}$ concentrations by site type, for each study area median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points. RB = regional background, UB = urban background and ST = street site.

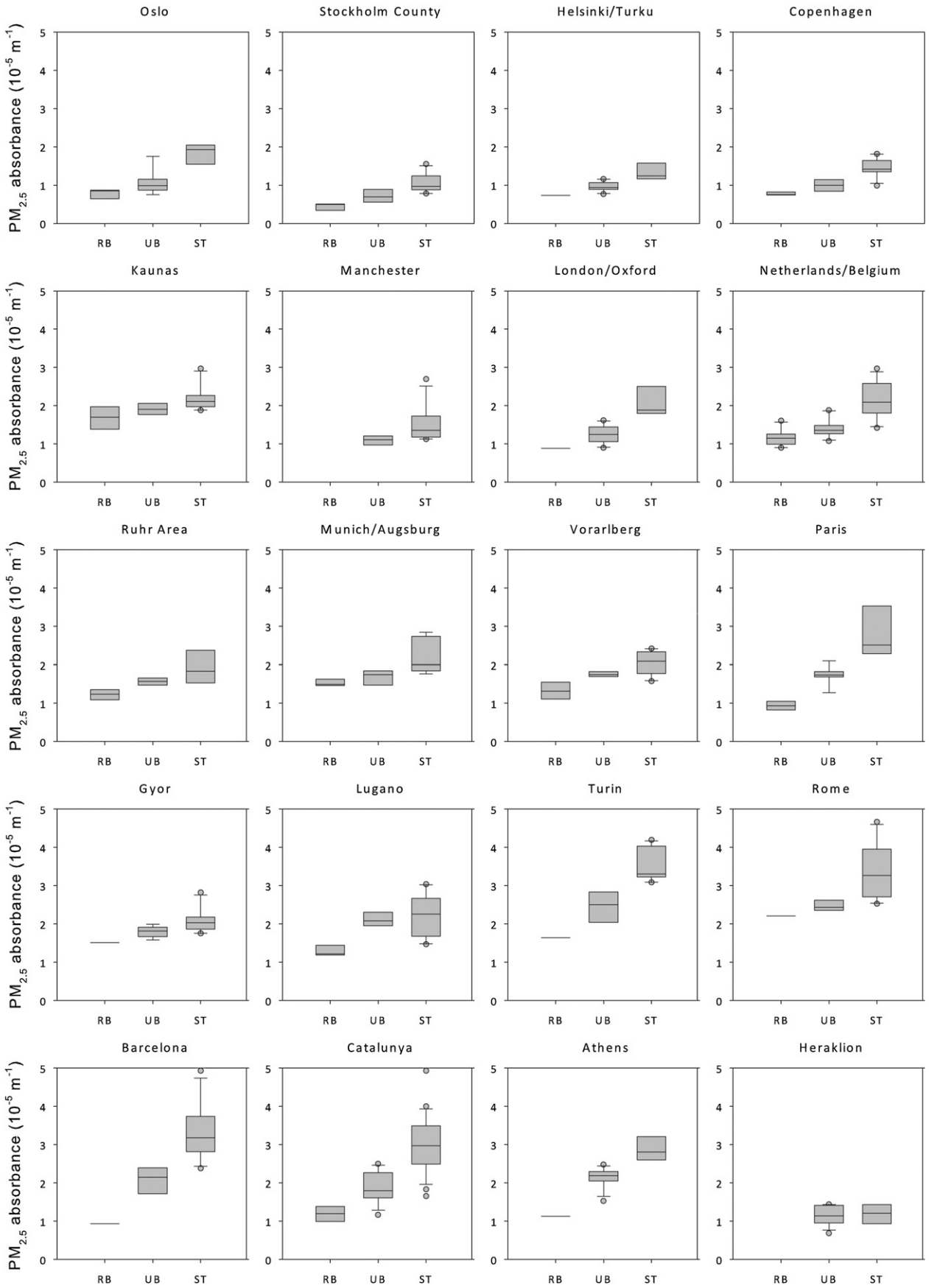

Fig. 4. Adjusted annual average $PM_{2.5}$ absorbance concentrations by site type, for each study area median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points. RB = regional background, UB = urban background and ST = street site.

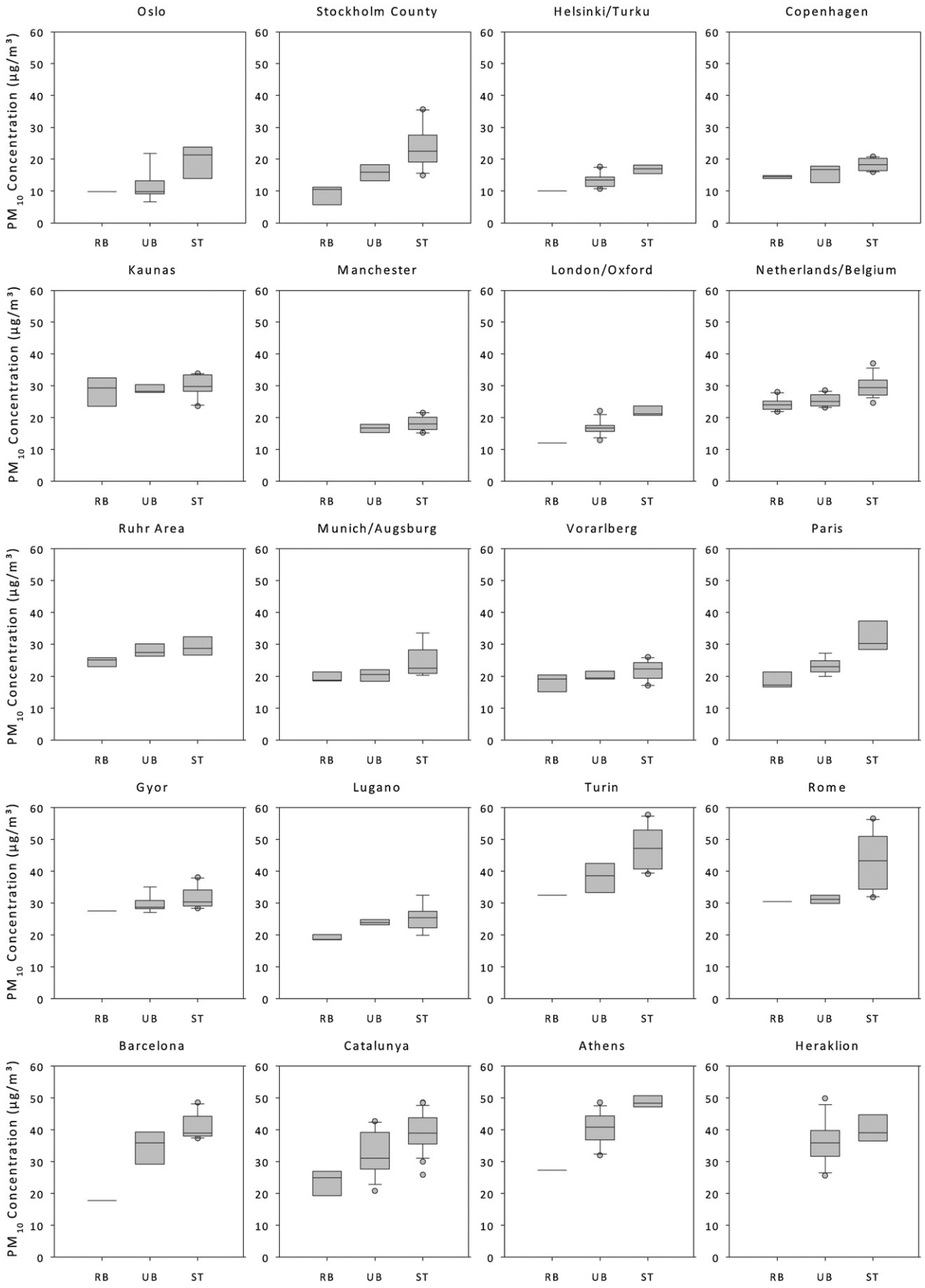

Fig. 5. Adjusted annual average PM_{10} concentrations by site type, for each study area median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points. RB = regional background, UB = urban background and ST = street site.

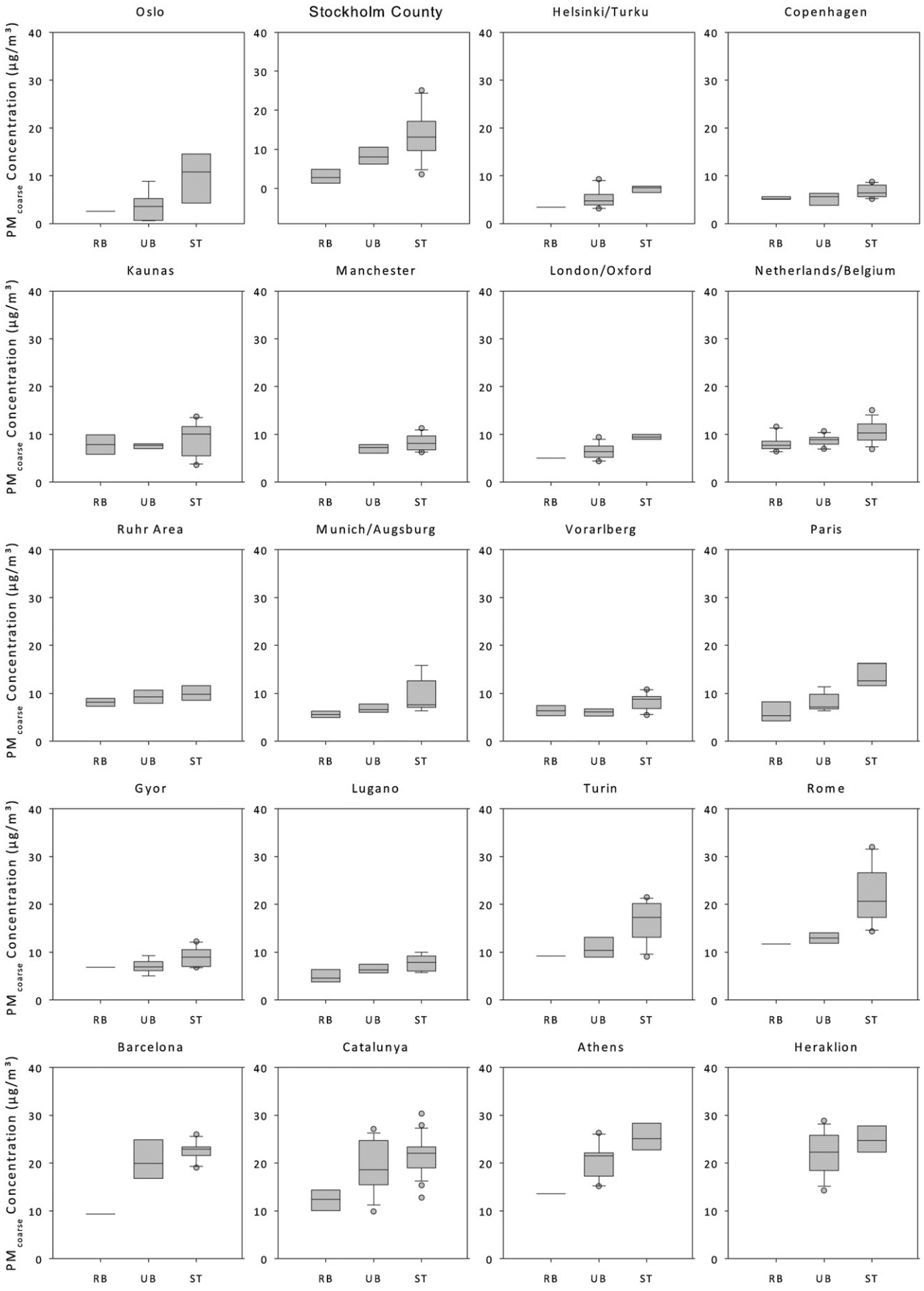

Fig. 6. Adjusted annual average PM_{coarse} concentrations by site type, for each study area median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points. RB = regional background, UB = urban background and ST = street site.

Table 3
Ratios between regional background (RB) and urban background (UB) average concentrations, and between street (ST) and urban background (UB) concentrations, for all study areas.

Study_area	$PM_{2.5}$		$PM_{2.5}$ absorbance		PM_{10}		PM_{coarse}	
	Ratio RB/UB	Ratio ST/UB	Ratio RB/UB	Ratio ST/UB	Ratio RB/UB	Ratio ST/UB	Ratio RB/UB	Ratio ST/UB
Oslo	0.83	1.28**	0.76*	1.73**	0.86	1.76**	0.47	3.41**
Stockholm County	0.81*	1.29**	0.62**	1.42**	0.56**	1.45**	0.32**	1.53*
Helsinki/Turku	0.81**	1.19**	0.74*	1.42**	0.73*	1.33**	0.61	1.56**
Copenhagen	0.86*	1.11	0.77**	1.47**	0.92	1.18**	1.10	1.38**
Kaunas	0.95	0.96	0.88	1.14*	0.97	1.03	1.03	1.13
Manchester	N/A	1.04	N/A	1.32**	N/A	1.11*	N/A	1.20*
London/Oxford	0.69*	1.26**	0.71	1.77**	0.72**	1.33**	0.80	1.44**
Netherlands/Belgium	0.97	1.15**	0.82**	1.52**	0.95	1.17**	0.92	1.19**
Ruhr_Area	0.88**	1.03	0.77**	1.19*	0.88**	1.04	0.87	1.06
Munich/Augsburg	1.07	1.15**	0.92	1.31**	0.98	1.21**	0.81	1.30*
Vorarlberg	0.85	0.98	0.76**	1.17**	0.92	1.10	1.05	1.36**
Paris	0.83*	1.24**	0.54**	1.65**	0.79*	1.37**	0.71*	1.61**
Gyor	0.92	1.00	0.84	1.15**	0.93	1.06	0.99	1.27**
Lugano	0.81**	1.02	0.60**	1.02	0.79**	1.05	0.75*	1.18
Turin	0.85	1.14**	0.67**	1.43**	0.85	1.24**	0.86	1.49**
Rome	1.07	1.21**	0.91	1.37**	0.99	1.37**	0.89	1.59**
Barcelona	0.60**	1.30**	0.45**	1.59**	0.52**	1.19**	0.46**	1.11
Catalunya	0.82**	1.25**	0.63**	1.57**	0.72**	1.20**	0.65**	1.16*
Athens	0.69**	1.19**	0.53**	1.36**	0.68**	1.23**	0.67**	1.27**
Heraklion	N/A	1.09	N/A	1.06	N/A	1.15*	N/A	1.19*
Mean	0.85	1.14	0.72	1.38	0.82	1.23	0.78	1.42

*Significant difference between the site types on $p < 0.10$ level, **significant on $p < 0.05$ level. N/A means that no regional sites were measured in this study area. Ratios were obtained from a regression model with log(concentrations) as dependent variables and site type as independent variable.

Especially for $PM_{2.5}$ absorbance and PM_{coarse} we saw significant contrasts within study areas. For $PM_{2.5}$ absorbance, the contrast within study areas was larger than between study areas. Concentrations at street sites were higher than at urban background sites for all components, but the ratios differed widely across the study areas. PM_{10} and $PM_{2.5}$ concentrations generally correlated well with NO_2 and absorbance, however, the ratios of absolute concentrations differed substantially.

The strength of this study is that we used a standardized approach for selecting sampling sites, a common study setup, the same equipment, identical sampling protocols and that all analyses were done by one central laboratory, so spatial contrasts could be compared both within and between the 20 study areas. This allowed us to assess the true variability across Europe of concentrations of fine and coarse particulate matter; the relation of various PM metrics (correlation and ratio); the variability of the increment at street sites relative to urban background. Our results indicate significant differences in the PM mixture across Europe which are not due to differences in measurement methods We used a gravimetric method to measure PM_{10} which is less affected by losses of

Table 4
Correlation (R^2) between annual average concentrations for different components.

Study_area	NO_2				$PM_{2.5}$			$PM_{2.5}$ absorbance		PM_{10}
	$PM_{2.5}$	$PM_{2.5}$ absorbance	PM_{10}	PM_{coarse}	$PM_{2.5}$ absorbance	PM_{10}	PM_{coarse}	PM_{10}	PM_{coarse}	PM_{coarse}
Oslo	0.24	0.74	0.34	0.29	0.61	0.62	0.37	0.48	0.34	0.93
Stockholm County	0.75	0.88	0.80	0.68	0.78	0.62	0.42	0.85	0.73	0.96
HelsinkiTurku	0.71	0.81	0.80	0.75	0.80	0.84	0.69	0.81	0.74	0.97
Copenhagen	0.40	0.86	0.60	0.50	0.44	0.75	0.22	0.54	0.36	0.71
Kaunas	0.04	0.55	0.17	0.06	0.23	0.50	0.20	0.30	0.00	0.11
Manchester	0.40	0.88	0.59	0.36	0.49	0.46	0.07	0.64	0.38	0.79
London/Oxford	0.84	0.88	0.82	0.39	0.79	0.86	0.29	0.83	0.46	0.67
Netherlands/Belgium	0.57	0.86	0.74	0.53	0.71	0.72	0.23	0.74	0.42	0.76
Ruhr_Area	0.69	0.89	0.65	0.50	0.80	0.91	0.67	0.68	0.46	0.91
Munich/Augsburg	0.29	0.87	0.67	0.68	0.50	0.63	0.28	0.78	0.68	0.87
Vorarlberg	0.04	0.59	0.35	0.52	0.34	0.62	0.02	0.69	0.47	0.53
Paris	0.86	0.90	0.91	0.86	0.91	0.95	0.81	0.92	0.85	0.95
Gyor	0.02	0.65	0.12	0.21	0.31	0.73	0.24	0.46	0.41	0.76
Lugano	0.66	0.58	0.83	0.60	0.76	0.83	0.28	0.82	0.48	0.70
Turin	0.65	0.87	0.67	0.51	0.77	0.81	0.47	0.75	0.54	0.87
Rome	0.73	0.89	0.75	0.67	0.79	0.86	0.69	0.80	0.70	0.96
Barcelona	0.90	0.91	0.69	0.32	0.94	0.80	0.40	0.74	0.35	0.83
Catalunya	0.72	0.89	0.63	0.41	0.83	0.73	0.39	0.73	0.46	0.88
Athens	0.49	0.85	0.70	0.66	0.71	0.74	0.44	0.79	0.65	0.91
Heraklion	0.18	0.63	0.37	0.42	0.20	0.78	0.59	0.40	0.45	0.96
Mean	0.51	0.80	0.61	0.50	0.64	0.74	0.39	0.69	0.50	0.80
Mean North	0.53	0.82	0.64	0.56	0.66	0.71	0.43	0.67	0.54	0.89
Mean West/Central	0.44	0.77	0.59	0.47	0.58	0.72	0.31	0.69	0.46	0.71
Mean South	0.50	0.80	0.64	0.52	0.64	0.75	0.34	0.73	0.52	0.78

Table 5
Median ratios between average concentrations of different components per study area.

Study_Area	$NO_2/PM_{2.5}$	$NO_2/PM_{2.5}$ absorbance	NO_2/PM_{10}	NO_2/PM_{coarse}	$PM_{2.5}/PM_{2.5}$ absorbance	$PM_{2.5}/PM_{10}$	$PM_{2.5}/PM_{coarse}$	$PM_{2.5}$ absorbance/ PM_{10}	$PM_{2.5}$ absorbance/ PM_{coarse}	PM_{10}/PM_{coarse}
Oslo	2.33	14.84	1.41	7.80	6.97	0.66	4.52	0.10	0.58	5.42
Stockholm County	2.11	21.30	0.95	2.02	10.67	0.49	1.16	0.05	0.10	2.16
Helsinki/Turku	2.11	16.54	1.24	3.18	8.05	0.60	1.61	0.08	0.20	2.61
Copenhagen	1.47	13.21	0.95	2.76	9.53	0.65	1.97	0.07	0.21	2.97
Kaunas	0.77	7.87	0.54	2.11	10.72	0.72	2.87	0.07	0.26	3.87
Manchester	2.75	20.12	1.54	3.58	7.71	0.57	1.34	0.08	0.18	2.34
London/Oxford	3.03	21.74	1.82	4.75	7.54	0.60	1.58	0.08	0.22	2.58
Netherlands/Belgium	1.66	17.73	1.08	3.18	11.33	0.66	1.96	0.06	0.18	2.96
Ruhr_Area	1.59	18.06	1.05	3.16	11.60	0.67	2.01	0.06	0.18	3.01
Munich/Augsburg	1.95	14.49	1.25	3.65	7.79	0.66	2.00	0.09	0.26	3.00
Vorarlberg	1.65	11.87	1.05	3.02	7.43	0.65	1.92	0.09	0.26	2.92
Paris	1.93	15.44	1.20	3.26	8.97	0.64	1.83	0.08	0.21	2.83
Gyor	0.67	7.73	0.49	1.92	11.92	0.74	2.96	0.06	0.25	3.95
Lugano	1.61	13.77	1.14	4.07	8.79	0.72	2.64	0.08	0.30	3.64
Turin	1.59	15.44	1.08	3.50	10.12	0.69	2.28	0.07	0.23	3.28
Rome	2.03	13.95	1.09	2.39	7.05	0.54	1.22	0.08	0.17	2.22
Barcelona	3.06	18.68	1.34	2.42	6.24	0.44	0.78	0.07	0.13	1.78
Catalunya	2.57	16.83	1.15	2.12	7.03	0.45	0.84	0.07	0.12	1.84
Athens	1.61	14.19	0.78	1.54	9.06	0.49	0.98	0.05	0.11	1.98
Heraklion	0.83	10.18	0.32	0.52	12.96	0.39	0.63	0.03	0.05	1.63
Mean	1.87	15.20	1.07	3.05	9.07	0.60	1.86	0.07	0.21	2.85
Mean North	2.01	16.47	1.14	3.94	8.81	0.60	2.32	0.08	0.27	3.29
Mean West/Central	1.76	14.88	1.12	3.27	9.38	0.66	2.11	0.08	0.23	3.11
Mean South	1.84	15.64	1.17	3.41	9.32	0.66	2.05	0.08	0.23	3.05

volatile components than the continuous monitors typically used in networks in Europe (Putaud et al., 2004; Van Dingenen et al., 2004). We conditioned filters at 37% RH before and after weighing following US EPA procedures. Our PM measurements are therefore less affected by particle-bound water than protocols which condition at 50% RH (Van Dingenen et al., 2004). The Harvard impactor is not a European reference sampler, but has been shown to agree well with reference samplers such as the PM_{10} High Volume sampler (Hoek et al., 1997). A limitation of our study is that measurements were conducted in two years and hence some of the variability between areas may be affected by different meteorological conditions. To assess this issue, all PM_{10} and $PM_{2.5}$ annual average concentrations in AIRBASE in the ESCAPE countries from 2009 to 2010 were evaluated. Only stations with more than 75% data capture in both years were included in the analysis. Overall, there was no difference in average concentration between 2009 and 2010. The mean (standard deviation) PM_{10} concentration was 22.4 (5.9) and 22.5 (5.5) $\mu g\ m^{-3}$ in 2009 and 2010 respectively. The mean (standard deviation) $PM_{2.5}$ concentration was 12.6 (3.5) and 13.3 (4.2) $\mu g\ m^{-3}$ in 2009 and 2010 respectively. Our temporal coverage of each site was limited to three 2-week samples. We adjusted the averages however with measurements at a reference site which measured for a whole year, to obtain a valid spatial contrast. Because of our focus on assessing human residential exposures, we did not include remote sites to assess the 'natural' background. Regional background sites were in villages, often near cities. Furthermore, street sites were chosen at the façade of homes, not on the kerbside.

4.1. Concentration contrasts between study areas

Across the study areas the fraction of regional background, urban background and street sites differed somewhat. Therefore, when comparing concentrations between areas, we focus mostly on comparing the urban background levels. The higher PM_{10} concentrations in the Southern and Eastern Europe are consistent with previous studies based upon routine monitoring networks (European Environment Agency (EEA) 2009; Beelen et al., 2009) and specific monitoring campaigns (Van Dingenen et al., 2004; Hoek et al., 1997; Lianou et al., 2011). We also observed some differences with the EEA PM_{10} map, e.g. our PM_{10} concentrations in Kaunas including regional background are substantially higher. This may be due to differences in PM_{10} sampling equipment between the routine networks in different countries, which limits comparison (European Environment Agency (EEA) 2009). The high PM_{10} concentrations in the eastern study areas were due to high $PM_{2.5}$ concentrations (below).

Less information is available from the literature about $PM_{2.5}$ and $PM_{2.5}$ absorbance spatial gradients in Europe. We found patterns for $PM_{2.5}$ and PM_{10}, which broadly agree with a study in 21 areas in Europe, measuring one urban background site per area over an entire year (Götschi et al., 2005; Hazenkamp-von Arx et al., 2004). Both our study and the Hazenkamp-study found high $PM_{2.5}$ concentrations in Turin. High traffic emissions alone are an unlikely explanation of these high concentrations, as $PM_{2.5}$ and $PM_{2.5}$ absorbance concentrations in Turin were substantially higher than those in the larger city of Rome. Turin is located in the Po-valley where specific meteorological conditions in combination with industrial, domestic and traffic emissions may play an important role. Similar patterns across Europe were also found in two smaller studies (Hoek et al., 2002a,b; Puustinen et al., 2007). Concentrations in Eastern Europe have been high in the past because of both industrial emissions and coal combustion (Houthuijs et al., 2001), but have since been reduced significantly (Heinrich et al., 2000). Although we included two moderately sized Eastern European cities, $PM_{2.5}$ and PM_{10} concentrations were still relatively high. This probably reflects the influence of large area sources in the region as absorbance levels (reflecting more local sources, particularly traffic) were relatively low – similar to those found in, for example Munich/Augsburg and Netherlands/Belgium.

The spatial gradient of PM_{coarse} differs from the $PM_{2.5}$ gradient, with high PM_{coarse} concentrations found in Heraklion, Athens and Barcelona. Turin has much lower levels of PM_{coarse}. The pattern is likely explained by the dryer climate in these areas, resulting in

more resuspension of coarse particles. Furthermore, the Mediterranean area is also affected by Sahara dust events (Perez et al., 2008; Zauli Sajani et al., 2011).

4.2. Spatial variability within study areas

Spatial variability within study areas was larger than between areas for $PM_{2.5}$ absorbance and was substantial for all other components. For the ESCAPE project, this is important as the main analyses of associations between air pollution and health will focus on within-study area variability. Overall contrasts (total range/mean) also depend on the distribution of sites, but we can clearly see that (in all areas) the spatial contrasts in $PM_{2.5}$ absorbance and PM_{coarse} are higher than those for $PM_{2.5}$ and PM_{10} (Table 2). The overall within area spatial variability is affected by differences in e.g. the number of street sites. All site types were selected to represent characteristic regional background, urban background or street sites for that area, so the ratio between the site types is more comparable between study areas.

4.3. Street/urban background contrasts

For all components, we found large variation of the street/urban background ratios between study areas with no clear pattern across Europe. These differences in ratios are probably explained by a combination of differences in actual traffic intensity, traffic composition (percentage diesel fuelled vehicles), local climate and meteorological factors (mixing height, temperature), local geography (mountains, lakes) and street configuration (e.g. street canyons and tree-lined streets). Contrasts between background and street sites in Manchester were lower than those in other areas for all components, because many selected street sites were further away from the road than in the other areas. The implication is that the use of exposure indicators, such as proximity to major roads or traffic intensity on nearby roads, in different study areas is associated with different actual contrasts in exposure. This limits comparison of health effect estimates related to major roadway proximity across studies (Jerrett et al., 2005a). A further discussion is found in the companion paper (Cyrys et al., 2012).

As has been reported previously (Janssen et al., 2011), we found that ratios between traffic and urban background sites were consistently lower for $PM_{2.5}$ than for $PM_{2.5}$ absorbance. This is likely due to the high background concentrations of fine particles related to long-range transported air pollution. $PM_{2.5}$ absorbance characterizes local soot emissions, especially from diesel vehicles and other combustion sources such as residential wood combustion. The traffic/urban background ratio for PM_{coarse} was similar to that of absorbance, illustrating the importance of non-tailpipe emissions (Johansson et al., 2009; Gietl and Klemm, 2009). Non-tailpipe emissions are due to resuspended dust from road surface material and wear of brakes, clutch and tyres. In Northern Europe, the sanding of icy roads and the use of studded tyres further increase non-tailpipe emissions. Traffic/urban background ratios were high in all areas, with a tendency towards higher ratios in the Northern European areas (e.g. Stockholm County).

4.4. Correlation between components

We found very high correlations of the spatial variation within areas between $PM_{2.5}$ and PM_{10}, and between NO_2, NO_x & $PM_{2.5}$ absorbance in most of the study areas. The implication is that in studies that assess the relationship between spatial variation of long-term average air pollution and health, the health effects of these components cannot be separated from each other within a single study area. However, as the ratio of these components varied significantly across Europe, within the ESCAPE study we will be able to make use of the difference in composition of the mixture to assess health effects of different components. Furthermore, the correlation between $PM_{2.5}$ and PM_{coarse} was substantially lower in most areas, probably allowing separation of these components. The high correlation between NO_2 and absorbance agrees with results from the TRAPCA study, carried out in 1999–2000 (Lewné et al., 2004). In TRAPCA, the correlation (R^2) between NO_2 and $PM_{2.5}$ absorbance for Munich, Netherlands and Stockholm was 0.76, 0.86 and 0.81 respectively. These correlations remained largely unchanged in ESCAPE, now with R^2 values of 0.87, 0.86 and 0.84 for Munich/Augsburg, Netherlands/Belgium and Stockholm County, respectively.

Correlations (R^2) between annual average concentrations of NO_2 and $PM_{2.5}$ were, however, more variable than reported within TRAPCA. Good correlations (R^2) between NO_2 and $PM_{2.5}$ were previously found in TRAPCA for all three areas 0.71, 0.80 and 0.64 (Lewné et al., 2004), while in ESCAPE the R^2 values for Munich and the Netherlands dropped to 0.29 and 0.57 respectively, while Stockholm remained stable at 0.64. The three study areas were larger in ESCAPE than in TRAPCA, which may affect the comparison, as well as changes over time in fleet composition, traffic intensity and emission rates. Many epidemiological studies on traffic have used NO_2 as a general indicator for traffic-related air pollution (HEI, 2010), but the highly variable correlations and concentration ratios between NO_2 and other metrics suggest that NO_2 might be a better general traffic indicator in some areas (Paris, Barcelona) than in others (Kaunas, Lugano, Vorarlberg).

5. Conclusion

We found clear spatial contrasts between $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10} and PM_{coarse}, both within and between the 20 study areas. While there were large differences in background concentration between areas for all components, within-area contrasts were particularly clear for $PM_{2.5}$ absorbance and PM_{coarse}. Significant differences were found between urban background sites and street sites for most study areas. High street/background ratios for PM_{coarse} specifically indicate the importance of non-tailpipe emissions. The highly standardized measurement of particle concentrations across Europe in ESCAPE will contribute to a consistent assessment of health effects across Europe.

Competing interest

The authors declare they have no competing financial interest.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2012.08.038.

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