

1 Variation of NO₂ and NO_x concentrations between and within

2 36 European study areas: results from the ESCAPE study

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79 **Running head:** Variation of NO₂ and NO_x concentrations between and within 36
80 European study areas

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82 study

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84

85 **Abbreviations:**

86 CV: Coefficient of Variation

87 ESCAPE: European Study of Cohorts for Air Pollution Effects

88 GIS: Geographic information systems

89 LUR: Land Use Regression

90 NO_x: nitrogen oxides [$\mu\text{g}/\text{m}^3$]

91 NO₂: nitrogen dioxide [$\mu\text{g}/\text{m}^3$]

92 NO: nitrogen monoxide [$\mu\text{g}/\text{m}^3$]

93 PM: particulate matter

94 PM_{2.5}: mass concentration of particles less than 2.5 μm in size

95 PM₁₀: mass concentration of particles less than 10 μm in size

96 RB: Regional Background site

97 SOP: Standard Operating Procedure

98 ST: Street site

99 TRAPCA: Traffic- Related Air Pollution and Childhood Asthma

100 UB: Urban Background site

101 ABSTRACT

102 The ESCAPE study (European Study of Cohorts for Air Pollution Effects) investigates
103 long-term effects on human health of exposure to air pollution in Europe. Various health
104 endpoints are analysed by using prospective cohort studies in the study areas. This paper
105 documents the spatial variation of measured NO₂ and NO_x concentrations between and
106 within 36 study areas across Europe.

107 In 36 study areas NO₂ and NO_x were measured using standardized methods between
108 October 2008 and April 2011. In each study area 14 to 80 sites were selected, which
109 represented a wide range of regional, urban and nearby traffic related pollution contrast.
110 The measurements were conducted for two weeks per site in three different seasons,
111 using Ogawa badges. Results for each site were adjusted for temporal variation using
112 data obtained from a routine monitor background site, which operated continuously, and
113 averaged.

114 Substantial spatial variability was found in NO₂ and NO_x concentrations between and
115 within study areas. Analysis of variance showed that 40% of the overall NO₂ variance is
116 attributable to the variability between the study areas and 60% is caused by the
117 variability within the study areas. The corresponding values for NO_x are 30% (between
118 the study areas) and 70% (within the study areas). The within-area spatial variability
119 was mostly determined by the differences between traffic and urban background
120 concentrations. The traffic/urban background concentration ratio varied between 1.09
121 and 3.16 across Europe. The NO₂ / NO_x ratio varied between 0.47 (Verona) and 0.72
122 (Heraklion) across study areas. In study areas in southern Europe the highest median
123 concentrations were observed (Barcelona: NO₂ 55 µg/m³), followed by densely
124 populated areas in Western Europe (Ruhr area, The Netherlands). The lowest
125 concentrations were observed in all areas in Northern Europe (e.g. Umeå: NO₂ 7
126 µg/m³).

127 In conclusion, we found significant contrast in annual average NO₂ and NO_x
128 concentration between and especially within 36 study areas across Europe.
129 Epidemiological studies should therefore characterize intra-urban contrasts. The use of
130 traffic indicators such as “living close to major road” as an exposure variable in
131 epidemiological studies results in different actual NO₂ contrasts.

132 **1. Introduction**

133 There is now increasing evidence from epidemiological studies that exposure to ambient
134 air pollution is associated with adverse health effects (Brunekreef and Holgate, 2002;
135 Heinrich et al., 2004; Pope and Dockery, 2006, WHO, 2006; R ckerl et al., 2011).

136 Adverse effects include pre-mature mortality and morbidity from cardiovascular and
137 respiratory causes. Plausible mechanisms for these associations based upon
138 experimental studies have been proposed, including particularly oxidative stress
139 (Brunekreef and Holgate, 2002; Brook et al., 2010; R ckerl et al., 2011). Most studies
140 from the USA have focussed on PM₁₀ and PM_{2.5} (Brook et al., 2010; Pope and Dockery,
141 2006). In European studies, several studies have also reported significant associations
142 between adverse health effects and NO₂ or NO_x concentrations (Brunekreef, 2007). In
143 these studies air pollution exposure was assessed at the residential address using
144 dispersion models, land use regression models and traffic indicator variables. Other
145 epidemiological studies on long-term exposure to NO₂ and other air pollutants
146 compared the health status of populations using the contrast in city-average air pollution
147 levels between different areas (e.g. Pope et al., 2002; Laden et al., 2006; Sunyer et al.,
148 2006; G tschi et al., 2008). These studies generally assigned one overall average
149 concentrations to all subjects living in each city. For NO₂ this likely results in
150 significant misclassification as high spatial variability within urban areas has been
151 documented previously for nitrogen dioxide (NO₂) in specifically designed studies
152 (Lebret et al., 2000; Monn, 2001; Lewne et al., 2004). There is a substantial number of
153 studies that have used traffic indicators as exposure variables, including distance to a
154 major road, traffic intensity on the nearest road (HEI, 2010). A major limitation of these
155 traffic indicators is that their value in characterizing actual air pollution exposure
156 contrasts may differ between study areas (Jerrett et al., 2005). Some studies have made
157 use of the spatial variation of air pollution within metropolitan areas (Gauderman et al.,
158 2005; Gehring et al., 2006; Morgenstern et al., 2008, Jacquemin et al., 2009; Modig et
159 al., 2009). These within-city studies often characterized air pollution with the
160 concentration of NO₂ and NO_x obtained from either spatially dense monitoring
161 networks, land use regression models based upon such networks or dispersion models
162 (Jerrett et al., 2005; Hoek et al., 2008; Modig et al., 2009; Levy et al., 2010).

163 In 1999, the European Commission established limit values for NO₂, NO_x, PM₁₀ and
164 some other air pollutants in the Air Quality Daughter Directive 1999/30/EC (EC, 1996),

165 which was replaced 2008 by the new Directive 2008/50/EC on ambient air quality and
166 cleaner air for Europe (EC, 2008). The existing air quality guidelines for NO₂ and PM₁₀
167 are currently being exceeded at many locations throughout Europe and Germany
168 (Giannouli et al., 2011; European Environment Agency 2006; Airbase 2007; Velders
169 and Diederer, 2009). There is therefore substantial interest at the EU policy level in the
170 health effects of current air pollution levels including NO₂, focussing especially on
171 European studies.

172 A comparison of NO₂ concentrations measured either in study specific monitoring
173 programme (Hazenkamp-von Arx et al., 2004) or in routine monitoring networks across
174 Europe (e.g. Airbase data used in Beelen et al., 2009) showed significant contrast across
175 Europe. The concentrations were generally lowest in Northern Europe and highest in the
176 major cities and Southern Europe.

177 NO₂ is often used as an indicator of the complex mixture of traffic-related air pollution
178 containing also fine and ultrafine particles. The ratio of NO₂ to other components e.g.
179 soot in emissions of motorized road traffic has changed in the last decade (Williams and
180 Carslaw, 2011). Specifically, the fraction of primary NO₂ emissions has increased.

181 In 2008 we embarked upon a European-wide study of long-term air pollution exposure
182 health effects. The ESCAPE study - **E**uropean **S**tudy of **C**ohorts for **A**ir **P**ollution
183 **E**ffects - assesses exposure-response relationships between long-term exposures to
184 ambient air pollution using prospective cohort studies in 15 different European
185 countries (<http://www.escapeproject.eu>).

186 As a key interest of epidemiological long-term studies is in within-urban variation of air
187 pollution and the most routine monitoring networks are not sufficiently dense to
188 characterize intra-urban concentration gradients, we decided to carry out study specific
189 monitoring, which was independent of routine monitoring networks. In all 36 study
190 areas, NO₂ and NO_x were measured with passive samplers. In 20 of these areas, we also
191 measured PM_{2.5}, PM_{2.5} absorbance, PM₁₀, and PM_{coarse} (Eeftens et al., submitted). The
192 measured average concentrations were combined with geographic predictors to develop
193 land use regression (LUR) models (Jerrett et al., 2005; Hoek et al. 2008). In all study
194 centres a common protocol was used to ensure high standardization of all procedures
195 across the 36 European study areas The standardization of the measurements and the
196 selection of the locations using a common protocol across a wide range of European

197 settings (i.e. >1000 monitoring sites across Europe) is one of the major strength of this
198 study.

199 The aim of this paper is to assess the variation of measured NO₂ and NO_x
200 concentrations between and within 36 European study areas. We further assessed the
201 variability across Europe of the increase of NO₂ and NO_x concentrations at traffic
202 stations versus urban background stations and the NO₂ / NO_x ratio. The companion
203 paper focuses on the PM measurement (Eeftens et al., submitted).

204

205 **2. Methods**

206 *2.1 ESCAPE exposure assessment*

207 The objective of the ESCAPE study is to investigate relationships of long-term air
208 pollution exposure and health (for further details please refer to Eeftens et al.,
209 submitted).

210 As indicated in Figure 1 in 20 study areas both particulate matter (PM_{2.5}, PM_{2.5}
211 absorbance, PM₁₀, and PM_{coarse}) and NO₂/NO_x were measured (NO_x + PM study areas)
212 whereas in 16 remaining study areas NO₂/NO_x only was measured (NO_x only study
213 areas). In all 36 study areas, NO₂ and NO_x were measured at 40 locations spread over
214 the whole study area; whereas the PM measurements were performed at 20 sites
215 selected out of 40 sites in total (Eeftens et al., submitted).

216 Exposure assessment was highly standardized within ESCAPE. All local centres used
217 the same passive sampler, namely the Ogawa badge (<http://www.ogawusa.com>). All
218 badges were prepared and analyzed by one central laboratory at Institute for Risk
219 Assessment Sciences (IRAS), Utrecht, using the ESCAPE Standard Operating
220 Procedure (SOP) available from the ESCAPE project website
221 (<http://www.escapeproject.eu/manuals/>). The SOP defined the field measurement
222 procedures including Quality Assurance and Quality Control and calculations. A study
223 manual provided details on site selection. A workshop was organized for all field
224 workers to further harmonize procedures. Site selection and calculations in a centrally
225 provided Excel file were centrally checked.

226

227

228 2.2 *Study area and site selection*

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

240 The measurement sites were classified as regional background, urban background and
241 street sites. A street site was considered a site in a major road carrying at least 10,000
242 vehicles per day. Measurements were typically made at the façade of the homes, as we
243 were interested in characterizing residential exposures. We hence did not measure at the
244 kerbside. An urban background site was defined as a site with fewer than 3,000 vehicles
245 per day passing within a 50m radius. The distinction between regional and urban
246 background was not strictly defined in the study manual, but typically involved
247 measurements in the smaller towns of the cohort.

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

257 An overview of the characteristics of study areas is presented in Table 1 and in more
258 detail in Online Supplement A.

259

260 2.3 *Sampling and analysis*

261 The standard operating procedure SOP is available from the ESCAPE project website
262 (<http://www.escapeproject.eu/manuals/>). NO₂ and NO_x were measured using Ogawa
263 diffusion badges. A detailed description of sampling and analysis has been published
264 previously (van Roosbroeck et al., 2006). The sampler contains two collection filters
265 that are coated with a reactive chemical, one for sampling NO₂ and one for sampling
266 NO_x (NO₂ plus NO). NO is calculated by subtraction. Ogawa badges were transported
267 from the central laboratory in individual plastic bags and cooled during transport and
268 storage. The analysis is spectrophotometrically based upon the Saltzman method (van
269 Roosbroeck et al., 2006). The preparation and chemical analysis of all Ogawa badges
270 was performed centrally in one laboratory (IRAS, Utrecht). From each batch of 40
271 filters obtained from the manufacturer, four filters were kept at IRAS laboratory as lab
272 blanks. These four lab blanks were analyzed on the same day as the exposed filters and
273 their results were subtracted from results of filters in the same batch.

274 To establish the agreement with the chemiluminescence method being the European
275 reference method, we compared the Ogawa diffusion badges during every 2-week
276 sampling period with a chemiluminescence monitor in each study area. The Ogawa
277 samplers were located in direct vicinity to the chemiluminescence monitor. The results
278 will be published soon in a separate publication.

279 2.4 *Sampling campaign*

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

288 In the NO_x only study areas the two week measurements were done almost
289 simultaneously at all sites – start day of all measurements was within a time period of
290 maximum three days. In contrast, due to equipment limitation for PM sampling, the
291 measurements in the NO_x + PM study areas were performed simultaneously only at 5

292 sites and one continuous reference site. Thus, for completion of one measurement
293 period four rounds of two week measurements were necessary. Each group of 5 sites
294 included different site types, e.g. regional, urban background and traffic. NO₂ and NO_x
295 in the NO_x + PM study areas was measured exactly at the same time as the PM
296 measurements were performed.

297 The reference site was used for sampling during all sampling periods, covering a full
298 year. This reference site was located at an urban or rural background location and
299 measured over the whole year. The site was used to adjust for temporal variation. If a
300 PM and/or NO_x measurement failed, the measurement was repeated, preferably in the
301 same season.

302 2.5 *Adjustment for temporal variation*

303 Air pollutants have a substantial temporal variation. Therefore adjustment for temporal
304 variability is essential if annual means of the measured NO₂ and NO_x concentrations are
305 calculated, particularly for the NO_x + PM areas with non-simultaneous measurements.
306 For details we refer to the PM companion paper (Eeftens et al., submitted). Briefly, the
307 difference between the concentration for a specific two week sampling period and the
308 annual average at the reference site was subtracted from each measurement. This
309 procedure is based upon procedures developed previously within the TRAPCA (Traffic-
310 Related Air Pollution and Childhood Asthma) study (Hoek et al., 2002). The annual
311 average of NO₂ and NO_x is calculated from the ESCAPE reference site for the NO_x +
312 PM areas. For the NO_x only sites, an urban or rural background station routine
313 monitoring data was used. We did not require a year-long reference campaign, because
314 measurements were already made simultaneous and only occasional missing data could
315 lead to bias in the spatial comparison of calculated averages. In Umeå where no
316 appropriate routine monitoring site was available, adjustment was made using the
317 average of the three sampling campaigns of all sites with complete data. This approach
318 only takes care of bias due to missing data and does not scale the data to an annual
319 average.

320 In the following this paper focuses on adjusted annual average NO₂ and NO_x
321 concentration. We report concentrations in µg/m³. This has implications e.g. for
322 reported NO₂/NO_x ratio. In case of equal NO₂ and NO concentrations in ppb, the NO₂/
323 NO_x ratio is 0.50 expressed in ppb and 0.60 in µg/m³. If the NO₂ is twice the NO
324 concentration in ppb, the NO₂/NO_x ratio is 0.67 expressed in ppb and 0.75 in µg/m³.

325 2.6 *Quality control*

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

335 2.7 *Data analysis*

336 Locally calculated adjusted annual averages were gathered centrally and their range and
337 distribution were calculated, and stratified by site type. To quantify the amount of
338 spatial variation relative to the background level, the interquartile range and total range
339 (maximum-minimum) were calculated as a percentage of the mean. For each study area,
340 we used ANOVA (SAS 9.2, PROC GLM) to test for significant differences between
341 urban background and street sites and (where applicable) between urban and regional
342 background sites. We also tested if urban background levels differed significantly
343 between study areas. The same analyses were performed for street sites and regional
344 background sites (where applicable), again using SAS 9.2, PROC GLM. Correlation R²-
345 values between components were determined using SAS 9.2, PROC REG. Percentages
346 of between and within area variance were determined using analysis of variance with
347 PROC VARCOMP, METHOD=reml.

348

349 **3. Results**

350 ESCAPE included study areas across Europe that were of substantially different size
351 (Figure 1, Table 1 and Online Supplement A). Please note that the study areas in Table
352 1 are sorted from the North to the South. We kept this order in all following tables and
353 figures. The size of the areas is given by the distribution of the included cohorts. Some
354 areas were restricted to a single city (e.g. Rome, Grenoble, Erfurt) or a metropolitan
355 area including more rural / suburban areas (Stockholm country, Athens). The study also
356 included large study areas such as the Ruhr area, Catalunya and the Netherlands /

357 Belgium. The included main cities were also of very different size, ranging from Umeå
358 (~100,000 inhabitants) to the largest European metropolitan areas such as Paris and
359 London with several million of inhabitants. Due to the differences between the study
360 areas the number of regional, urban background and street sites in each study area is
361 also different.

362 3.1 *Quality control*

363 Detection limits were low for NO₂ for all study areas and very few samples were below
364 the limit of detection (Table 2). Detection limits were slightly higher for NO_x but in
365 nearly all study areas very few samples were below the limit of detection. Only in
366 Turin, Catalunya and Albacete a sizable fraction was below the detection limit. This
367 was partly due to unexplained outliers which were all included in the detection limit
368 calculations presented in Table 2. Samples below the detection limit were retained at
369 their original value.

370 Precision expressed as the Coefficient of Variation (CV) calculated from field
371 duplicates (Eeftens et al., submitted) was lower than 10% in 31 areas for NO₂ and in 34
372 areas for NO_x (out of 36). The average CV was 9.2% for NO₂ and 6.1 for NO_x,
373 respectively. The areas with the largest CV (especially for NO₂) were Grenoble, Oslo,
374 Granada and Huelva. In Granada only three and in Huelva two duplicates were
375 obtained. Precision was in 24 study areas poorer for the NO₂ measurements compared to
376 NO_x, but the difference was mostly only minor.

377 3.2 *Spatial contrast of average NO₂ and NO_x concentrations*

378 Unadjusted average concentrations and average concentrations adjusted for temporal
379 variation were very highly correlated (Online Supplement B, Table B1). Squared
380 correlations (R^2) were generally above 0.95, indicating that temporal adjustment
381 resulted only in small changes to the calculated average concentrations. For the NO_x
382 only areas, this reflects that sampling was performed simultaneously and that there were
383 few measurements that failed due to stolen badges or sampling and analysis errors. For
384 sites with three valid observations, the adjustment involves the same scaling to an
385 annual average. For the sites with less than three a different scaling may occur, e.g. if
386 the winter measurement has failed, the adjustment to the average is typically larger than
387 for the sites with valid measurements in the three seasons. For the NO_x + PM study
388 areas, where measurements were not conducted simultaneously, measurements of two-

389 week samples in three seasons were apparently sufficient to obtain a fairly stable
390 estimate of spatial contrasts, in agreement with findings of the TRAPCA study using the
391 same approach (Lewne, 2004).

392 The distribution of adjusted annual average concentrations for NO₂ and NO_x is shown in
393 Figures 2a and 2b as well as in Online Supplement B, Tables B2 and B3.

394 3.3 *Spatial variability within study areas*

395 In many study areas the contrast of individual averages within the study area was as
396 large as the contrast in median concentration across the study areas (Figures 2a and 2b).
397 Substantial spatial variability was found for NO₂ and NO_x concentrations within study
398 areas (refer also to Online Supplement B, Tables B2 and B3). The average range for
399 NO₂ (difference between the highest and the lowest annual average in one specific study
400 area) was 54 µg/m³. The lowest range was observed in Kaunas, Gyor and Bradford NO_x
401 (28 µg/m³) as well as in Heraklion (29 µg/m³). The largest spatial variability for NO₂
402 was found in Catalunya (97 µg/m³) followed by London and Barcelona (95 µg/m³). The
403 average range for NO_x was 136 µg/m³. The lowest NO_x range was measured again in
404 Heraklion (44 µg/m³) followed by Huelva (58 µg/m³) and Munich (72 µg/m³). The
405 largest spatial variability for NO_x was observed in Marseille (254 µg/m³) followed by
406 Verona (251 µg/m³), London (239 µg/m³) and Paris (236 µg/m³).

407 The results of a restricted maximum-likelihood variance component analysis (SAS
408 PROC VARCOMP, METHOD=reml) show that the within study areas variance of NO₂
409 (60.1% of the total variance) is considerable larger than the variance attributable to the
410 differences between the study areas (39.9% of the total variance). The difference for
411 NO_x is even more pronounced; variance component for within study area variability is
412 1005.5 (70% of the total variance), and for between study variability 430.2 (30% of the
413 total variance).

414 The type of the measurement sites (regional background, urban background and street
415 sites) plays also an important role in the overall variability of NO₂ and NO_x. After
416 including the variable “type of the measurement site” to the variance analysis the results
417 show that 30.4% of the overall NO₂ variance is attributable to the variability between
418 the study areas, 37.4% of the variability is caused by different site type and 32.3% could
419 be traced to the variability within the site types. The corresponding values for NO_x are

420 23.4% (variability between the study areas), 36.5% (variability because of different site
421 type) and 40.1% (variability within the site types).

422 The distribution of annual averages of NO₂ and NO_x concentrations by site type, for
423 each study area is shown in Figures 3 and 4, respectively. The NO₂/NO_x concentrations
424 at urban background sites are in general lower than at street locations. The range of
425 NO₂/NO_x concentrations is lowest for regional background sites, followed by urban
426 background sites, whereas the largest variability across the sites was observed for street
427 sites.

428 Table 3 presents the ratio of concentrations measured at regional versus urban
429 background and the ratio of traffic to urban background sites for NO₂ and NO_x,
430 respectively. In almost all study areas the differences between the site types were
431 statistically significant (p<0.05 level).

432 In all study areas, the concentrations at traffic stations were higher than at urban
433 background stations; the street site vs. urban background site ratio was 1.63 for NO₂ and
434 1.93 for NO_x on average for all sites over all study areas. The ST / UB ratio ranged
435 substantially from 1.09 to 3.16 for NO₂. High ratios were found especially in Umeå,
436 Oslo, Mideast Spain and Granada. In all areas the NO_x ST / UB ratio was larger than for
437 NO₂. The NO_x ST / UB ratio ranges from 1.14 in Lugano to 4.24 in Umeå and 2.77 in
438 Paris.

439 NO₂ concentrations were lower at regional background sites compared to urban
440 background concentrations; the mean ratio for all sites over all study areas was 0.63 for
441 NO₂ and 0.60 for NO_x, respectively. However, the RB / UB concentration ratio varied
442 substantially across study areas, from 0.24 in London to 0.91 in Gyor (Table 3). Low
443 ratios were found in London, Umeå and Mid-East Spain. The highest ratios were found
444 in the Ruhr area and Gyor. The pattern for NO_x was similar.

445 3.4 Spatial variability across Europe

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

449 Substantial differences in annual average NO₂ and NO_x concentration across Europe
450 were found (Online Supplement B, Tables B2 and B3). The lowest annual average NO₂
451 concentration was found in Umeå, a medium sized city in Northern Sweden.

452 Concentrations in the large North-European cities (Helsinki, Stockholm, Oslo, and
453 Copenhagen) were relatively low, but similar to the smaller cities in southern
454 (Heraklion, Crete) and central Europe (Erfurt, Gyor, Kaunas). The highest
455 concentrations were measured in the Mediterranean area, especially Barcelona and
456 Turin. The pattern for annual NO₂ levels measured on street locations follows more or
457 less the pattern observed for the urban background locations, except for Paris, Lyon and
458 Granada. The largest differences between the NO₂ concentrations measured on street
459 sites and urban background sites were observed for Paris (31 µg/m³) and Mediterranean
460 areas such as Granada, Turin, Lyon, Barcelona and Catalunya (>31 µg/m³), whereas the
461 lowest difference between the two site types were detected for Lugano, Kaunas and
462 Bradford.

463 The pattern for NO_x was similar to that of NO₂.

464 3.5 NO₂ – NO_x relationship

465 In all study areas a high correlation between NO₂ and NO_x was found (Table 4). Table 4
466 presents also the average NO₂ to NO_x ratios measured in rural, urban background and
467 street sites in each study area. The average NO₂ to NO_x ratio ranged from 0.51 to 0.72
468 for urban background sites in the different study areas. The lowest ratios were found in
469 Verona, Geneva and Basque Country. The highest ratio was found in Copenhagen. At
470 traffic stations the NO₂ to NO_x ratio ranged from 0.42 to 0.73, with the highest value
471 found in Heraklion. Overall there was no clear North-South gradient in the ratio. Mean
472 ratios at urban background sites were 0.62, 0.63 and 0.61 for the Northern-European,
473 central-western and southern European areas, respectively.

474 In most areas, the NO₂ to NO_x ratio was smaller at street sites than at urban background
475 sites. On average, the ratio was 0.69, 0.62 and 0.54 for regional background, urban
476 background and street sites respectively. Without Mid-East Spain the ratio was 0.65 for
477 regional background. The difference in ratio varied significantly across Europe, e.g. in
478 Copenhagen, Paris and the Netherlands the ratio was much lower at street sites than at
479 urban background sites, whereas in e.g. Basel, Geneva there was little difference,
480 possibly reflecting differences in primary NO₂ emissions across Europe.

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483

484 4. Discussion

485 Overall we found significant contrast in annual average NO₂ and NO_x concentration
486 between and especially within 36 study areas across Europe. NO₂ concentrations at
487 street locations were on average between 1.22 and 3.6 times higher than at urban
488 background stations. The NO₂ / NO_x ratio varied between 0.47 and 0.72 across study
489 areas. Concentrations were generally lower in Northern than in Southern Europe.

490 4.1 ST / UB contrast

491 The ST / UB contrast observed in our study for NO₂ and NO_x concentrations was higher
492 and more consistent than for the particle metrics across all study areas (Eeftens et al.,
493 submitted). For PM_{2.5}, PM_{2.5} absorbance (soot), PM₁₀ and PM_{coarse} we found ratios of
494 1.14, 1.38, 1.23 and 1.42 respectively. This was not due to differences in the included
495 study areas, as the NO₂ and NO_x ST/UB ratio in the areas where PM was measured were
496 1.54 and 1.80 respectively. Such pronounced contrast between traffic and background
497 environments was observed also for other traffic related pollutants, including ultrafine
498 particles (Cyrys et al., 2003a; Tuch et al., 2006; Puustinen et al., 2007; Cyrys et al.,
499 2008; Hoek et al., 2011). Furthermore, the spatial variation of NO₂ is more comparable
500 to the spatial variation of other traffic related primary pollutants (soot) and less to those
501 of partly secondary pollutants such as PM_{2.5} or PM₁₀. As shown in the companion paper
502 for PM_{2.5}, PM₁₀ and PM_{2.5} absorbance (Eeftens et al., submitted) the R² between NO₂
503 and PM_{2.5} absorbance (as indicator for traffic related soot) is generally high: 0.80 (range
504 0.55-0.91), while the correlation between NO₂ and PM_{2.5} is much lower: R² = 0.50
505 (range 0.02-0.90). The contrast between regional and urban background sites was more
506 consistent and higher than observed in the companion paper for PM_{2.5}, PM₁₀ and
507 absorbance in almost all areas (Eeftens et al., submitted). Also the temporal variation of
508 NO and NO₂ concentrations is often strongly correlated with those of other traffic
509 related air pollutants, such as CO, Black Carbon and ultrafine particles (Cyrys et al.,
510 2003b; Hagler et al., 2009; Sabaliauskas et al., 2012). Moreover, nitrogen oxides have
511 been found to be the most important predictor variables for ultrafine particles in the
512 urban air (Paatero et al., 2005). It reveals the role of nitrogen oxides as marker for
513 traffic related air pollutants. Due to the close temporal correlation with other
514 combustion related pollutants NO₂ has been often used in epidemiological studies as a
515 marker for traffic exhaust. Its concentration is measured easily and relatively cheaply,

516 but one should keep in mind that it serves only as a surrogate for a set of sources and
517 resulting mixture of air pollutants.

518 4.2 *Variability within study areas*

519 The observation of high within study area variability of NO₂ concentrations compared
520 to between study area variability agrees with previous observations in four urban areas
521 cities in western and central Europe (Lebret et al., 2000), 16 cities of the European
522 Community Respiratory Health Survey (ECRHS) (Götschi et al., 2008) and 8 areas of
523 the longitudinal Swiss cohort study on air pollution and lung and heart disease in adults
524 in Switzerland (SAPALDIA) (Liu et al., 2012).

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

546 The observed pronounced within-area spatial contrast for NO₂ and NO_x is in a large part
547 attributable to the differences between street and urban background sites. The ratio of
548 the ST / UB concentrations varied widely across the study areas. The further implication

549 of this variability for epidemiological studies is that the use of traffic indicator variables
550 such as living close to a major road presents a different contrast in actual air pollution
551 exposure in different cities. This likely contributes to the observed heterogeneity of
552 estimated health effects in studies using traffic indicators (Vardoulakis et al., 2003;
553 Heinrich et al., 2004).

554 The relatively high ST / UB ratios in Northern European cities, particularly in Umeå,
555 are likely due to the low urban background concentrations in these countries. The
556 variability in ratios is probably explained by a combination of the following factors:
557 traffic intensity, fraction of heavy duty vehicles, emission factor of the car fleet related
558 to e.g. age and fuel type of cars and possibly street configuration. In interpreting the
559 (often modestly increased) magnitude of the ratio of concentrations measured at street
560 sites and urban background sites, it has to be taken into account that measurements were
561 not made at kerbsides, as is often the case in routine monitoring network street sites.
562 The average ratio further reflects an average street, not the busiest street in the area.
563 Figures 3 and 4 illustrate that there is significant variability across street sites.

564 The RB / UB ratios for NO_x were more consistent and higher than observed in the
565 companion paper for PM_{2.5}, PM₁₀ and absorbance in almost all areas (Eeftens et al.,
566 submitted). PM₁₀ and PM_{2.5} have a high regional background concentration and local
567 sources increase concentrations only modestly, however absorbance as marker for soot
568 is strongly affected by local sources including traffic. The higher contrast for NO₂
569 compared to soot could be related to the differences of atmospheric lifetime for NO_x and
570 soot. Soot is enriched in submicrometer particle fraction having a longer atmospheric
571 lifetime and could be accumulated there. Higher emissions of NO_x from road traffic or
572 other combustion sources compared to the background could also play a role. The
573 slightly higher ratio of traffic/urban background for NO₂ compared to soot argues in
574 favour of this explanation, though this pattern is less clear than the urban-rural contrast.
575 The use of after treatment technology that traps soot from diesel vehicles but increases
576 NO₂ because of catalytic oxidation of collected soot may have contributed to this (Grice,
577 2009; Williams and Carslaw, 2011).

578 At 323 sites (out of 1485) in 31 study areas (out of 36), the annual average NO₂
579 concentration measured at a site exceeded 40 µg/m³, the EU air quality guideline for the
580 annual average (in Catalunya at 50 sites, Barcelona at 33 sites, and Turin at 31 sites).
581 The comparison with the annual average guideline is limited because we sampled only

582 during three 2-week periods. However, a reference site was used to adjust all measured
583 concentrations to an annual average concentration.

584 4.3 *Contrasts across Europe*

585 The general pattern across Europe agrees with previous studies based upon study
586 specific monitoring programme (Hazenkamp-von Arx et al., 2004) or upon routine
587 monitoring stations (Beelen et al. 2009). The high NO₂ concentrations in Southern
588 European countries could be due to high traffic intensity, a large fraction of diesel-
589 powered vehicles and a higher conversion of NO to NO₂ because of relatively high
590 temperatures and ozone concentrations. Alternatively, the more densely built
591 Meditererean cities could lead to higher concentrations. High air pollution
592 concentrations in Turin and the Po-valley in general have been reported before
593 (Hazenkamp-von Arx et al., 2004). Turin is located mainly on the left bank of the Po
594 River and surrounded by the Alpine arch. It favors thermal inversions, characterized by
595 low surface wind speeds (stagnation of the air) and trapping air pollutants in the lower
596 layers of the atmosphere. In general these unfavorable meteorological conditions are
597 characteristic for the whole Po Valley al all. The combination of the stagnant air
598 conditions with high emissions due to heavy traffic and high population density is the
599 cause of a large number of days exceeding air quality standards and some very strong
600 pollution episodes in this area (Minguzzi et al., 2005; de Meij et al., 2009; Lonati et al.,
601 2010; Fattore et al., 2011). Some of the variability across the study areas is probably
602 also due to weather variability between the two years.

603 Similar North-South gradient across Europe was also observed for PM_{2.5} and PM₁₀
604 (Hazenkamp-von Arx et al., 2004; Eeftens et al., submitted). However in the NO₂
605 pattern a city size is also clearly visible. NO₂ concentrations in the moderately sized
606 cities of Heraklion, Crete, Gyor and Kaunas were similar to those in the Northern large
607 cities of Helsinki, Stockholm and Oslo, whereas the PM₁₀ and PM_{2.5} concentrations
608 were substantially lower in the Northern cities. This reflects the larger impact of long
609 range transported secondary aerosol on the PM_{2.5}/PM₁₀ levels and, in contrast, the
610 predominant role of local source emissions on the NO_x concentrations. Another example
611 is that NO₂ concentrations in London exceeded concentrations in the Netherlands and
612 the Ruhr area whereas the PM_{2.5} and PM₁₀ concentrations showed the opposite pattern.

613 By the comparison of the NO₂ and NO_x concentrations across the study areas one
614 should keep in mind that the study areas are of substantially different size and type

615 (which is given by the different distribution of the existing study cohorts). Some study
616 areas consist of one single city, some include more rural and/or suburban locations and
617 some others cover whole regions (Catalunya) or even countries (the Netherlands /
618 Belgium). Also the included cities differ regarding their size from small cities like
619 Umeå to the largest European metropolitan areas such as Paris or London. Because of
620 those differences also the number of regional, urban background and street sites in each
621 study area is different.

622 4.4 *NO₂ to NO_x ratio*

623 The major sources of NO₂ and NO_x are motorized road traffic, industry, shipping and
624 heating. Nitrogen oxides are emitted as NO and NO₂. In the atmosphere NO reacts with
625 ozone to form secondary NO₂. The observed NO₂ to NO_x ratio varied between study
626 areas and site types within study areas. The typically higher ratios observed at urban
627 background sites reflect enhanced transformation of NO to NO₂ through equilibrium
628 reactions with ozone at those sites. The relatively modest difference between traffic and
629 urban background sites may reflect the increase in primary NO₂ emissions close to street
630 sites. Recent studies reported evidence of increasing NO₂ to NO_x ratio from road traffic
631 emissions due to an increase in primary NO₂ emissions observed in several urban areas
632 in Europe. Primary NO₂ emissions have therefore gained importance compared to the
633 ozone / NO_x equilibrium (Keuken et al., 2009; Mavroidis and Chaloulakou, 2011). This
634 increase have been attributed to the more common use of diesel-fuelled vehicles, since
635 they emit a higher fraction of NO₂ compared to gasoline-fuelled vehicles (Grice et al.,
636 2009; Anttila et al., 2011; Carslaw et al., 2011). In addition, the after-treatment devices
637 (such as oxidation catalyst) implemented for reducing particulate matter emissions by
638 diesel vehicles contribute to increasing fraction of primary NO₂ in NO_x (Williams and
639 Carslaw, 2011; Mavroidis and Chaloulakou, 2011). For diesel-fuelled vehicles equipped
640 with catalytic diesel particulate filters, primary NO₂ fractions of around 40 - 50% are
641 reported (Carslaw et al., 2007).

642 Significant variability of the fraction of primary NO₂ in traffic emissions across Europe
643 was reported in the studies, but there is less information on NO₂ / NO_x concentration
644 ratios.

645 Due to higher ozone levels and a higher percentage of diesel vehicles in the car fleet, we
646 expected higher NO₂ to NO_x ratio in southern Europe. We did not find this, possibly
647 because of more street canyons within Southern European cities, in which due to poor

648 dispersion ozone may become a limiting factor (Vardoulakis et al. 2003). For Athens, it
649 was suggested that the fraction of primary NO₂ is not increasing (as is the case for other
650 urban areas in Europe) as diesel passenger cars are not allowed there and particle after-
651 treatment technologies are not applied in Greece (Mavroidis and Chaloulakou, 2011).

652

653 **5. Conclusion**

654 We found significant contrast in annual average NO₂ and NO_x concentration between
655 and especially within 36 study areas across Europe. NO₂ concentrations at traffic
656 stations were on average between 1.22 and 3.6 times higher than at urban background
657 stations. The NO₂ / NO_x ratio varied between 0.47 and 0.72 across study areas.

658 Concentrations were generally lower in Northern than in Southern Europe, but a clear
659 impact of city size was also found.

660 Epidemiological studies should therefore characterize intra-urban contrasts. The use of
661 traffic indicators such as “living close to major road” as an exposure variable in
662 epidemiological studies results in different actual NO₂ contrasts.

663

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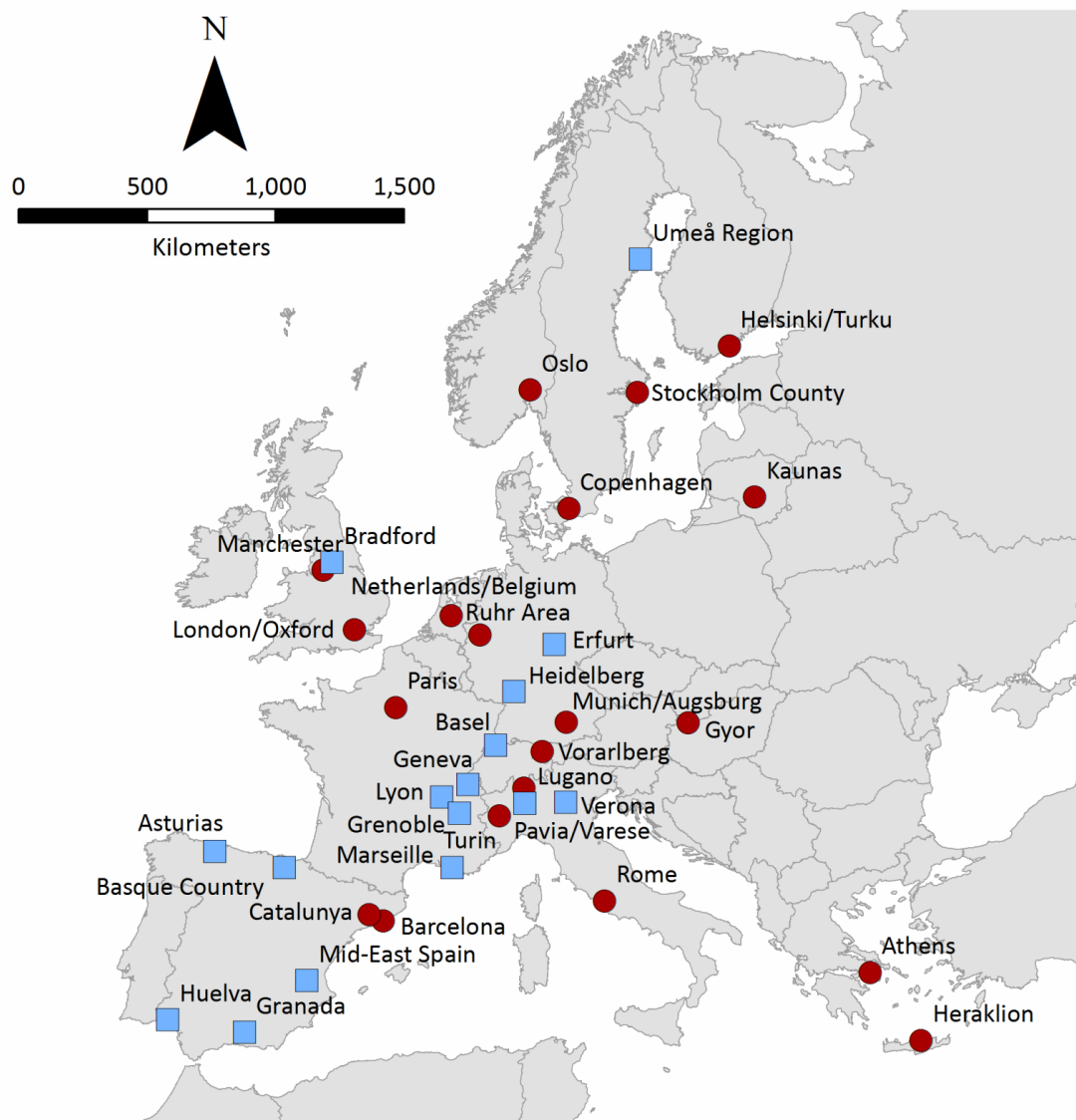
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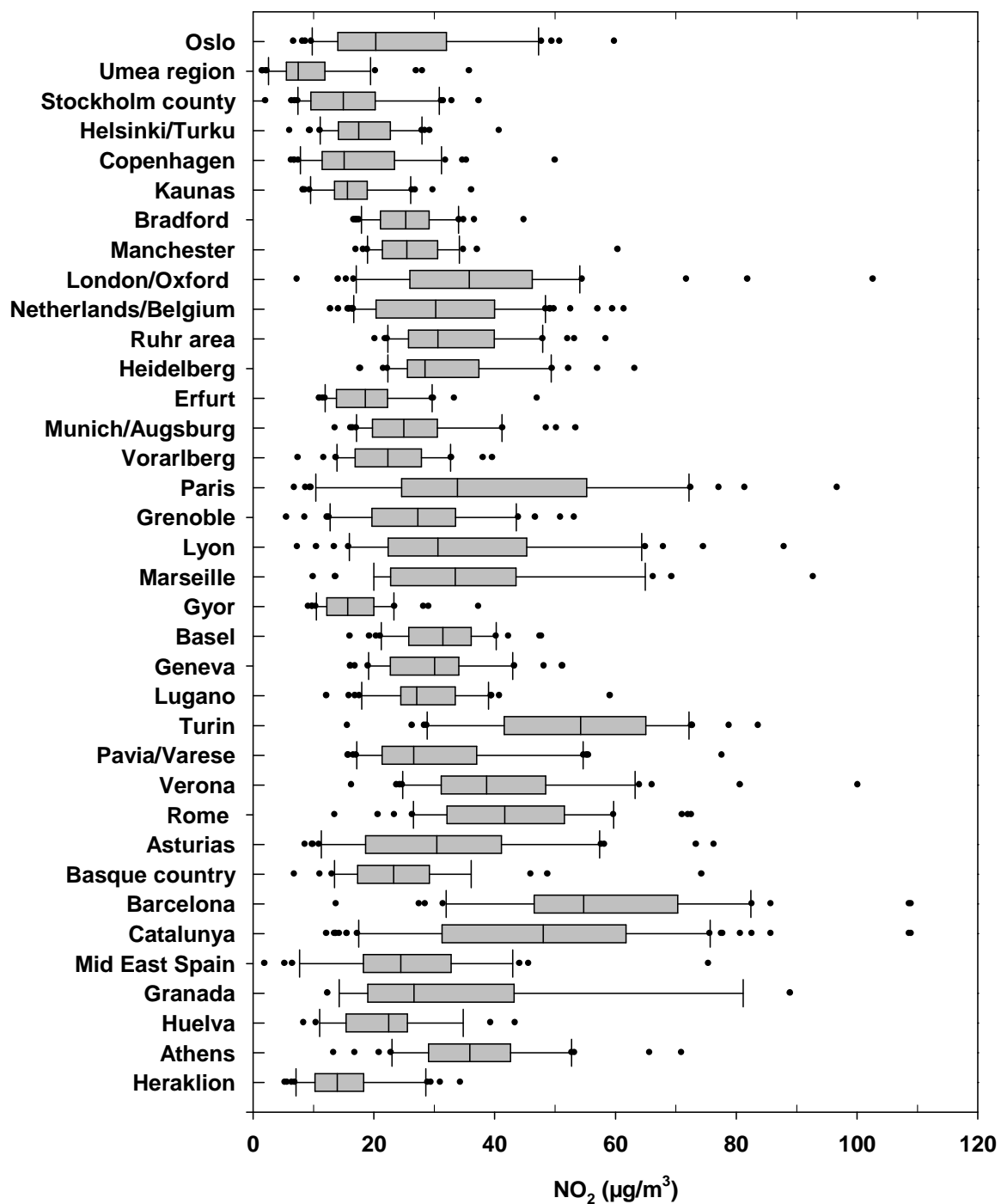
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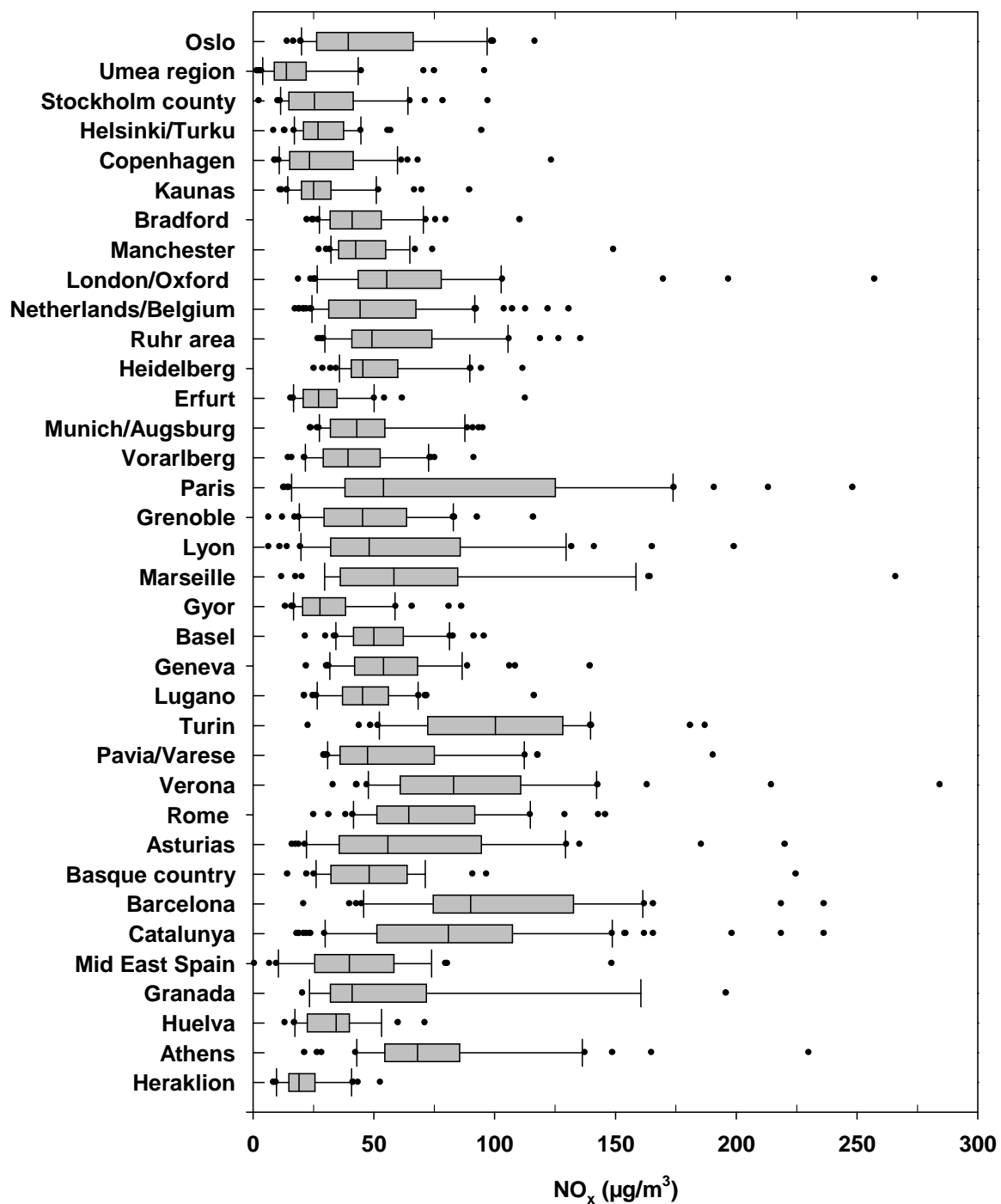
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873 Figure 1: ESCAPE study areas; dark circles mark the study areas where both PM and
874 NO_x were measured. Blue squares indicate the study areas where only NO_x was
875 measured



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Figure 2a: Distribution of annual average concentration of NO₂ 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.



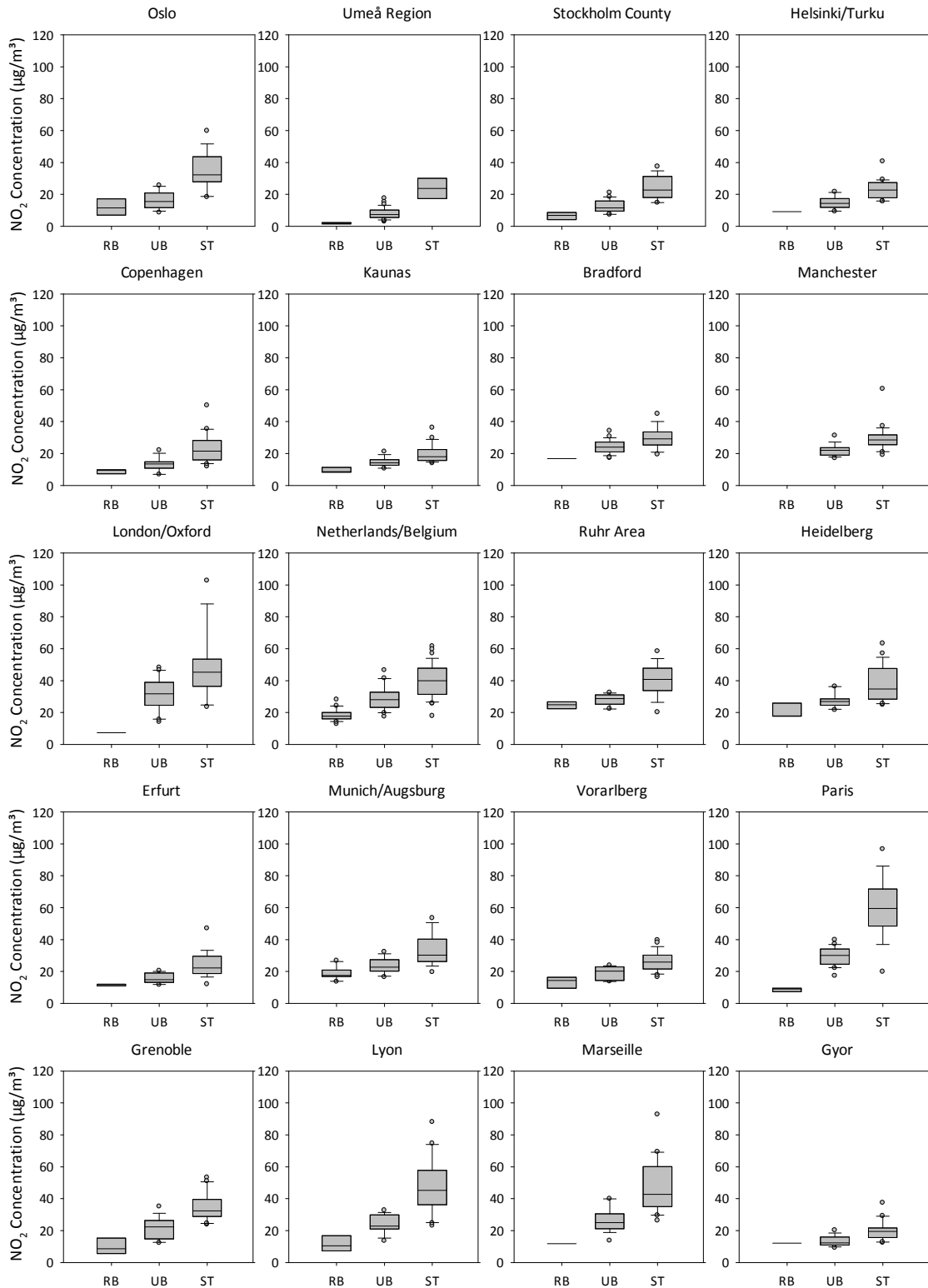
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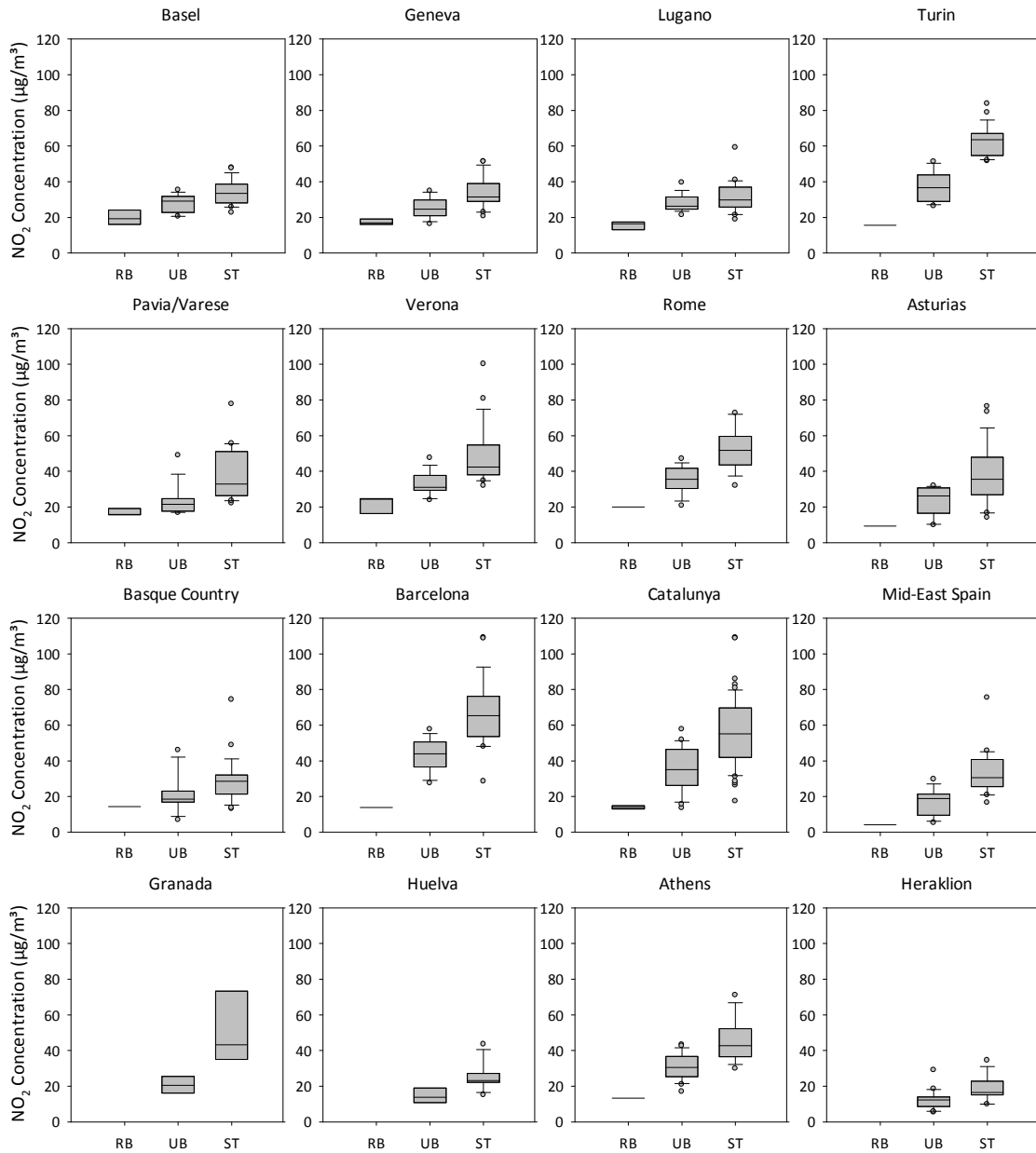
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884 Figure 2b: Distribution of annual average concentration of NO_x for each study area

885 separately. Median, 25th and 75th percentiles are shown in the box, whiskers indicate

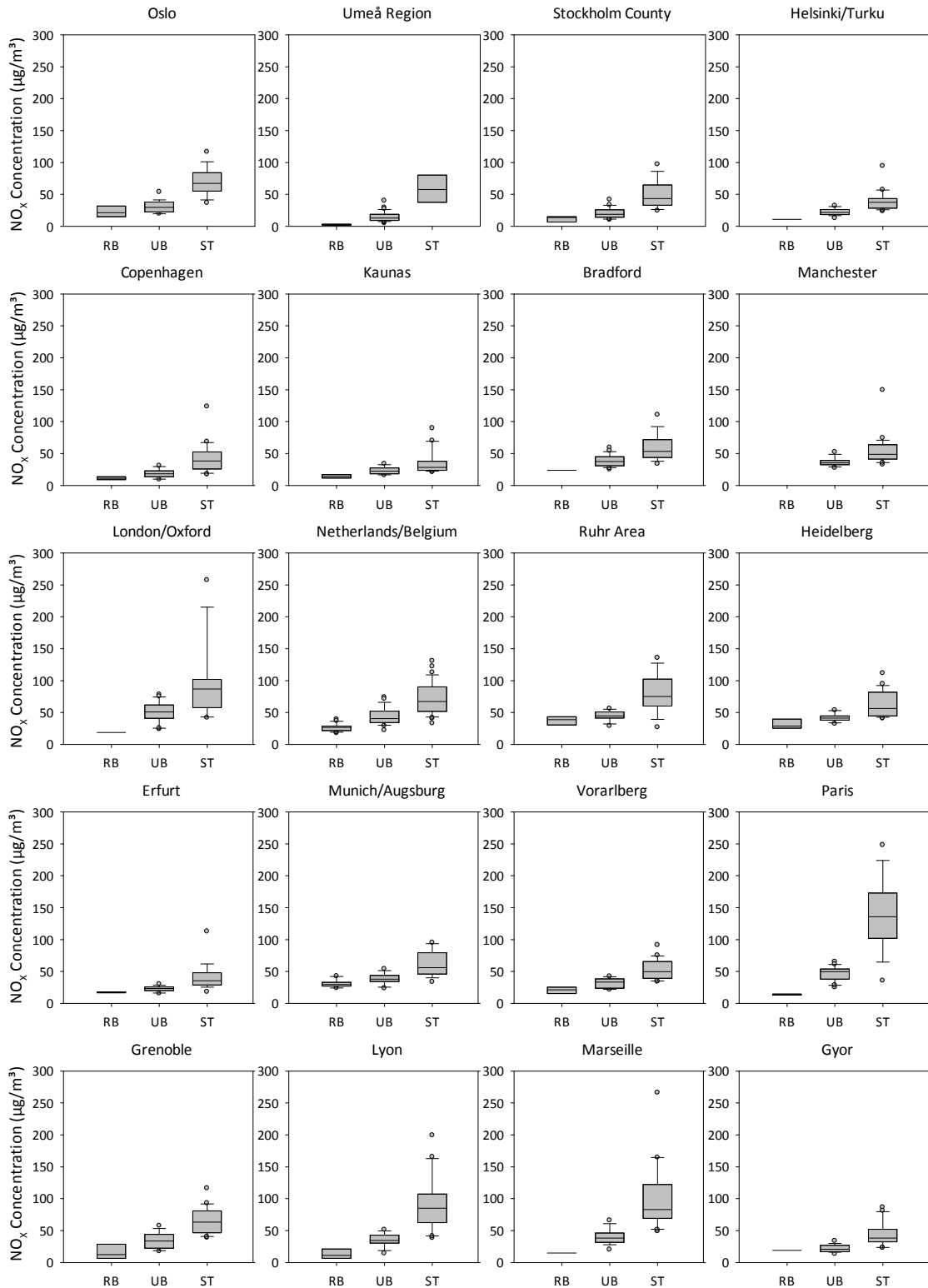
886 10th and 90th percentiles and individual outliers are shown as points.

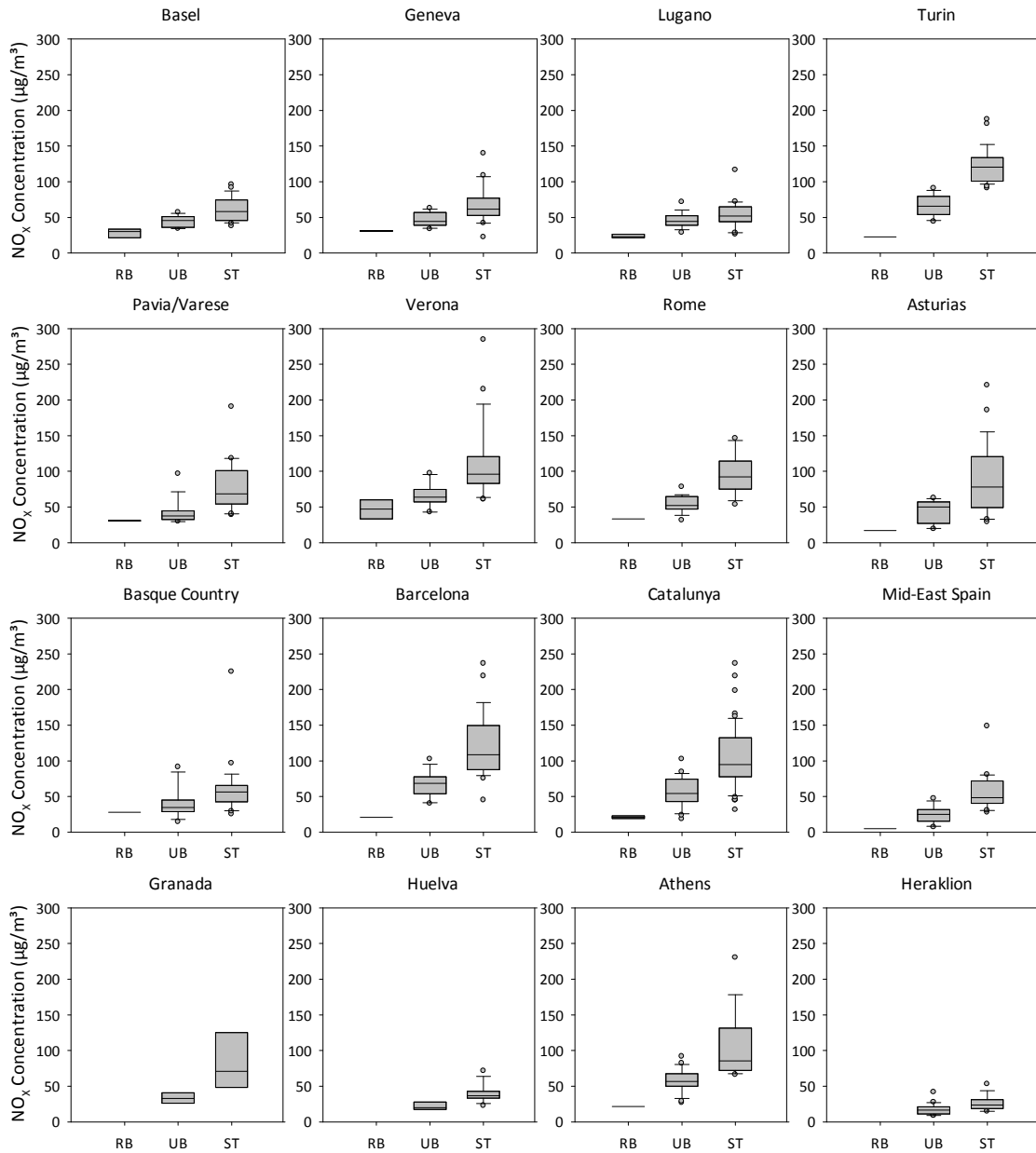




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Figure 3: Annual averages of NO_2 concentrations by site type, for each study area (for measurement period please refer to Table 1). Median, 25th and 75th percentiles are shown in the box, whiskers indicate 10th and 90th percentiles and individual outliers are shown as points. RB = Regional Background, UB = Urban Background and ST = Street locations.





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Figure 4: 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.

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: ² 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
	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
	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 neighboring 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 ¹	Greater Athens area, 16 municipalities; Athens	PM+ NO _x	2	21-04-2010 – 27-04-2011	40	1 / 22 / 17
	Heraklion ¹	Heraklion prefecture; Heraklion	PM+ NO _x	1	18-02-2009 – 16-02-2010	40	0 / 21 / 19

¹ 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.² RB = regional background / UB = urban background / ST = street site

909 Table 2: Detection limits and precision for NO₂ and NO_x measurements

	Field blanks						Field duplicates			
	Number field blanks	Average field blank (µg/m ³)		Detection limit (µg/m ³)		N samples below the detection limit (total number of valid samples)		Number duplicates	CV (%)	
Study area	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**
Copenhagen	20	0.3	-0.2	1.2	3.1	0 (143)	0 (143)	20	5.6	7.6
Kaunas*	24	0.1	0.8	0.6*	5.7*	0 (146)	3 (146)	24	8.5	10.4**
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*	4.6*	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*	9.5*	0 (137)	4 (137)	23	7.7	5.1
Turin	24	0.3	1.7	1.4*	10.0*	0 (144)	36 (144)	24	9.6	10.0**
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:***										
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:***										
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*	28.1*	1 (142)	6 (142)	27	7.7	7.8
Catalunya	23	0.5	3.7	3.3*	28.1*	1 (309)	39 (309)	27	7.7	7.8
Mid-East Spain:***										
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**	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

910 Detection limit calculated as three times the standard deviation of field blanks. CV= coefficient of variation calculated
 911 from field duplicates.

912 * Detection limit (DL) affected by one blank for which no explanation was found. Without this one blank, NO_x
 913 DL and number of samples below DL become 2.0 µg/m³ and 0 (Munich); 6.7 µg/m³ and 27 (Turin); 2.7 µg/m³
 914 and 0 (Kaunas); 7.2 µg/m³ and 0 (Barcelona); 7.2 µg/m³ and 0 (Catalunya); 2.8 µg/m³ and 0 (Lugano);

915 ** CV affected by one poor duplicate, for which no explanation was found. Without this duplicate CV becomes
 916 3.8% for NO_x (Helsinki); 5.4% for NO_x (Kaunas); 3.1% for NO_x (Turin); 4.7% for NO₂ (Valencia);

917 *** in three subareas different teams performed fieldwork, field duplicates / blanks treated analysed separately

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Table 3: Ratios between regional background and urban background concentrations, and between street and urban background concentrations, for all study areas.

StudyArea	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**

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*=significant difference between the site types on p<0.10 level
**=significant on p<0.05 level

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Table 4: Correlation and ratio between NO₂ and NO_x (R-squared).

	Study area	Correlation between NO ₂ and NO _x (R ²)*	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	1.93**	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

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Ratios between NO₂ and NO_x for all sites, regional background sites and street sites.

*All p-values were below <0.0001.

**=This value is different because in this particular case the ratio was only based on 2 regional background sites of which one had a negative NO concentration and hence a NO_x concentration which was lower than NO₂.