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Comparing land use regression and dispersion modelling to assess residential exposure to ambient air pollution for epidemiological studies

Kees de Hoogh [a,b,c,*], Michal Korek [c], Danielle Vienneau [a,b], Menno Keuken [e], Jaakko Kukkonen [f], Mark J. Nieuwenhuijsen [g,h,i], Chiara Badaloni [j], Rob Beelen [k], Andrea Bolignano [l], Giulia Cesaroni [j], Marta Cirach Pradas [g,h,i], Josef Cyrys [m,n], John Douros [o], Marloes Eeftens [a,b,k], Francesco Forastiere [j], Bertil Forsberg [p], Kateryna Fuks [q], Ulrike Gehring [k], Alexandros Gryparis [r], John Gulliver [c], Anna L Hansell [c,s], Barbara Hoffmann [q,t], Christer Johansson [u], Sander Jonkers [e], Leena Kangas [f], Klea Katsouyanni [r,v], Nino Künzli [a,b], Timo Lanki [w], Michael Memmesheimer [x], Nicolas Moussiopoulos [o], Lars Modig [p], Göran Pershagen [d], Nicole Probst-Hensch [a,b], Christian Schindler [a,b], Tamara Schikowski [a,b,q], Dorothee Sugiri [q], Oriol Teixidó [y], Ming-Yi Tsai [a,b,z], Tarja Yli-Tuomi [w], Bert Brunekreef [k,aa], Gerard Hoek [k], Tom Bellander [d,ab]

[a] Swiss Tropical and Public Health Institute, Basel, Switzerland
[b] University of Basel, Basel, Switzerland
[c] MRC-PHE Centre for Environment and Health, Department of Epidemiology and Biostatistics, Imperial College London, London, United Kingdom
[d] Institute of Environmental Medicine, Karolinska Institutet, Stockholm, Sweden
[e] Netherlands Organization for Applied Research, Utrecht, The Netherlands
[f] Finnish Meteorological Institute, Helsinki, Finland
[g] Center for Research in Environmental Epidemiology (CREAL), Barcelona, Spain
[h] IMIM (Hospital del Mar Research Institute), Barcelona, Spain
[i] CIBER Epidemiología y Salud Pública (CIBERESP), Spain
[j] Epidemiology Department, Lazio Regional Health Service, Rome, Italy
[k] Institute for Risk Assessment Sciences, Utrecht University, P.O. Box 80178, 3508 TD Utrecht, The Netherlands
[l] Environmental Protection Agency, Lazio Region, Italy
[m] Helmholtz Zentrum München, German Research Center for Environmental Health, Institutes of Epidemiology I and II, Neuherberg, Germany
[n] University of Augsburg, Environmental Science Center, Augsburg, Germany
[o] Laboratory of Heat Transfer and Environmental Engineering, Aristotle University of Thessaloniki, Aristotle University, Thessaloniki, Greece
[p] Department of Public Health and Clinical Medicine, Occupational and Environmental Medicine, Umeå University, Sweden
[q] IUF Leibniz Research Institute for Environmental Medicine, University of Düsseldorf, Düsseldorf, Germany
[r] Department of Hygiene, Epidemiology and Medical Statistics University of Athens, Medical School, Athens, Greece
[s] Directorate of Public Health and Primary Care, Imperial College Healthcare NHS Trust, London, UK
[t] Medical Faculty, Heinrich-Heine University of Düsseldorf, Düsseldorf, Germany
[u] Department of Applied Environmental Science, Stockholm University, Stockholm, Sweden
[v] Department of Primary Care & Public Health Sciences and Environmental Research Group, King's College London, United Kingdom
[w] Department of Environmental Health, National Institute for Health and Welfare (THL), Kuopio, Finland
[x] Rhenish Institute for Environmental Research (RIU), Köln, Germany
[y] Energy and Air quality Department, Barcelona Regional, Barcelona, Spain
[z] Department of Environmental and Occupational Health Sciences, University of Washington, Seattle, WA, United States
[aa] Julius Center for Health Sciences and Primary Care, University Medical Center Utrecht, Utrecht, The Netherlands
[ab] Centre for Occupational and Environmental Medicine, Stockholm County Council, Stockholm, Sweden

* Corresponding author at: Department of Epidemiology and Public Health, Swiss Tropical and Public Health Institute, Socinstrasse 57, CH-4051, Basel, Switzerland. Tel.: +41 61 284 8749; fax: +41 61 284 8105.
E-mail address: c.dehoogh@unibas.ch (K. de Hoogh).

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A B S T R A C T

Background: Land-use regression (LUR) and dispersion models (DM) are commonly used for estimating individual air pollution exposure in population studies. Few comparisons have however been made of the performance of these methods.

Objectives: Within the European Study of Cohorts for Air Pollution Effects (ESCAPE) we explored the differences between LUR and DM estimates for NO_2, PM_{10} and $PM_{2.5}$.

Methods: The ESCAPE study developed LUR models for outdoor air pollution levels based on a harmonised monitoring campaign. In thirteen ESCAPE study areas we further applied dispersion models. We compared LUR and DM estimates at the residential addresses of participants in 13 cohorts for NO_2; 7 for PM_{10} and 4 for $PM_{2.5}$. Additionally, we compared the DM estimates with measured concentrations at the 20–40 ESCAPE monitoring sites in each area.

Results: The median Pearson *R* (range) correlation coefficients between LUR and DM estimates for the annual average concentrations of NO_2, PM_{10} and $PM_{2.5}$ were 0.75 (0.19–0.89), 0.39 (0.23–0.66) and 0.29 (0.22–0.81) for 112,971 (13 study areas), 69,591 (7) and 28,519 (4) addresses respectively. The median Pearson *R* correlation coefficients (range) between DM estimates and ESCAPE measurements were of 0.74 (0.09–0.86) for NO_2; 0.58 (0.36–0.88) for PM_{10} and 0.58 (0.39–0.66) for $PM_{2.5}$.

Conclusions: LUR and dispersion model estimates correlated on average well for NO_2 but only moderately for PM_{10} and $PM_{2.5}$, with large variability across areas. DM predicted a moderate to large proportion of the measured variation for NO_2 but less for PM_{10} and $PM_{2.5}$.

1. Introduction

A large number of epidemiological studies have shown a clear association between long-term ambient air pollution exposure and adverse health effects (WHO, 2013). Several of these studies estimated individual air pollution exposures from stationary monitoring data, e.g. by using the nearest air pollution monitor to represent the pollution in entire cities (Dockery et al., 1993) to more complex approaches including spatial interpolation and kriging (Brauer et al., 2008; Künzli et al., 2005). Such methods provide estimates of large-scale spatial differences in air pollution concentrations, but are less effective in assessing intra-urban variation particularly when the number of monitoring sites is small. Recent studies have focused on intra-urban variation of air pollution, using indicators or proxies such as distance to the nearest road as well as pollutant levels estimated by land use regression (LUR), dispersion modelling (DM) including Chemical transport models (CTM) and hybrid models (HEI, 2010).

The LUR method, first developed by Briggs et al. (1997), uses least squares regression to combine monitored data with Geographic Information System (GIS)-based predictor data reflecting pollutant sources, to build a prediction model applicable to non-measured locations, e.g. residential addresses of cohort members. LUR modelling has been increasingly used in epidemiological studies because it is relatively cheap and can be easily implemented on the basis of purpose-designed monitoring campaigns or routinely measured concentrations and appropriate geographic predictors of air pollution sources (Hoek et al., 2008).

DMs are based on detailed knowledge of the physical, chemical, and fluid dynamical processes in the atmosphere. DMs use information on emissions, source characteristics, chemical and physical properties of the pollutants, topography, and meteorology to model the transport and transformation of gaseous or particulate pollutants through the atmosphere to predict, e.g., ground level concentrations (Holmes and Morawska, 2006; Kukkonen et al., 2012). Gaussian based DMs were originally developed as air quality management tools but have also been used in environmental epidemiology to model long-term exposures (Bellander et al., 2001; Wu et al., 2011). Chemical Transport Models have also been used to model short- and long-term exposure periods (Hennig et al., 2014). Few studies to date have conducted comparisons between LUR and DMs for their performance in estimating exposures (Beelen et al., 2010; Cyrys et al., 2005; Dijkema et al., 2011; Gulliver et al., 2011; Marshall et al., 2008; Sellier et al., 2014). These studies included different models, spatial resolution, pollutants and study areas, factors likely to have contributed to inconsistent findings within individual studies. As both LUR and DM are applied in epidemiology, there is a need for more comparison studies of these methods, addressing their respective advantages and strengths depending on the specific air pollution and health-related questions which are sought to be answered.

We compare LUR and DM to assess spatial variation of annual average ambient air pollution estimates at residential addresses within the framework of the European Study of Cohorts for Air Pollution Effects (ESCAPE), not taking into account population activity patterns or indoor air pollution. The ESCAPE study developed LUR models to estimate exposure at the residential addresses of cohort participants based on uniform monitoring campaigns and uniform modelling approaches in 36 study areas (Beelen et al., 2013; Cyrys et al., 2012; de Hoogh et al., 2013; Eeftens et al., 2012a,b). To several of these study areas we apply DM or use existing DM output, allowing for an in depth comparison to better understand the differences and/or agreements between LUR and DM estimates for use in epidemiological studies with long-term exposures. We include a range of exposure environments and populations across Europe, and focus, in particular, on the differences in estimated exposure at the individual participant level which is most relevant for interpretation of epidemiological studies.

2. Materials and methods

We estimated annual average outdoor air pollution concentrations for NO_2 in 13, PM_{10} in 7 and $PM_{2.5}$ in 4 of the 36 European cities/areas included in the ESCAPE study using both LUR and DM (Umeå region, Sweden; Stockholm County, Sweden (PM_{10}); Helsinki—Vantaa region, Finland ($PM_{2.5}$); Bradford, UK; London, UK (PM_{10}); Netherlands (PM_{10} & $PM_{2.5}$); Ruhr Area (PM_{10} & $PM_{2.5}$), Germany; Basel, Switzerland; Geneva, Switzerland; Lugano, Switzerland (PM_{10}); Rome, Italy ($PM_{2.5}$); Barcelona, Spain (PM_{10}); Athens, Greece (PM_{10})). The selection of study areas was based on the availability of existing dispersion models. A general discussion of these two modelling approaches is reported elsewhere (Hoek et al., 2008; Özkaynak et al., 2013).

We conducted several comparisons, depending on the comparability of the model outputs. The main comparison between the methods was made at the residential address of cohorts participants (referred to as LUR-DM). We also compared the DM estimates with measured concentrations at the ESCAPE monitoring sites. This was an independent validation, as monitoring data from the ESCAPE sites were not used as input data in the DM models. Recent studies have documented that the model R^2 and the leave-one out cross-validation R^2 overestimate the predictive ability of LUR models at independent sites (Basagaña et al., 2012; M. Wang et al., 2013). Therefore we cannot directly compare the explained variance of the LUR models with the explained variance of the dispersion models. Furthermore, we did not have a sufficiently large set of independent monitoring data available within the study areas to serve as an independent test set for both LUR and DM.

2.1. Description of cohorts

We used address locations of cohort participants as the basis for the LUR-DM comparison by study area. The majority of cohorts in this analysis were also used in the ESCAPE health studies: EPIC in Umeå (SE), SDPP, 60 years cohort, SALT and SNAC in Stockholm (SE), FINRISK in Helsinki (FI), Born in Bradford (UK), EPIC-Oxford in London (UK), PIAMA in the Netherlands, Heinz Nixdorf Recall (HNR) study in the Ruhr area (DE), SAPALDIA in Basel, Geneva and Lugano (CH) and SIDRIA in Rome (IT). For Barcelona (ES), we chose the larger population of the ARIBA cohort ($n = 8{,}402$), rather than the ECRHS cohort ($n = 297$) used in ESCAPE. Due to confidentiality, address locations of the EPIC cohort in Athens (GR) were not available; instead we used 1500 randomly selected addresses across the study area to act as a cohort surrogate. Most of the study areas were large cities and the surrounding suburban or rural communities; however, some of the cohorts covered larger regions, such as PIAMA in the Netherlands. In total, we used 112,971 address locations over 13 cohorts.

2.2. Land use regression modelling

The ESCAPE study involved harmonised monitoring campaigns for NO_2 in 36 study areas and $PM_{10}/PM_{2.5}$ in 20 study areas, as described in Cyrys et al. (2012) and Eeftens et al. (2012a). In brief, in each study area a measurement campaign was carried out during three 2-week periods within one year. The complete monitoring period across all study areas was between 2008 and 2011. Ogawa badges were used for monitoring of NO_2 and Harvard Impactors were used for monitoring of PM. Care was taken to select site locations to incorporate relevant intra-urban spatial variation in traffic and land use characteristics. Adjusted annual mean concentrations for each site were then estimated with the aid of measurement data from an all-year running reference site in an urban background location in each study area.

Based on these measurements, LUR models were developed in each study area following a standardised approach (Beelen et al., 2013; Eeftens et al., 2012b). Geographical Information Systems (GIS) predictor variables were collected for all study areas centrally (EU-wide datasets including CORINE land cover, EuroStreets road network, altitude and population density) and locally (traffic data and, where available, more detailed land cover data). Circular buffers with radii of 25, 50, 100, 300, 500, and 1000 m were used to calculate traffic and road variables for each monitoring location. For land use and population, buffers of 100, 300, 500, 1000, and 5000 m were calculated. LUR models were developed combining the adjusted annual means and the GIS predictor data in each study area following a stringent set of rules. Linear regression was performed in a stepwise logical standardised approach, detailed by Eeftens et al. (2012b). Predictors giving the highest adjusted R^2 were subsequently added to the model if they conformed to the direction of effect defined a priori and added more than 1% to the adjusted R^2. Final models were checked for p-value (removed when p-value >0.10), co-linearity (variables with Variance Inflation Factor (VIF) >3 were removed and model rerun) and influential observations (models with Cook's D >1 were further examined). The final models were evaluated by leave-one-out cross validation (LOOCV).

Model structure, model R^2 and LOOCV R^2 of the LUR models in the included 13 study areas are shown in Table A.1. LUR model predictions at the cohort address were based on predictor values restricted to the range of observed values at the monitoring sites, in order to prevent extrapolation beyond the range for which the model was developed.

2.3. Dispersion modelling

DM was applied in the 13 study areas by third parties using input data including traffic flow, road geometry, other non-traffic pollution sources (e.g. industrial and agricultural sources), meteorological parameters and concentrations measured at regional and urban background sites. In ten of the 13 study areas a Gaussian plume DM was used: Airviro in Stockholm and Umeå Region; CAR-FMI in Helsinki; ADMS-Urban in Bradford, London and Barcelona; CAR and Pluim Snelweg (motorway) in the Netherlands; Pollumap DM 2010 in Basel, Geneva, and Lugano. Two areas used Eulerian or chemical transport models: EURAD-CTM in Ruhr area; Flexible Air quality Regional Model (FARM) in Rome and one used a Computational Fluid Dynamic (CFD) model; MEMO/MARS-aero in Athens. Information about the DM by study area is shown in Table 1. Models differed in the sources included (all models including traffic sources but some additionally including industry and agricultural sources), the scale of assessment and the representation of regional background (most used routine monitoring data). The effective spatial scale of the receptor-oriented methods depends on several factors, e.g. the precision of the spatial description of sources and topography, and could not be estimated. DM estimates were extracted to the addresses of the cohorts involved.

2.4. Statistical analyses

Exposure estimates from LUR and DM were compared at the address level. We calculated Pearson (R) and Spearman (Rho) correlation coefficients and show scatterplots of the relationship. The LUR and DM exposure estimates were also categorised into quintiles as epidemiological studies often use categorical analyses to relax the assumption of a linear association. Kappa coefficients were calculated to assess the level of agreement beyond chance. Bland–Altman plots were produced to further investigate the agreement between the two methods, specifically to test whether the difference between LUR and DM depends on the absolute concentrations. In addition, the correlation between the DM estimates and monitored concentrations at the ESCAPE monitoring sites was calculated (R and Rho) and visualised in scatterplots (DM-MON).

Statistical analysis was carried out in STATA version 11.0 (StataCorp LP, College Station, Texas, USA).

3. Results

3.1. Comparison of LUR and DM at address level

Distributions of LUR and DM predictions at the cohort addresses, the correlation and Kappa coefficients are shown in Table 2. Fig. 1 shows scatterplots of LUR and DM predictions.

3.1.1. NO_2

LUR and DM estimates of NO_2 levels for cohort members were available for the 13 study areas at a total of 112,971 residential addresses. The correlation (Pearson R) between LUR and DM estimates of NO_2 levels at cohort addresses varied from 0.19 (Athens) to 0.89 (The Netherlands; Fig. 1, Table 2). The Spearman rank correlation (R) ranged from 0.21 to 0.90. The median Pearson and Spearman correlation coefficients were 0.75 and 0.77 respectively, indicating overall good agreement. The agreement by quintiles ranged from 24% to 62%. Kappa statistics ranged from 0.005 to 0.52 (Table 2).

The overall median of estimated NO_2 concentrations was slightly higher for LUR (21.4 $\mu g/m^3$) than for DM predictions (17.3 $\mu g/m^3$). The difference between LUR and DM median estimates was up to 11.9 $\mu g/m^3$ (Rome; Table 2). In the areas with the largest differences between LUR and DM estimates, the DM/CTM modelled an average concentration over an area of 0.25–1 km^2, in contrast to LUR which modelled concentrations at individual address (receptor) points. The

Table 1
Details of atmospheric dispersion models used to predict air pollution concentrations in each study area.

Study area	Name of dispersion model	Type	Pollutants	Geographical resolution output	Year output	Regional background	Sources	Street canyon	Reference(s)
Umeå region, SE	Airviro Gauss dispersion model	Gaussian plume	NO_2	50 × 50 m	2010	Monitoring[a]	T, P, R[b]	No	SMHI (1993)
Stockholm County, SE	Airviro Gauss dispersion model	Gaussian plume	NO_2, PM_{10}	25 × 25 m in urban, 500 × 500 m in rural area	2009	Monitoring[a]	T	Yes	SMHI (1993)
Helsinki-Vantaa region. FI	CAR-FMI (Contaminants in the Air from a Road – Finnish Meteorological Institute)	Gaussian plume	NO_2, $PM_{2.5}$	At unique receptor points	2010	Monitoring[a]	T	No	Kukkonen et al. (2001), Karppinen et al. (2000)
Bradford, UK	ADMS-Urban	Gaussian plume	NO_2	At unique receptor points	2009	Monitoring[a]	T, A	No	Carruthers et al. (2000)
London, Oxford, UK	ADMS-Urban	Gaussian plume	NO_2, PM_{10}	10 × 10 m	2011	Monitoring[a]	T, A	Yes	Carruthers et al. (2000)
Netherlands	GCN (Generic Concentrations in the Netherlands), for the regional/urban background, Pluim Snelweg for the motorways and provincial roads, CAR model for the urban roads	Gaussian plume	NO_2, PM_{10}, $PM_{2.5}$	25 × 25 m	2009	Model[c]	T, P, A	Yes, included in CAR	Velders et al. (2013) Wesseling and Visser (2003) Wesseling and Sauter (2007)
Ruhr Area, DE	EURopean Air Pollution Dispersion (EURAD) model system	Dispersion and chemical transport model	NO_2, PM_{10}	1 × 1 km	2006-2008	Monitoring	T, P, R, A	No	Memmesheimer et al. (2004)
Basel, Geneva and Lugano, CH	Pollumap dispersion model 2010	Gaussian plume	NO_2 (All), PM_{10} (Lugano only)	100 × 100 m	2010		T, A	No	SAEFL (2003) Gariazzo et al. (2007)
Rome, IT	Flexible Air quality Regional Model (FARM)	Eulerian chemical transport model	NO_2, $PM_{2.5}$	1 × 1 km	2007		T, A	No	Gariazzo et al. (2007) Finardi et al. (2009)
Barcelona, ES	ADMS-Urban	Gaussian plume	NO_2, PM_{10}	5 × 5 m for NO_2, 100 × 100 m for PM_{10}	2008	Monitoring[a]	T, P, R, A	Yes	Carruthers et al. (2000)
Athens, GR	MEMO/MARS-aero	Eulerian chemical transport model	NO_2, PM_{10}	500 × 500 m	2008	Model		No	Moussiopoulos et al. (2012)

[a] Monitoring data from regional background station.
[b] T = traffic; P = point sources; R = residential heating; A = area source for all non-traffic sources.
[c] Combination of monitoring and modelling at 1 × 1 km scale.

relative difference between the median NO_2 LUR and DM predictions was however not large in these areas (<~30%).

The estimated ranges of NO_2 concentrations differed for the two methods, with some study areas showing a distinctly narrower range for LUR estimates compared to DM estimates (Bradford and the Netherlands) and other study areas showing a larger range for LUR estimates than for DM estimates (Ruhr Area, Athens, Lugano, Barcelona, and London).

3.1.2. PM_{10}

PM_{10} concentrations were modelled with LUR and DM for 69,591 residential addresses in 7 study areas. The correlation between LUR and DM was generally lower and the differences in levels larger than for NO_2 (Table 2, Fig. 1). A large difference of 20 μg/m^3, for instance, was found between median PM_{10} concentrations for LUR and DM in Athens, whereas the differences in the Netherlands and Lugano were small (0.3 and 1.2 μg/m^3 respectively). The median Pearson and Spearman correlation coefficients between LUR and DM estimates were 0.39 and 0.49 respectively. Lugano, the Netherlands and London showed the highest correlations (Pearson) between the 2 methods (R = 0.66, 0.56 and 0.52 respectively). In several of the LUR predictions the impact of truncation to the highest value of predictor variables at the monitoring sites is visible, e.g. in the Netherlands (Fig. 1). In Stockholm, the dispersion model had a lower bound, defined by the measured regional background used as input in the model. The percentage of agreement by quintiles ranged from 25 to 55%.

3.1.3. $PM_{2.5}$

Estimated $PM_{2.5}$ concentrations were modelled in four study areas (Helsinki—Vantaa region, the Netherlands, the Ruhr Area, and Rome) for a total of 28,159 residential addresses. In the Netherlands there was a high correlation (Pearson R = 0.81), with similar median $PM_{2.5}$ concentrations for both methods, but with a larger range for DM estimates (14.5 μg/m^3) compared to LUR estimates (6.2 μg/m^3). The other three study areas showed low correlations between the LUR and DM estimates.

The Bland–Altman plots (Fig. A.1) were inspected to assess the agreement over the concentration range between the two methods. The majority of points were located within +/− 2 times the standard deviation; however, there were quite different patterns for the different study areas and pollutants. Fig. A.1 shows that bias rarely is zero (only Basel (NO_2), Netherland (NO_2, PM_{10}, and $PM_{2.5}$), Lugano (PM_{10}) and Helsinki, and Rome ($PM_{2.5}$) have an absolute mean difference of less than 1 μg/m^3). Secondly the upper- and lower-limits of the 95% range

Table 2
Descriptive and comparison statistics of LUR and dispersion estimates (μg/m^3) at cohort address for NO_2, PM_{10} and $PM_{2.5}$.

		LUR predictions (μg/m^3)				DM predictions (μg/m^3)				Comparison of LUR with DM						
										Continuous: DM = Constant + Slope × LUR					Quintiles	
Study area	N[a]	Median	P_{05}	P_{95}	P_{95}–P_{05}	Median	P_{05}	P_{95}	P_{95}–P_{05}	Spearman's Rho	Pearson *R*	Constant	Slope	RMSE	Agreement (%)[b]	Kappa
NO_2																
Umeå region, SE[c]	4575	6.8	4.1	16.4	12.2	12.5	5.6	20.6	15.0	0.782	0.792	5.17	0.93	2.63	48.3	0.352
Stockholm County, SE[c]	39409	9.6	6.4	20.9	14.5	6.5	3.3	18.1	14.9	0.791	0.856	−1.98	0.93	2.46	48.9	0.361
Helsinki–Vantaa region, FI[c]	5871	16.0	9.0	25.5	16.5	9.0	7.0	17.0	10.0	0.762	0.745	2.01	0.52	2.34	43.7	0.297
Bradford, UK[c]	20919	24.0	18.9	29.0	10.1	18.3	14.0	26.5	12.5	0.820	0.667	−1.62	0.86	3.06	49.2	0.365
London, UK[c]	7089	33.3	21.7	45.5	23.8	32.0	21.1	42.6	21.4	0.836	0.798	8.55	0.70	4.05	55.2	0.441
Netherlands[c]	7295	22.7	12.7	33.9	21.2	24.0	11.4	38.2	26.8	0.901	0.891	−2.37	1.13	3.70	61.8	0.523
Ruhr Area, DE[d]	4809	29.6	23.4	38.6	15.2	37.5	30.8	44.1	13.3	0.428	0.389	28.45	0.30	3.51	31.0	0.138
Basel, SU[c]	1118	29.0	18.3	34.3	16.0	30.5	21.4	34.4	13.1	0.771	0.768	11.11	0.65	2.71	48.9	0.362
Geneva, SU[c]	737	26.4	16.2	38.7	22.6	31.7	24.4	36.0	11.7	0.708	0.657	21.73	0.36	2.84	41.4	0.267
Lugano, SU[c]	1090	26.6	11.8	39.2	27.3	30.9	22.9	34.8	12.0	0.773	0.819	20.43	0.37	1.97	50.2	0.377
Rome, IT[d]	10157	38.1	25.5	56.1	30.5	50.0	31.5	59.4	27.8	0.406	0.386	33.35	0.36	7.65	29.4	0.120
Barcelona, ES[c]	8402	57.1	38.5	85.1	46.6	54.0	39.7	78.4	38.7	0.687	0.688	21.41	0.59	8.84	43.3	0.292
Athens, GR[d]	1500	36.0	23.4	59.5	36.0	47.0	36.5	56.4	19.8	0.207	0.188	42.86	0.10	6.35	23.9	0.005
All	112971	21.4				17.3										
PM_{10}																
Stockholm County, SE[c]	39409	15.1	6.2	20.4	14.2	10.0	7.8	16.6	8.8	0.378	0.367	6.83	0.29	2.82	31.2	0.140
London, UK[c]	7089	16.9	14.9	20.9	6.1	21.7	20.7	23.0	2.4	0.554	0.517	17.94	0.22	0.65	55.2	0.441
Netherlands[c]	7295	24.6	23.8	27.1	3.3	24.9	20.4	27.2	6.7	0.625	0.556	−4.88	1.16	1.91	42.0	0.275
Ruhr Area, DE[d]	4809	27.5	25.3	31.6	6.3	18.0	15.1	22.5	7.4	0.328	0.346	5.97	0.43	2.18	24.8	0.060
Lugano, SU[c]	1087	23.3	18.0	27.4	9.4	24.5	20.4	25.9	5.5	0.575	0.659	13.87	0.43	1.25	39.8	0.248
Barcelona, ES[c]	8402	39.0	37.0	47.5	10.6	37.4	35.7	44.2	8.5	0.495	0.393	24.14	0.35	2.62	33.1	0.163
Athens, GR[d]	1500	47.0	24.7	64.1	39.4	27.0	23.4	30.3	7.0	0.272	0.233	24.70	0.046	2.36	26.5	0.080
All	69591	16.6				15.1										
$PM_{2.5}$																
Helsinki–Vantaa region, FI[c]	5871	8.0	5.6	9.1	3.5	8.5	8.2	9.3	1.1	0.215	0.252	7.85	0.093	0.37	25.8	0.073
Netherlands[c]	7295	16.5	15.4	17.3	1.9	16.8	13.1	18.6	5.6	0.879	0.812	−20.40	2.23	0.41	50.4	0.380
Ruhr Area, DE[d]	4809	18.3	16.9	20.4	3.5	14.7	13.1	16.7	3.6	0.391	0.327	8.21	0.35	1.12	28.0	0.100
Rome, IT[d]	10544	18.9	17.3	23.3	6.0	20.1	16.5	21.6	5.0	0.252	0.223	16.03	0.19	1.53	26.5	0.081
All	28159	17.4				16.8										

[a] Number of residential addresses in the participating cohorts.
[b] Percentage of residential addresses falling in the same quintile.
[c] Spatial resolution of DM estimates ≤100 × 100 m.
[d] Spatial resolution of DM estimates ≥500 × 500 m.

differ widely between the study areas. Fig. A1 also shows which of the two methods tends to provide higher or lower concentration estimates. For example NO_2 estimates in Bradford are mostly higher with LUR (95% range = −12.7 to 0.7 μg/m^3) while the opposite is true in Umeå (95% range = −0.6 to 9.8 μg/m^3). For the coarse-scale models and the three Swiss models, the DM model predictions were lower than the LUR predictions for the highest concentrations, mostly traffic locations.

3.2. Comparison DM with ESCAPE monitoring results

Correlations between dispersion modelled annual average concentrations and adjusted annual average concentrations based on measurements at the ESCAPE monitoring sites are shown in Table 3 (scatterplots in Fig. 2, Table A.2). Pearson *R*'s correlation coefficients ranged from 0.09 (Athens) to 0.86 (Umeå) for NO_2, with a median of 0.74. Dispersion models that aimed to predict at specific receptor points or predict with a very small resolution of <100 × 100 m predicted NO_2 concentrations better than the coarser Eulerain/CFD models. The median correlation for PM_{10} (0.58, ranging from 0.36 (Barcelona) to 0.88 (London)) was lower than for NO_2, which again was mainly driven by the difference in scale. Among the four study areas with a DM for $PM_{2.5}$, the two models that estimated at unique receptor points or on a small spatial scale (Helsinki–Vantaa region, Netherlands) predicted measured concentrations with correlations of 0.66 and 0.54 (Pearson), respectively. Correlations for the larger scale models were 0.39 (the Ruhr Area) to 0.61 (Rome). For most of the study areas Spearman correlations were moderate to high (ranges: NO_2 0.15 to 0.88; PM_{10} 0.47 to 0.70 and $PM_{2.5}$ 0.49 to 0.70). For the majority of the study areas DM thus tend to predict a fairly large proportion ($R > 0.6$) of the variation across the measurement sites. Scatter plots of the DM-MON comparison are shown in Fig. 2. The regression lines for NO_2 generally follow the 1:1 line, whereas regression lines for PM_{10} and $PM_{2.5}$ show departures from the 1:1 line. Relatively large differences in NO_2 concentrations were found only in Umeå (DM > measured) and Helsinki–Vantaa region (DM < measured), though in both areas the correlation was higher than 0.6. PM_{10} concentrations were higher than the model predictions in Athens, though the correlation was reasonable. Fig. 3 illustrates that the agreement between LUR and DM at the cohort addresses increases with increasing correlation between the DM and measured concentrations at the monitoring sites. The agreement between LUR and DM did not depend on the LOOCV of the LUR model.

4. Discussion

To our knowledge, this is the first study to compare LUR and DM for assigning air pollution exposures to a large number of residential addresses in different geographical areas. In general, a distinction between two types of DM can be made: one estimates receptor-specific concentrations (Gaussian) and the other estimates average concentrations for an area (Eulerian/CFD). This has potential implications on the comparability of air pollution estimates at the address level and for the downstream epidemiology.

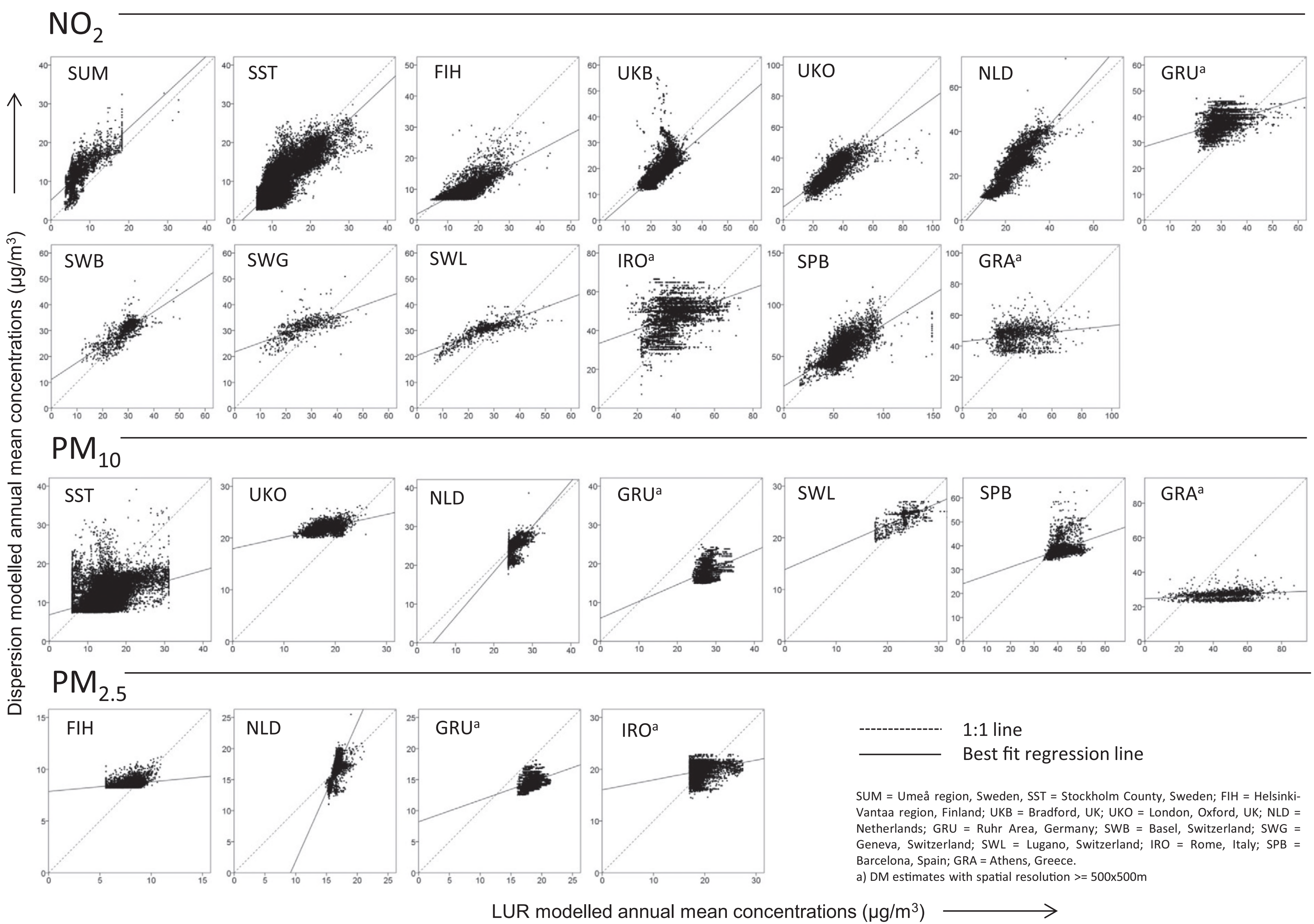

Fig. 1. Scatter plots of DM (*y*-axis) against LUR (*x*-axis) estimates (μg/m^3) at residential addresses for NO_2, PM_{10} and $PM_{2.5}$.

Table 3
Descriptive and comparison statistics of DM estimates and measurements (μg/m^3) at ESCAPE monitoring sites for NO_2, PM_{10} and $PM_{2.5}$.

Study area	N[a]	Monitored concentrations at ESCAPE sites (μg/m^3)			DM predictions at ESCAPE sites (μg/m^3)			Comparison of DM predictions with measured concentrations at ESCAPE sites				
		Median	Min	Max	Median	Min	Max	Spearman's Rho	Pearson R	Constant	Slope	RMSE
NO_2												
Umeå region, SE[b]	20	9.3	5.3	35.8	15.5	7.4	31.0	0.878	0.858	-5.36	1.02	3.88
Stockholm County, SE[b]	39	14.8	2.1	33.0	13.0	2.9	25.3	0.775	0.755	4.41	0.84	4.94
Helsinki—Vantaa region, FI[b]	25	19.7	12.2	28.5	10.6	6.6	26.7	0.753	0.658	12.64	0.63	3.78
Bradford, UK[b]	40	25.2	16.7	36.7	19.8	13.0	38.0	0.806	0.743	11.99	0.62	3.59
London, UK[b]	27	39.7	29.2	102.7	37.7	23.0	79.9	0.681	0.849	-10.83	1.39	9.06
Netherlands[b]	68	28.0	12.8	57.1	27.7	11.1	47.1	0.897	0.852	1.05	0.99	5.45
Ruhr Area, DE[c]	29	31.2	22.2	58.4	39.2	28.5	50.2	0.459	0.391	5.47	0.72	8.98
Basel, SU[b]	40	31.4	16.1	47.8	31.8	21.4	35.2	0.492	0.598	-14.10	1.46	5.98
Geneva, SU[b]	41	30.1	16.1	51.3	31.2	20.4	40.9	0.642	0.540	-5.90	1.17	7.66
Lugano, SU[b]	42	27.1	12.2	59.2	31.7	23.4	39.3	0.749	0.764	-33.72	2.00	5.37
Rome, IT[c]	40	41.7	13.6	72.6	50.0	26.6	62.1	0.568	0.614	-8.76	1.08	10.96
Barcelona, ES[b]	40	54.7	13.8	109.0	51.2	28.5	78.5	0.805	0.754	-4.61	1.15	13.40
Athens, GR[c]	40	35.9	13.3	71.0	40.4	34.4	52.2	0.154	0.089	27.86	0.20	12.04
PM_{10}												
Stockholm County, SE[b]	19	18.5	5.7	35.6	15.2	7.5	19.3	0.472	0.580	11.33	1.09	5.65
London, UK[b]	13	18.4	16.1	31.2	22.3	21.5	30.7	0.484	0.877	-13.47	1.46	2.03
Netherlands[b]	34	26.2	21.9	33.0	25.7	20.8	30.5	0.671	0.696	3.74	0.88	2.18
Ruhr Area, DE[c]	15	27.4	22.5	33.3	18.2	15.3	32.3	0.521	0.392	22.95	0.25	2.90
Lugano, SU[b]	18	23.9	18.5	32.5	24.1	20.1	25.3	0.552	0.668	-8.77	1.38	2.67
Barcelona, ES[b]	20	38.6	17.8	48.5	36.7	34.5	51.3	0.699	0.356	10.71	0.71	6.82
Athens, GR[c]	20	42.9	27.3	58.0	24.5	22.8	30.3	0.522	0.397	9.42	1.32	6.81
$PM_{2.5}$												
Helsinki—Vantaa region, FI[b]	13	8.9	7.9	10.4	8.8	8.2	10.1	0.703	0.657	1.39	0.86	0.64
Netherlands[b]	34	17.4	12.7	21.0	17.5	13.4	21.0	0.485	0.540	8.4	0.52	1.54
Ruhr Area, DE[c]	15	18.5	15.5	21.1	14.9	13.0	25.1	0.492	0.387	15.8	0.17	1.48
Rome, IT[c]	18	18.5	14.2	27.0	20.5	16.6	21.9	0.598	0.612	−11.0	1.53	2.74

[a] Number of ESCAPE monitoring sites.
[b] Spatial resolution of DM estimates ≤100 × 100 m.
[c] Spatial resolution of DM estimates ≥500 × 500 m.

Overall, agreement between DM and LUR was quite strong for NO_2 in 7 out of 13 study areas, (Pearson $R > 0.70$). Lower agreement was found for PM_{10} and $PM_{2.5}$. Agreement between LUR and DM at the address level was higher for areas where the DM correlated more strongly with the measurements.

4.1. Prediction of measured concentrations at monitoring sites

Gaussian DMs generally predicted the spatial variation of NO_2 at monitoring sites well, reflecting the small-scale variation of this pollutant. On the other hand Eulerian/CFD DMs that modelled average NO_2 concentrations on a coarser spatial scale reflected larger scale variations of urban background within cities. Most models also predicted the concentration levels well (within about 30%), partly due to the incorporation of measured regional background concentrations. Prediction of PM was less effective, similar to LUR models (Beelen et al., 2013; Eeftens et al., 2012b).

As we did not have independent data available for a sufficiently large number of locations in our cities, we cannot make a solid comparison between the two models' predictive ability for the study areas. The correlations between DM and measured concentrations were however lower than for the LUR models (median LOOCV R^2 was 0.80 (0.55), 0.77 (0.34) and 0.61 (0.33) for LUR (DM) NO_2, PM_{10} and $PM_{2.5}$ respectively (Table A.2)). This does not necessarily imply better performance at unmeasured locations. The model R^2 only represents the predictive ability at the monitoring sites and recent studies have documented that the LOOCV R^2 used in LUR studies only partly compensates for the over-fitting. Hold-out validation R^2 has been shown to be potentially 20–40% lower than the model R^2, with larger differences observed for LUR models based on a smaller number of sites (Basagaña et al., 2012; M. Wang et al., 2013).

The RMSE of the comparison between DM and measurements (Table 3) was larger than the RMSE of the comparison between DM and LUR (Table 2). Although based on different locations, this might indicate that both models may have similar errors in explaining measurements.

Several previous studies have compared LUR and DM at monitoring sites. Beelen et al. (2010) found moderate agreement ($R = 0.55$) between LUR and DM estimates for annual average NO_2 concentrations at a 100 × 100 m grid in the Rijnmond area of the Netherlands with the URBIS performing better than the LUR model ($R = 0.77$ vs 0.47) at 18 independent sites. This is likely because the LUR model was developed for the whole of the Netherlands and lacking specific local information for the Rijnmond area. A study in Amsterdam (NL) by Dijkema et al. (2011) compared NO_2 concentrations estimated by 2 LUR models (regional and city specific) against the Dutch CAR dispersion model. All models explained between 50 and 60% of the variance, although CAR overestimated at background and underestimated at traffic monitoring sites. In Vancouver, Canada, Marshall et al. (2008) compared LUR and a 4 × 4 km chemical transport DM (CMAQ) to estimate NO, NO_2, CO and ozone. They found that LUR was better in predicting the small spatial variations at the neighbourhood scale, whereas DM tended to be better in predicting the urban scale variations. Cyrys et al. (2005) also compared LUR and dispersion modelling for NO_2 and $PM_{2.5}$ in Munich, Germany, at 40 monitoring sites and at 1669 addresses. The model estimates correlated well at the 40 monitoring sites and addresses ($R > 0.79$). Gulliver et al. (2011) compared LUR and DM at 52 routine monitoring stations in London (UK) using a grouped jack-knife approach Results showed that LUR ($R^2 = 0.47$) outperformed DM ($R^2 = 0.28$). Most recently Sellier et al. (2014) compared LUR and DM estimates for NO_2 at cohort addresses in Nancy and Poitiers (France) finding a good correlation between the two methods ($R = 0.87$).

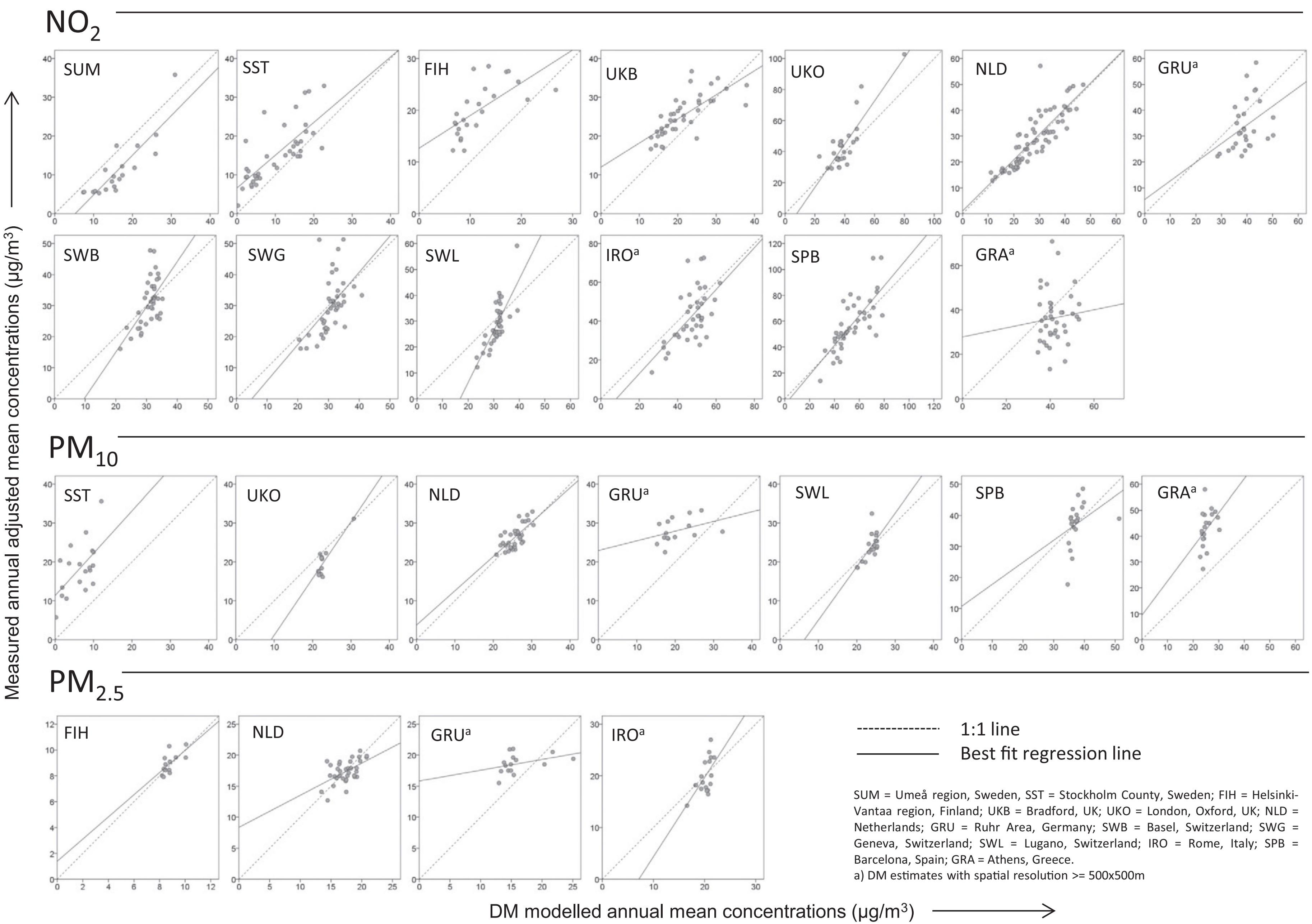

Fig. 2. Comparison of measured annual adjusted concentrations (*x*-axis) against DM (*y*-axis) estimates (μg/m^3) at ESCAPE monitoring sites.

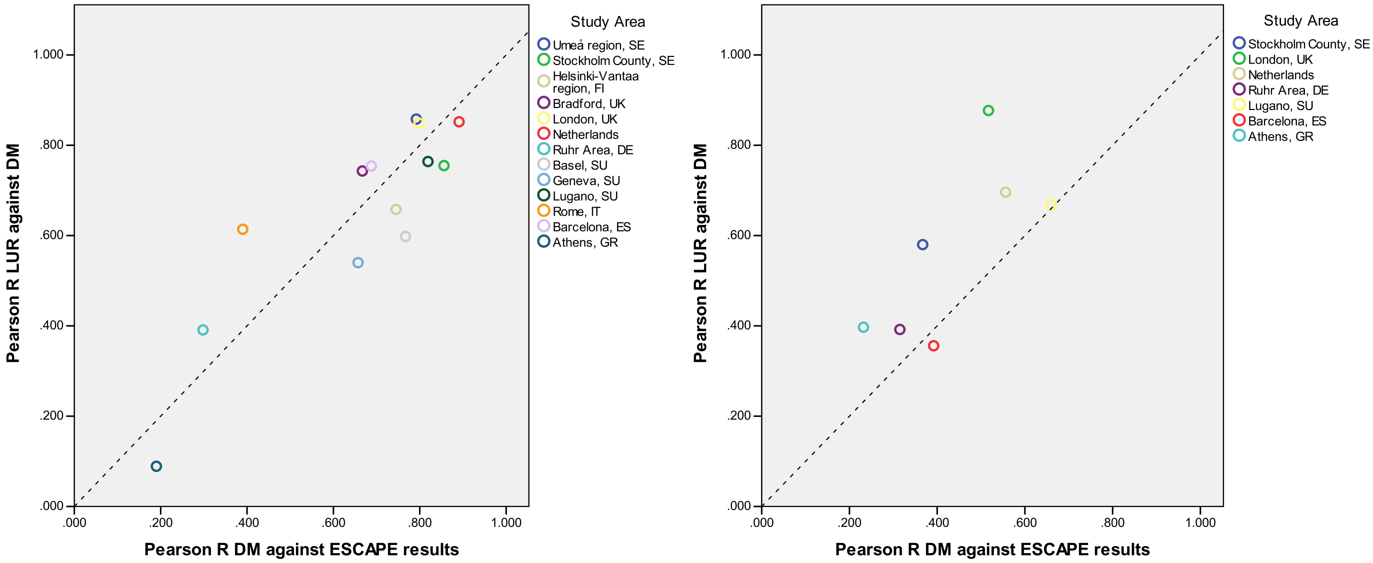

Fig. 3. Scatterplots of Pearson *R*'s between the LUR–DM and DM–ESCAPE comparisons for both NO_2 and PM_{10}.

4.2. Predictions of address level exposure

Despite the different modelling approaches of LUR and DM, the agreement in predicting NO_2 concentrations at cohort addresses was relatively good in most study areas. This is probably due to the importance of traffic affecting small-scale spatial variation of NO_2 in the predominantly urban areas. DMs have been developed extensively for modelling NO_2 traffic sources, and LUR models are most effective for modelling traffic because of the availability of predictor variables such as traffic intensity and distance to major roads. In a recent paper of PM composition, LUR models predicted traffic-related components (Black carbon, Cu, Fe) much better than elements for which non-traffic sources were dominant e.g. Ni, V and S (de Hoogh et al, 2013).

Compared to NO_2, the lower agreement between DM and LUR predictions for PM_{10} is likely due to a combination of random error related to the smaller spatial variation of PM_{10}, the lower predictive power of both models to predict concentrations and the smaller number of monitoring sites available to develop LUR models (20 PM versus 40 NO_2 in most areas). In general, the spatial variation of the measured PM and the predictions by both models was smaller than for NO_2, consistent with observations of a high regional background contribution to fine particle concentrations and a smaller influence of local sources (Eeftens et al., 2012b). In several areas, for both models, the spatial variation of PM was relatively small compared to the prediction errors as reflected by the root mean squared error.

Some of the differences in agreement between the two models at the cohort addresses were caused by the different model types. The Eulerian/CFD models used in the Ruhr, Rome and Athens areas correlated less strongly with LUR estimates than the Gaussian models for both NO_2 and PM_{10}. This is probably in part caused by the coarser resolution used by the Eulerian/CFD models compared to the Gaussian models which therefore better predicting receptor-specific concentrations as modelled in LUR. In epidemiological studies using the Ruhr Area model, the coarser resolution dispersion model was therefore supplemented with distance to major roads to account for the small-scale variation (Hoffmann et al., 2009). Fig. 3 illustrates that the agreement at the cohort addresses depended on how well the DM predicted the measurements at the ESCAPE monitoring sites. In addition to scale of the model, the complexity and size of the urban environment likely affect how well DM and LUR can predict spatial patterns. DMs for Mediterranean cities have some additional challenges such as describing local flows in coastal areas with complex terrain, as well as accounting for the intricacies of boundary layer development. In the case of Athens, emissions have exhibited large variability (inter-annual as well as spatial) over the last couple of years due to the effects of the economic crisis. Therefore, the amount of emission uncertainty involved in the Athens calculations has conceivably played a key role in the DM calculations. Interestingly, the LOOCV for the LUR models was also relatively low in Athens.

DM and LUR models generally explained a lower fraction of measured spatial variation of PM_{10} compared to NO_2 (Table 3 and Tables A.1 and A.2). The continental and regional scale chemical transport models commonly underestimated both the measured $PM_{2.5}$ and PM_{10} concentrations at the ESCAPE monitoring sites, which were designed to capture specifically the variation in traffic-related pollutants and therefore oversampled high traffic sites. Other reasons might include missing or under-estimated source categories (such as wild-land fires, desert dust, biogenic sources, non-exhaust emissions from traffic, shipping, fugitive dust, and sea salt), and by missing or inadequately treated processes in the models (such as the formation of secondary organic aerosols). Because of the urban character of the study areas, all the Gaussian models used measured concentration values at regional background stations; the above mentioned PM modelling deficit for chemical transport models does not therefore influence the predicted results in those cases. However, some dispersion models clearly underpredicted PM_{10} concentrations at the ESCAPE monitoring sites, in case of Stockholm, Ruhr Area and Athens, as can be seen based on the results presented in Fig. 2. For those models predicting average concentrations on a larger scale (i.e. Ruhr Area, Athens) this is a logical consequence of the fact that these models are not designed to predict concentrations at traffic sites. Consistently, the Ruhr Area Eulerian DM model predicted NO_2, PM_{10} and $PM_{2.5}$ better ($R = 0.53, 0.69, 0.68$ respectively) when the traffic sites were excluded.

As previously mentioned, LUR models are less effective for sources other than traffic (de Hoogh et al., 2013). The simple dispersion assumptions in LUR models apply better to traffic emissions than industrial point emissions, emitted at potentially hundreds of metres above ground. In Bradford, our NO_2 LUR model under-predicted at a number of residential addresses which were located in one residential area with a high activity of chemical processes. While this emission source was included in the ADMS-Urban model emission inventory, the LUR model for Bradford did not include an industry variable, because no ESCAPE monitoring sites were located near industrial sources.

A discussion about the Bland–Altman plots and Kappa-coefficients can be found in the Appendix (p. 4).

4.3. Implications for epidemiological studies

The overall high correlation between LUR and (fine scale) DM for NO_2 suggests that similar effects may be obtained if both approaches are applied in epidemiological studies to assess associations with health. However, if predicted concentration ranges differ, the size of the effect estimates may be different. The lower correlation for PM suggests that health effect estimates could be more different when applied in epidemiological studies. It remains important, however, to test directly in epidemiological studies differences in effect estimates related to exposure models. A recent study from Sellier et al. (2014) which applied four different exposure methods, including LUR and DM, to a cohort in Nancy and Poitiers (France), showed some differences in estimated health effect despite moderate to high correlations between NO_2 exposure estimates at the cohort level

The ESCAPE study was specifically designed to investigate health effects of long term air pollution exposure, using standardised LUR as the method of choice. Both LUR and DM are equally equipped to predict long term exposures, but an advantage of DM is that it can more easily deal with different time periods (e.g. hours, days, weeks, years and decades, also in retrospect) by using diagnostic or real-time emission and meteorological data. LUR models estimating daily concentrations have been developed and applied (Gryparis et al., 2014) but their evaluation and use are still limited. LUR model application is further restricted to the time period and geographical area of the monitoring campaign, although some recent studies suggest that LUR models in some circumstances can be transferred both back in time as well as geographically (Gulliver et al., 2013; R. Wang et al., 2013). An advantage of LUR models, however is that exposure estimates can be generated for absorbance, UFP, elemental composition (de Hoogh et al., 2013; Eeftens et al., 2012b) for which few dispersion models are available.

DMs can also be used for evaluating the contributions originating from various sources or source categories at selected locations. A specific strength of DM is its use for retrospective evaluations as well as for scenarios for the future. DM, however is also inherently source specific and as such requires several accurate input datasets like emission inventories, and ideally, pre-processed representative meteorological data, a thorough discussion of which has been presented by Kukkonen et al. (2012). Although the initial development of a LUR model takes some time, the subsequent application to residential addresses is fairly light in terms of computing power and time. DM on the other hand needs a lot more expertise to run and is relatively heavier in data demand and running time.

5. Conclusions

Dispersion model estimates for outdoor NO_2 with high spatial resolution showed, in most countries, high correlation with measured values and with the corresponding land-use regression estimates for cohort addresses. This implies that both methods may be useful for epidemiological studies of small-scale variations of outdoor combustion-related air pollution, typically from road traffic. The agreement for PM levels was considerably lower than for NO_2, probably reflecting smaller spatial variation, less precise source characterization and/or lack of related land use descriptors. The agreement between LUR and dispersion models with lower spatial resolution was reduced. These Eulerian/CFD DMs provide average concentrations in a small area, thus modelling a different aspect of person-specific exposure. The influence of data requirements and whether the methods tend to give different results in epidemiological studies need to be further explored.

Conflict of interest

All authors declare no actual or potential conflict of financial or other interests.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.envint.2014.08.011.

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