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Comparison of Land-Use Regression Modeling with Dispersion and Chemistry Transport Modeling to Assign Air Pollution Concentrations within the Ruhr Area

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Abstract: Two commonly used models to assess air pollution concentration for investigating health effects of air pollution in epidemiological studies are Land Use Regression (LUR) models and Dispersion and Chemistry Transport Models (DCTM). Both modeling approaches have been applied in the Ruhr area, Germany, a location where multiple cohort studies are being conducted. Application of these different modelling approaches leads to differences in exposure estimation and interpretation due to the specific characteristics of each model. We aimed to compare both model approaches by means of their respective aims, modeling characteristics, validation, temporal and spatial resolution, and agreement of residential exposure estimation, referring to the air pollutants $PM_{2.5}$, PM_{10}, and NO_2. Residential exposure referred to air pollution exposure at residences of participants of the Heinz Nixdorf Recall Study, located in the Ruhr area. The point-specific ESCAPE (European Study of Cohorts on Air Pollution Effects)-LUR aims to temporally estimate stable long-term exposure to local, mostly traffic-related air pollution with respect to very small-scale spatial variations ($\leqslant$100 m). In contrast, the EURAD (European Air Pollution Dispersion)-CTM aims to estimate a time-varying average air pollutant concentration in a small area (*i.e.*, 1 km^2), taking into account a range of major sources, e.g., traffic, industry, meteorological conditions, and transport. Overall agreement between EURAD-CTM and ESCAPE-LUR was weak to moderate on a residential basis. Restricting EURAD-CTM to sources of local traffic only, respective agreement was good. The possibility of combining the strengths of both applications will be the next step to enhance exposure assessment.

Keywords: air pollution; Land use regression; chemistry-transport dispersion-model

1. Introduction

A large number of epidemiological studies have shown associations between short-and/or long-term exposure to outdoor air pollution and adverse health effects [1]. Traditionally, adverse health effects of air pollution have been divided into effects of short-term variations in air pollution concentrations, mainly influenced by meteorology, and effects of long-term exposure to air pollution, where contrasts rely on spatial variation of air pollution concentrations. Early approaches on assessing exposure to air pollution used average air pollution concentrations of the nearest monitoring station as a surrogate of personal exposure, assuming homogeneity among air pollution concentrations within the area surrounding the monitoring station, or even within the whole city [2]. Considering short-term health effects in ecological time-series studies on air pollution and mortality, it seems reasonable to assume such a spatially-uniform temporal elevation or reduction in air pollution concentration because they are dependent on the underlying meteorological conditions. When considering long-term health effects on an individual basis, however, the spatial and spatio-temporal variations are of great importance given that outdoor air pollution concentrations vary on a small spatial scale, e.g., within 100 m of a busy road [3]. More recent epidemiological studies have, thus, approached such small-scale intra-urban variation of air pollution concentrations by using different types of models, such as Land Use Regression (LUR) models, Dispersion Models (DM), chemistry Transport Model Models (CTM), a combination of DM+CTM (DCTM), hybrid models, or other alternatives [4,5].

The LUR method, first developed by Briggs *et al.* [6] in the Small Area Variations In Air quality and Health (SAVIAH study), uses linear (least squared) regression models to predict monitoring air pollution data with Geographic Information System (GIS)-based data reflecting pollutant conditions. Compared to other approaches, LUR models were built to predict temporally-stable long-term air pollution concentrations applicable to the smallest spatial scale (point-specific), e.g., home residences.

DMs are in general mathematical simulation models to estimate air pollution concentrations by means of numerical descriptions of deterministic (physical, chemical, and fluid dynamical) processes of the dispersion of air pollutants in the ambient atmosphere, and typically include data on emissions, meteorological conditions, and topography [3].

CTMs model the variability in space and time of chemical concentrations in the atmosphere, using three-dimensional numerical models to simulate processes of emission, transport, chemical transformation, diffusion and deposition, using emissions, meteorological information, and land use as input. Most often DMs and CTMs (DCTM) are combined in practice, resulting in spatio-temporal estimations. Usually DMs and CTMs estimate air pollution concentrations on a coarser spatial scale compared to the point-specific LUR, e.g., a grid of 1 or 5 km^2.

LUR models were developed to estimate exposure concentration at the finest spatial resolution and have been increasingly used in epidemiological studies due to their relatively low cost and easy implementation, developed either on the basis of purpose-designed monitoring campaigns or routine monitoring measurements and appropriate geographic predictors of sources [7]. In contrast, DCTMs have been developed for air quality, *i.e.*, prediction, regulation and management, putting high demands on data requirements, costs and the complexity of modeling [6].

So far, only a few studies compared the performance of LUR and dispersion modeling for estimating exposure to nitrogen dioxide (NO_2). While some studies suggested that LUR models explained small-scale variations in air pollution concentrations as well or even better than various dispersion models [8–10], Beelen *et al.* [11] showed that the dispersion models performed better than LUR models regarding monitored and modeled concentrations on several validation sites. Most recently, de Hoogh *et al.* [12] investigated agreement between LUR and DM modeling approaches aiming to estimate residential exposure to NO_2 and particulate matter (PM) with an aerodynamic diameter $\leqslant$10 μm and $\leqslant$2.5 μm (PM_{10}, $PM_{2.5}$) within the European Study of Cohorts for Air Pollution Effects (ESCAPE). Comparisons across 4–13 cohorts, including the Heinz Nixdorf Recall (Risk Factors, Evaluation of Coronary calcium and Lifestyle) (HNR) study, located in the Ruhr area in Germany, yielded moderate to good correlations between LUR and DM (or DCTM) for NO_2 (0.39–0.90) and for

PM_{10} and $PM_{2.5}$ (0.23–0.81). However, single correlation coefficients for the HNR study were below 0.4 for all three pollutants [12], raising the question of comparability of the two different exposure modelling approaches. So far, most studies on the comparison of different modeling strategies focused on the residential agreement of estimated exposure concentrations, disregarding the potential reasons for the disagreement between different modelling approaches, as well as respective strengths and limitations. Although all exposure metrics are equally used as a surrogate of personal exposure in epidemiological studies, exposure modeling is strongly influenced by the spatial and temporal variation of exposure and exposure sources [5]. Furthermore, aims, application, input data but also the complexity of models might differ, yielding not only different exposure estimates but consequently different health effect estimates in terms of magnitude and/or statistical significance [5,13].

In the Ruhr area in Germany, the location of multiple epidemiological studies, e.g., the Heinz Nixdorf Recall study, air pollution concentrations have been modeled with a LUR model as part of the European Study of Cohorts for Air Pollution Effects (ESCAPE-LUR), as well as with a European Air Quality and Dispersion Model which is a DCTM (EURAD-CTM) as part of several research projects investigating health effects of residential air pollution exposure. In this article, we aim to compare the ESCAPE-LUR model and the EURAD-CTM model focusing on their respective strengths and limitations. To do so, we compare model approaches by means of their respective aim, application characteristics, validation, temporal, and spatial resolution and by means of residential agreement. In addition, we evaluated the agreement of modeled air pollution concentrations by EURAD-CTM and measured air pollution concentrations at ESCAPE-LUR monitoring sites for overlapping time windows. Air pollutants of interest are $PM_{2.5}$, PM_{10}, and NO_2.

2. Methods

2.1. Study Area

The Heinz Nixdorf Recall (Risk Factors, Evaluation of Coronary calcium and Lifestyle) (HNR) study area covers a region of approximately 600 km^2 and is located in the highly urbanized Ruhr Area in the west of Germany, including the cities of Mülheim, Essen, and Bochum. In addition to that the HNR study area is located within N3, one of the smallest sequential nests developed for the air pollution modelling purposes of EURAD-CTM. We used locations (x,y) (Gauss–Krüger coordinates) of 4809 residences, located within the HNR study area. According to the Ruhr Regional Association, land use in the area can be roughly divided into agricultural (~40%), built-up (~40%), and forest and other regions (e.g., water) (~20%) [14]. The population density of the Ruhr area is about 2100 inhabitants per 1 km^2, and in terms of traffic density the area is one of the densest in the whole of Europe (Figure 1). As an urban area, almost one fifth of the working population is occupied in the industrial sector. Among many industrial areas, the majority of steel and coal industry is located in Duisburg, in the west of the Ruhr area, including the biggest steelwork in Europe. Furthermore, Europe's largest inland harbor is located directly west of the study area in Duisburg. Intensive shipping takes place on the Rhine, which flows through Duisburg from south to north.

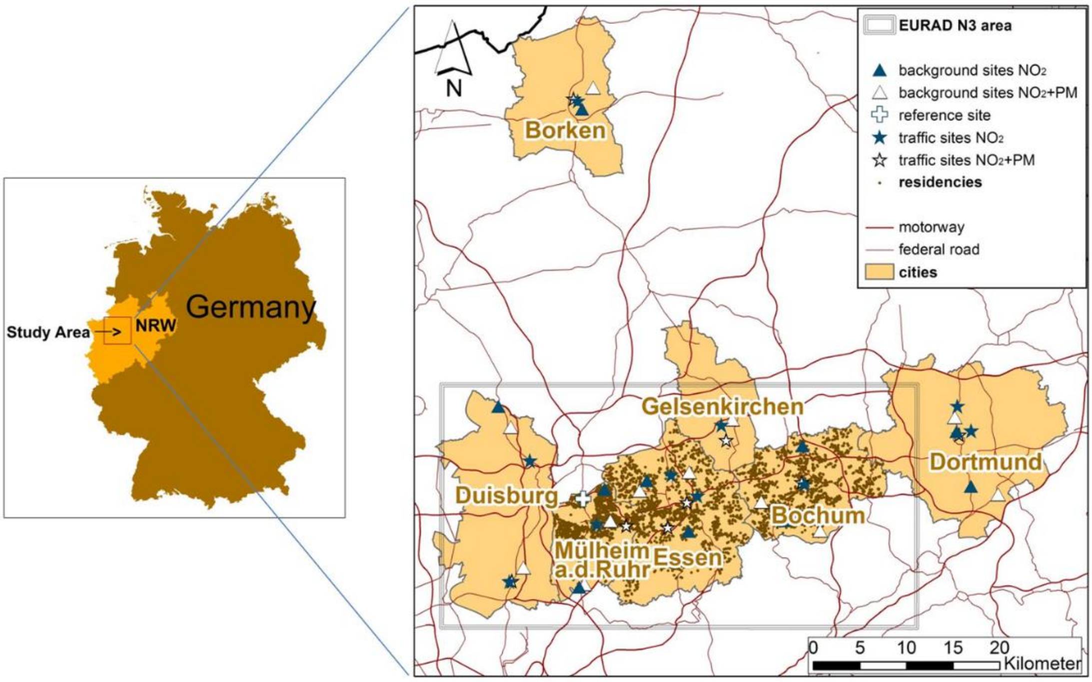

Figure 1. Study area, residences, and monitoring sites.

2.2. Exposure Assesment

2.2.1. EURAD-CTM

The EURAD-CTM model [15] is a validated time dependent three-dimensional chemistry transport model [16–19] developed to predict daily concentrations of air pollutants on a horizontal grid resolution of 1 km^2 (Table 1). The EURAD-CTM model system is a multi-layer, multi-grid model system for the simulation of transport, chemical transformation, and deposition of tropospheric constituents [20], and consists of five major parts (Figure S1): (1) the meteorological driver version 3 (MM5V3) [21]; (2) two pre-processors for preparation of meteorological fields and observational data; (3) the EURAD Emission Model EEM [22], and (4) the Chemistry Transport Model (CTM); including (5) a model for aerosol dynamics in Europe (MADE) [16,18,23,24]. An additional procedure includes data assimilation on an hourly basis, using routine measurement data of monitoring sites in North Rhine-Westphalia (NRW) provided by the local environmental agency: State Agency for Nature, Environment, and Consumer Protection (LANUV-NRW) [25–27] (intermitted 3d-var) (Figure S1). EURAD-CTM calculations are performed using a one-way nesting scheme to take long-range transport into account. Nested grid domains ranged from a European scale (N0: 125 km), to central Europe (N1: 25 km), to NRW (N2: 5 km) in Germany, to the Ruhr area (N3: 1 km), while the vertical resolution is the same for all model domains (40 m) ([18,20]). In addition to long-range transport, the formation of atmospheric gases and PM is also included in the model, *i.e.*, the formation of secondary particles in the atmosphere from primary emitted gaseous pollutants from NO_2, sulfur dioxide (SO_2), ammonia (NH_3), and Volatile Organic Compounds (VOC) during the transport [19]. Long-range transport and formation of secondary particles in the atmosphere can contribute considerably to the particle mass concentration in NRW and the Ruhr area, e.g., more than 50% [28]. The EURAD-CTM is driven by emissions due to anthropogenic and biogenic sources [29]. Anthropogenic emissions are taken from officially-available databases as EMEP-grid [30] for Europe and from the LANUV-NRW. The EURAD-CTM emission input is further structured with respect to different source categories according to the Selected Nomenclature for Sources of Air Pollution (SNAP-97) [31], including traffic, industry, and other source categories.

Table 1. Characteristics of the ESCAPE-LUR and EURAD-CTM approaches to estimate air pollution concentrations.

-	Land use regression (ESCAPE-LUR)	European Air Quality and Dispersion Chemistry Transport Model (EURAD-CTM)
Model Type	Linear regression model, to predict annual averages derived from selected monitored concentrations with land use data	Mesoscale chemistry transport model involving emissions, transport, diffusion, chemical transformation, wet and dry deposition, and sedimentation of gases and aerosols
Aim & Application	Estimation of long-term traffic-related air pollution for population-based exposure studies and epidemiological health outcome analyses	1) Air pollution modeling (forecasts, episode analysis, trend analysis, reduction scenarios) and Chemical data assimilation studies for Europe, Central Europe and several German States; 2) Exposure estimation in population-related exposure studies
Model Input	1) Data: • Annual mean AP concentration (for details see Table S1); • Land use density in 100, 300, 500, 1000, and 5000 m buffers: ○ Industry ○ Seaport ○ urban green ○ semi-natural ○ forested areas ○ number of inhabitants • Traffic data in 25, 50, 100, 300, 500, and 1000 m buffers: ○ distance ○ (heavy) traffic intensity on the nearest road and nearest major road ○ (heavy) traffic load on all roads and major roads)	1) Data: • Model area projection topography • Land use • Meteorological initial and boundary values • Anthropogenic emission data (according to the Selected Nomenclature for Sources of Air Pollution (SNAP-97)) • Chemical initial and boundary values, • Long-range transport, • Photolysis frequencies. 2) Procedures (Figure S3): • Mesoscale meteorological model (MM5) driven by global meteorological fields provided by NCEP (http://www.ncep.noaa.gov/), • EPC, anthropogenic and non-anthropogenic emission modules (EEM-A, EEM-B), • Aerosol dynamics module (MADE), • Data assimilation [a]
Modelled Air Pollutants	**$PM_{2.5}$, PM_{10}, NO_2** (additional pollutants: $PM_{2.5}$ absorbance, PM coarse, NO, NOx)	**$PM_{2.5}$, PM_{10}, NO_2** (additional pollutants: PM_1, O_3, SO_2, CO, PNC, NH_4, NO_3, SO_4, BC, EC)
Temporal Resolution (Output)	Yearly mean concentration (October16, 2008 until October 15, 2009)	Any temporal resolution > day within October 2000 until December 2003 and January 2006 until December 2008 is possible; e.g., 7-,14-, 21-,28-,91-,182-, and 365-day mean concentration
Model Validation	a) Goodness of fit (cf. Table S2): $PM_{2.5}$ (R^2 = 0.85), PM_{10} (R^2 = 0.66), NO_2 (R^2 = 0.88) b) Leave-one-out cross-validation: $PM_{2.5}$ (R^2 = 0.74), PM_{10} (R^2 = 0.59), NO_2 (R^2 = 0.82)	Validation for daily mean concentration in N3 area with routine measurements (mean bias, correlation); year: a) Before data assimilation: PM_{10} (−6.5, 0.45); 2006 NO_2 (4.0, 0.39); 2007 b) After data assimilation PM_{10} (−0.9, 0.93); 2006 NO_2 (0.6, 0.95); 2007
Spatial Resolution	Point-specific	1 km × 1 km grid
Additional Features	1) XRF-Model for air pollutant constituents 2) Back-extrapolating back in time and for specific time windows	Source-specific air pollutant concentrations (only local traffic (TRA), only local industry (IND))

[a] only for PM_{10} and NO_2 for the considered time period.

Output of the EURAD-CTM calculations consists of chemical compounds, such as atmospheric particle mass, number density, and particle size distribution, as well as concentration of atmospheric gases, photo oxidants, and a set of volatile organic compounds on an hourly basis for each grid. EURAD-CTM estimates of PM_{10} and NO_2 concentrations are assimilated using measurements from all available routine monitoring sites within the region of interest. For the Ruhr area there exists a maximum of ten monitoring sites, including different air pollution data bases [25]. Using ArcView 9.2, location of residences were assigned to a 1 × 1 km^2-grid and then matched to the corresponding grid-based air pollutant concentration, allowing both short-term (daily mean concentrations) and long-term (annual mean concentrations) assignment of exposure. The basis of daily mean concentration allows us to calculate exposure for any temporal resolution with a minimum of one day. Model runs for the EURAD-CTM within N3 were done for the examination periods of the HNR study (2000–2003 and 2006–2008). Thus, we are able to assign exposure concentrations of yearly-mean concentrations for the years 2001, 2002, 2003, 2006, 2007, and 2008 and personalized exposure concentrations of 1-, 7-, 28-, 91-, 182-, and 365-day mean concentrations prior to the date of examination.

As an add-on feature it was possible to model source-specific Air Pollution (AP) concentration with EURAD-CTM [28]. Briefly, within EURAD-CTM we estimated AP concentration suppressing local sources within the smallest grid domain (N3), such as traffic and industry by setting to them to zero (AP_{noTRA} or AP_{noIND} respectively). We then calculated local traffic-specific or industry-specific AP by taking the difference $AP_{TRA} = AP - AP_{noTRA}$ or $AP_{IND} = AP - AP_{noIND}$, respectively. In earlier studies, we applied this method to compare the health effects of PM, emitted from local traffic and local industrial sources within the Ruhr area on levels of highly-sensitive C-reactive protein, a marker of systemic inflammation [32].

2.2.2. ESCAPE-LUR

LUR models were developed to estimate temporally-stable spatial-variant concentrations of long-term exposure to traffic-related air pollutants as part of the ESCAPE study (Table 1). Following the definition of LUR describes a standardized model building procedure developed within the ESCAPE study, here the ESCAPE-LUR. The ESCAPE-LUR defines a linear prediction model for an air pollutant concentration, including annual mean air pollution concentrations as a dependent variable and geographic data on traffic, industry, and population density as potential predictors (independent variables). Predictor data were collected in a Geographical Information System (GIS), based on CORINE 2000 definitions [33]. The procedure of model development was standardized within the ESCAPE study and included a forward selection of predictors based on the incremental improvement in R^2 [34–36]. A predictor was added if addition of the predictor yielded an improvement of R^2 by more than 1%, if the coefficient conformed to the pre-specified direction, and if the direction of previously selected predictors did not change. In addition, predictors with a p-value > 0.1 were removed, while predictors with a variance inflation factor (VIF) > 3 and Cook's Distance (Cook's D) >1 were further investigated. To avoid extrapolation, estimated concentrations were truncated at the highest observed value. Annual air pollution concentrations were based on a measurement campaign in the study area of interest, including three periods of a 14-day measurement to cover all seasons (cold, warm, and one intermediate temperature season) from October 2008 until October 2009. The reason for the choice of 14-days was the settings design of the ESCAPE-LUR measurement campaign, which was conducted with discontinuous particle measurement devices (Harvard impactors). Measurements were conducted at 20–40 monitoring sites, placed at locations which were characteristic of traffic and background pollutant concentrations to measure PM (at 20 sites) and NO_2 (at 40 sites) (Figure 1, Table S1). One additional background reference site was chosen to measure PM and NO_2 continuously during a complete year (starting in October 2008) so that all discontinuous site-specific measurements could be adjusted to derive a long-term annual average. Measurement data from the reference site was only used for adjustment and not for ESCAPE-LUR model development. A separate LUR model was developed for each air pollutant and validated via Leave-One-Out Cross Validation (LOOCV), excluding one monitoring site at a time. Other choices of model validation are possible, e.g., hold-out cross validation, which has recently been proposed to perform better [37]. However, in this manuscript we hold onto the ESCAPE-LUR.

Since ESCAPE included two cohorts located within NRW, namely the HNR study and the Study on the influence of air pollution on lung function, inflammation, and aging (SALIA), the ESCAPE-LUR measurement campaign was combined for both studies and ranged from the urban Ruhr area to the more rural city of Borken (Figure 1) [34,36]. ESCAPE-LUR for $PM_{2.5}$ included heavy traffic load (1 km buffer), industry (5 km buffer), population density (1 km buffer), and the x-coordinate of the location of interest as predictors with an explained variance of $R^2 = 0.85$ (LOOCV-$R^2 = 0.74$) (Table S2) [34]. ESCAPE-LUR for PM_{10} included heavy traffic load (1 km buffer) and population density (1 km buffer) with an explained variance of $R^2 = 0.66$ (LOOVC-$R^2 = 0.59$) (Table S1) [34], ESCAPE-LUR for NO_2

included industry (5 km buffer), population density (100 m buffer), inland or seaport (5 km buffer) and traffic load (100 m buffer) with an explained variance of $R^2 = 0.88$ (LOOVC-$R^2 = 0.82$) (Table S1) [36]. (Heavy) traffic load referred to total (heavy-duty) traffic load of all roads in a buffer (sum of (traffic intensity × length of all segments)), industry referred to industrial, commercial, and transport units in a certain buffer; inland or seaport referred to the respective area within a buffer and population density to the number of inhabitants in a certain buffer. Uncertainty was evaluated as residuum's mean squared error in the LOOCV-approach, which was 0.61 for $PM_{2.5}$, 1.44 for PM_{10}, and 3.19 for NO_2.

Based on the coordinates of residence, located within the study area, annual mean concentrations were estimated using the ESCAPE-LUR prediction models and the relevant GIS predictors. In order to estimate AP concentration back in time, LUR modeling offers the method of back-extrapolation using a ratio or absolute difference method. Briefly, routine monitoring data should be available in order to account for differences of AP concentrations back in time [38]. Within the ESCAPE study, back-extrapolated AP estimations referred to a two year average (± 365 days of the examination day) in order to avoid any time-specific outliers. An additional feature offered by ESCAPE-LUR is the possibility to estimate exposure concentration as an average per month or trimester, e.g., before pregnancy, which might be of interest when investigating birth cohorts.

2.3. Statistical Analysis

Conducted statistical analysis referred to air pollutants $PM_{2.5}$, PM_{10}, and NO_2, estimated using the EURAD-CTM and the ESCAPE-LUR model. First, we described EURAD-CTM grid-based concentrations for the whole HNR study area for the years 2001–2003 and 2006–2008 by mean and standard deviation (mean ± SD) as well as minimum and maximum (Min, Max). Secondly, we described residence-based exposures derived from the EURAD-CTM and from the ESCAPE-LUR by mean ± SD (Min, Max) and Person's correlation coefficients for the most closely matched annual time-window: year 2008 for EURAD-CTM *vs.* annual mean ESCAPE-LUR (*i.e.*, based on measurements from October 2008 until October 2009). Considered air pollutants were $PM_{2.5}$, PM_{10}, and the gas NO_2. In addition, we calculated Spearman's correlation coefficient between 14-day mean air pollution concentrations measured at ESCAPE measurement sites (traffic and background) and 14-day mean air pollution concentrations calculated by EURAD-CTM for the grid cells that included an ESCAPE measurement site within the time period of October 2008–December 2008.

To evaluate an overall agreement between routinely measured air pollution concentrations, we compared annual mean concentrations of three routine monitoring stations provided by LANUV, located within the Ruhr area, and thus within EURAD specific grid cells (gc), with annual estimated air pollution concentrations estimated by EURAD-CTM and ESCAPE-LUR. Details of routine measurement stations are given in Table S3. Referred monitoring sites are the above mentioned reference site in Mülheim-Styrum (STYR) (gc: 679), an additional background site, located in Essen-Vogelheim (EVOG) (gc: 942), and one traffic site, located at a highly trafficked road in Essen (VESN) (gc: 690). For the comparison with the EURAD-CTM we considered annual mean concentrations from January 1, 2008 until December 31, 2008, while for the comparison with the ESCAPE-LUR we considered annual means from October 16, 2008 until October 15, 2009 in order to match the time window of the ESCAPE measurement campaign. Annual mean concentrations modeled by the ESCAPE-LUR referred to the location (coordinate points) of monitoring sites. In addition to that we calculated Pearson's correlation coefficients between daily measurements of LANUV monitoring sites and daily estimations by EURAD-CTM for the year 2008.

With regard to different temporal resolution, we compared EURAD-CTM air pollution concentration estimates to measured air pollution concentrations on a monthly basis to yearly mean concentrations (2006, 2007, and 2008) estimated by EURAD-CTM in two of the above mentioned grid cells (679 and 690). In contrast we visualized time-dependent measurements of the two corresponding routine monitoring sites (STYR and VESN) on a monthly basis as well as the temporally stable air pollution concentration estimated by ESCAPE-LUR for the specific locations of routine monitoring sites. For ESCAPE-LUR values we used the original, not back-extrapolated values, since during the study period of 2006–2008, no substantial changes of long-term air pollutant concentrations were observed at the routine monitoring sites, therefore not having a meaningful influence on the back-extrapolated values.

With respect to the additional feature of source-specific estimation of air pollution concentrations, we further investigated the correlation of traffic-specific and industry-specific EURAD-CTM (EURAD-CTM_{TRA} and EURAD-CTM_{IND}, respectively) and ESCAPE-LUR concentrations at residence as well as at locations of specific ESCAPE measurement sites.

Statistical analysis were carried out with the statistical software R version 3.1.3 (2015-03-09) [39].

3. Results and Discussion

3.1. Comparison of Residence-Based EURAD-CTM and ESCAPE-LUR

Residence-based air pollution concentrations (for 4809 residences within the HNR study area) estimated by EURAD-CTM as yearly-mean air pollution concentrations for the years 2001–2003 (not including 2000 since modeling did not start before October 2000), 2006–2008 and estimated yearly mean air pollution concentrations by ESCAPE-LUR as well as back-extrapolated ESCAPE-LUR air pollution concentration estimates are presented in Table 2 for $PM_{2.5}$, PM_{10}, and NO_2 and visualized in Figure 2 for the year 2008 (EURAD-CTM) and October 2008–October 2009 (ESCAPE-LUR), respectively.

Table 2. Description of residence-based air pollutant exposure estimates $PM_{2.5}$, PM_{10}, and NO_2 from EURAD-CTM and ESCAPE-LUR for 4809 residences within the HNR study area.

–	$PM_{2.5}$ Mean ± SD (Min, Max)	PM_{10} Mean ± SD (Min, Max)	NO_2 Mean ± SD (Min, Max)
EURAD-CTM (μg/m³)			
2001 year-mean	16.6 ± 1.5 (14.0, 21.6)	21.2 ± 2.9 (17.0, 30.1)	42.2 ± 4.2 (28.2, 55.4)
2002 year-mean	16.8 ± 1.4 (14.3, 21.2)	20.4 ± 1.9 (16.7, 27.0)	39.3 ± 3.8 (27.5, 50.2)
2003 year-mean	18.2 ± 1.4 (15.5, 22.7)	22.4 ± 3.3 (17.8, 32.4)	42.7 ± 4.1 (30.1, 56.1)
2006 year-mean	16.2 ± 1.3 (13.9, 21.2)	21.0 ± 3.7 (16.5, 34.2)	40.0 ± 4.8 (27.1, 57.2)
2007 year-mean	15.7 ± 1.3 (13.4, 20.3)	19.8 ± 2.9 (15.7, 30.8)	37.7 ± 4.5 (26, 53.7)
2008 year-mean	14.6 ± 1.1 (12.5, 19.0)	18.0 ± 2.3 (14.9, 25.1)	37.5 ± 3.9 (26.3, 47.9)
ESCAPE-LUR (μg/m³)			
back-extrapolated (2-year averages)	–	30.3 ± 2.1 (25.5, 38.7)	30.5 ± 5.0 (19.3, 62.0)
Year 2008–2009	18.4 ± 1.0 (16.0, 21.4)	27.7 ± 1.8 (23.9, 34.7)	30.1 ± 4.9 (19.8, 62.4)
Difference (μg/m³)			
ΔESCAPE-LUR (2008–09) EURAD-CTM (2008)	3.7 ± 1.3 (−0.7, 7.0)	9.8 ± 2.4 (0.9, 16.5)	−7.4 ± 4.9 (−26.8, 18.9)

On a residential basis, estimated $PM_{2.5}$ and PM_{10} concentrations revealed a consistent decline since 2006 (Table 2). Considering the back-extrapolated ESCAPE-LUR and ESCAPE-LUR, we also observed a decline over time. Observed declines are accounted for by ongoing nation- and state-wide air quality regulations.

Comparing EURAD-CTM (2008) and ESCAPE-LUR (2008–09), however, we saw that the overall mean of the ESCAPE-LUR was considerably higher compared to the overall yearly-mean of EURAD-CTM ($\Delta PM_{2.5}$ = 3.7 ± 1.3 μg/m^3 and ΔPM_{10} 9.8 ± 2.4 μg/m^3, respectively). Ranges for $PM_{2.5}$ estimated by EURAD-CTM were slightly smaller than estimated by ESCAPE-LUR (5.4 *vs.* 6.5 μg/m^3), while ranges for PM_{10} were more similar for both models (10.8 *vs.* 10.0 μg/m^3). Smaller ranges of air pollution concentrations from EURAD-CTM are not unexpected due to the smoothing pattern within 1 km^2.

Explanations for the difference in mean concentrations for PM might be a consequence of the finer spatial resolution of the ESCAPE-LUR, since high exposure peaks in a very close proximity to busy roads are better captured with this model than with the EURAD-CTM, especially considering that residences are usually located close to the roads and not randomly distributed across a certain area.

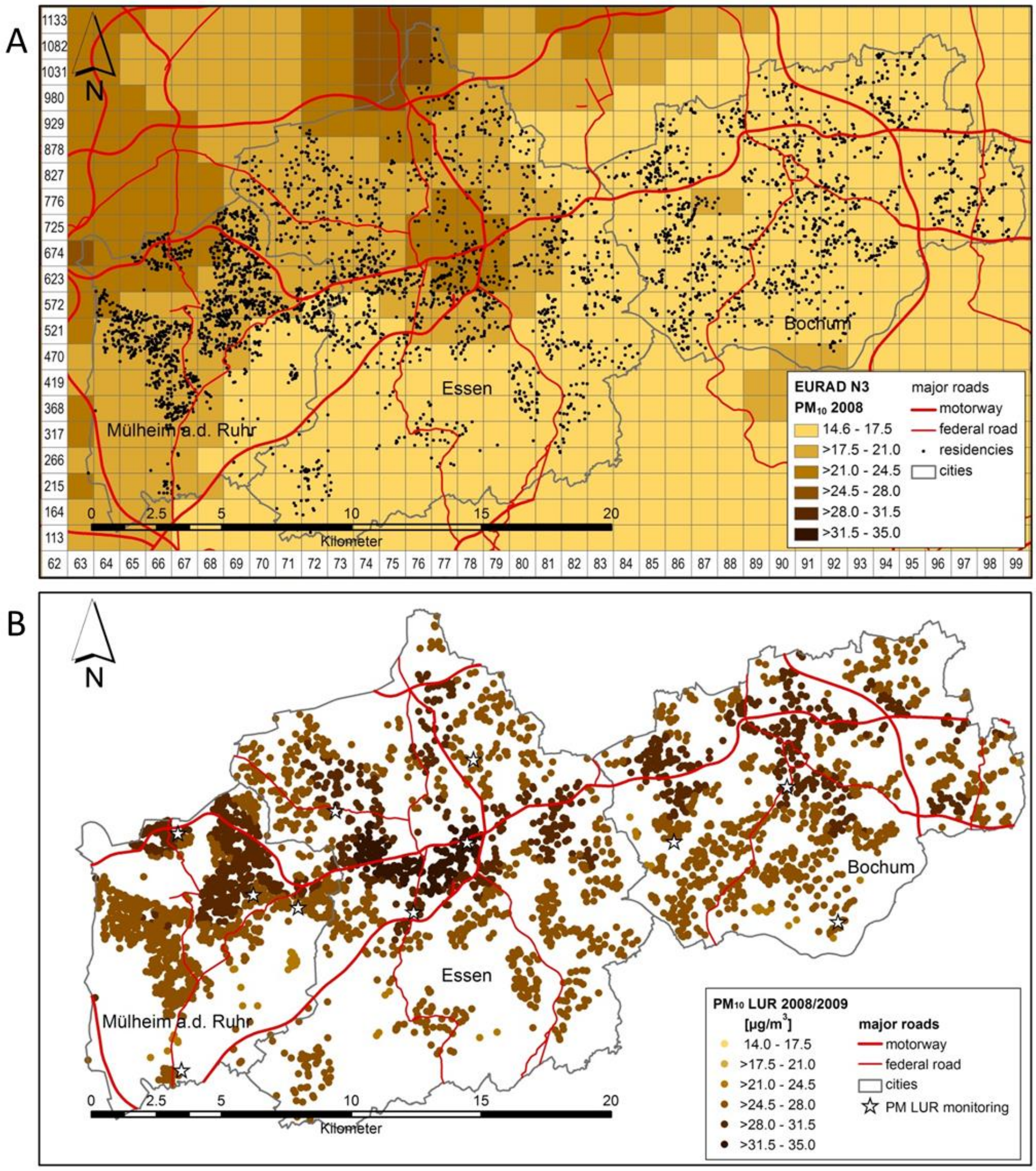

Figure 2. Spatial distribution of EURAD-CTM (1 km^2, yearly mean 2008, (**A**)) and ESCAPE-LUR (yearly mean October 2008–October 2009, (**B**)) at 4809 residences within the HNR study area for PM_{10}.

Pearson's correlation coefficients between models were rather weak for both, $PM_{2.5}$ and PM_{10}, with 0.33. This rather weak correlation has been reported earlier [12] and is not unexpected due to the different spatial resolution but also due to the different spatial distribution of PM concentrations for the two modelling approaches within the study area (Figure 2 and Figure S2): while we observed a west-to-east gradient for EURAD-CTM with higher concentrations in the west, estimated concentrations of ESCAPE-LUR revealed only a slight west-to-east gradient, which was prominently overlapped by an additional decreasing north-to-south and local hot spots, e.g., in Essen at a motorway intersection. In our study area the decreasing west-to-east gradient mirrors the distribution of industrial locations, e.g., metallurgical-industry and Europe's largest inland harbour in Duisburg, located to the west of the study area (Figure 1), as well as transported emissions from other countries in the west of study area, e.g., the Netherlands or Great Britain. The decreasing north-to-south gradient on the other hand is consistent with the population density and the location of major arterial roads within our study area [32].

NO_2 concentrations estimated by EURAD-CTM showed an overall decrease between 2001 with 42.2 $\mu g/m^3$ and 2008 with 37.7 $\mu g/m^3$, while a change between the ESCAPE-LUR and the back-extrapolated ESCAPE-LUR was not observed. Yet, in contrast to PM, temporally-stable NO_2 concentrations estimated by ESCAPE-LUR were systematically lower than estimated by EURAD-CTM ($\Delta NO_2 = -7.4 \pm 4.9$ $\mu g/m^3$). One explanation for this difference could be a misrepresentation of industrial sources within the ESCAPE modeling approach: "industry" referred to industrial, commercial and transport units in a certain buffer, giving no information of the emission of such sources. Ranges of concentrations, however, were twice as big for the ESCAPE-LUR compared to the EURAD-CTM (42.4 *vs.* 21.9 $\mu g/m^3$), probably driven by greater small-scale variations due to point-specific estimates and the consideration of traffic load within a buffer of 100 m. Unlike spatial gradients for $PM_{2.5}$ and PM_{10}, we observed a more pronounced northwest-to-southeast-gradient for EURAD-CTM for NO_2, while the distribution of NO_2 by ESCAPE-LUR did not reveal a clear gradient, but local hot spots near major roads or motorway intersections (Figure S2). Similar to PM, correlation between EURAD-CTM NO_2 and ESCAPE-LUR NO_2 was rather weak with a correlation coefficient of 0.4.

3.2. Comparison of Estimated and Measured Air Pollution Concentrations

3.2.1. Comparison between 14-Day Mean ESCAPE-LUR Measurements and EURAD-CTM Estimates

In order to evaluate EURAD-CTM estimates we compared estimated 14-day mean AP concentrations by EURAD-CTM to available 14-day measurements taken during the ESCAPE measurement campaign. Descriptive statistics and correlation coefficients of these 14-day mean measured air pollution concentrations at ESCAPE measurement sites (background, traffic (*cf.* Table S2), and both) and the respective 14-day mean air pollution concentrations estimated by EURAD-CTM in the corresponding grid cells are shown in Table 3 for air pollutants $PM_{2.5}$, PM_{10}, and NO_2.

Table 3. Description of 14-day mean measured air pollution concentrations at ESCAPE measurement sites (background and/or traffic) and 14-day mean air pollution concentration estimations of EURAD-CTM in the corresponding grid cells for $PM_{2.5}$, PM_{10}, and NO_2.

Background	**ESCAPE Site ($\mu g/m^3$)**	**EURAD-CTM ($\mu g/m^3$)**	**Spearman Correlation Coefficient (*r*)**
	Mean ± SD	**Mean ± SD**	
$PM_{2.5}$ (N = 9)	17.78 ± 2.40	19.80 ± 5.80	0.34
PM_{10} (N = 9)	26.12 ± 4.70	23.29 ± 5.98	0.93
NO_2 (N = 16)	37.85 ± 6.21	50.82 ± 10.07	0.34
traffic			
$PM_{2.5}$ (N = 6)	19.75 ± 3.75	21.78 ± 6.96	0.43
PM_{10} (N = 6)	29.26 ± 4.95	26.97 ± 7.68	0.37
NO_2 (N = 13)	50.43 ± 9.83	58.04 ± 10.33	0.60
Background + traffic			
$PM_{2.5}$ (N = 15)	18.57 ± 3.05	20.59 ± 6.13	0.45
PM_{10} (N = 15)	27.37 ± 4.89	24.77 ± 6.71	0.77
NO_2 (N = 29)	43.49 ± 10.13	54.06 ± 10.65	0.55

Overall, 14-day mean EURAD-CTM estimates for $PM_{2.5}$ are slightly higher than mean of 14 daily measurements at the ESCAPE sites, while EURAD-CTM estimates for PM_{10} are slightly lower and EURAD-CTM estimates for NO_2 are considerably higher, especially regarding the ESCAPE background site (Table 3).

The highest correlation coefficient (*r*) was observed for PM_{10} between EURAD-CTM and ESCAPE background sites (r = 0.93), while the lowest correlation was observed for PM_{10} between EURAD-CTM and ESCAPE traffic sites (r = 0.37). This finding is not unexpected, regarding the aim, input, and construction of the two modeling approaches (Table 1): the EURAD-CTM aims to assess an average concentration in a 1 km^2 grid cell, taking into account long-range transport rather than locally-emitted pollution, in contrast to the ESCAPE-LUR, which was specifically designed to assess mostly traffic-related differences in exposure concentration. For $PM_{2.5}$, however, we did not observe a clear distinction between background and traffic sites, whereas correlation coefficients for NO_2 were higher between EURAD-CTM and ESCAPE traffic sites (r = 0.60) than between EURAD-CTM and ESCAPE background sites (r = 0.34). One reason for the low to moderate correlation between $PM_{2.5}$ modeled by EURAD-CTM and $PM_{2.5}$ measured at ESCAPE sites could be the lack of the assimilation procedure within EURAD-CTM, since $PM_{2.5}$ has only been measured at routine monitoring sites since 2009. So, for the considered period of time, estimated $PM_{2.5}$ was only assimilated indirectly taking a (constant) proportion of PM_{10} and $PM_{2.5}$ into account.

Overall, correlations between EURAD-CTM estimates and measured concentrations at all ESCAPE measurement sites were moderate for $PM_{2.5}$ (r = 0.45) and NO_2 (r = 0.55), and high for PM_{10} (r = 0.77) and, therefore, slightly better than comparing residence-based modeled air pollution concentrations between EURAD-CTM and ESCAPE-LUR.

3.2.2. Comparison between Routinely-Monitored and Estimated Air Pollution Concentrations

Overall correlations between daily measurements at routine monitoring sites and EURAD-CTM estimations over one year (2008) were strong for PM_{10} and NO_2 (>0.8) and moderate for $PM_{2.5}$ (0.66–0.74) for both, background and traffic monitoring site (Table 4). This finding is a consequence of the assimilation procedure within EURAD-CTM for PM_{10} and NO_2.

Taking into account absolute annual values, we observed several findings: annual averages for January 2008 until December 2008 differ considerably from annual averages from 16 October 2008 to 15 October 2009 (ESCAPE measurement period), for PM (Table 4). Generally, PM concentrations throughout Germany were at a minimum in 2008, as reported by the Federal Environment Agency [40]. This finding points to the importance of a fine temporal resolution even in medium- and long-term exposure estimations.

Considering uncertainty, the EURAD-CTM estimations underestimated PM and overestimated NO_2 at background monitoring sites, while the ESCAPE-LUR estimations agreed well for $PM_{2.5}$ (all sites) and PM_{10} (background sites), but tended to underestimate NO_2 concentrations considerably (Table 4). The latter is supported by mean squared errors of the LOOCV, which were remarkably higher for NO_2 than for PM. Furthermore, we observed considerable disagreement between predicted ESCAPE-LUR PM_{10} and measured PM_{10} at the routine monitoring traffic-site. This finding might be a consequence of the disagreement between PM_{10} measured at the routine monitoring site and the measured PM_{10} at the closest ESCAPE site (26.64 *vs.* 32.70 $\mu g/m^3$), which were located only 2.2 m away from each other.

Table 4. Yearly mean air pollution concentrations measured at routine monitoring sites (background (BG) and traffic (TRAFFIC)), provided by LANUV, modeled by EURAD-CTM (for the respective grid cell), modeled by ESCAPE-LUR (at the location of the routine monitoring sites) and measured adjusted yearly mean at the closest ESCAPE site plus Pearson's correlation coefficient between LANUV daily measurements and EURAD-CTM daily estimations for $PM_{2.5}$, PM_{10}, and NO_2.

Air Pollutant (μg/m^3)	LANUV Monitor (2008)	EURAD-CTM (2008)	Pearson Correlation Coefficient (LANUV*EURAD-CTM)	LANUV Monitor (October 2008–October 2009)	ESCAPE-LUR Prediction (October 2008-October 2009)	Closest ESCAPE-Measurement Site
Mülheim-Styrum (BG) (grid cell: 679)						
$PM_{2.5}$	17.90	16.33	0.66	20.71	19.50	19.00 [a]
PM_{10}	25.24	23.21	0.88	28.20	28.86	29.00 [a]
NO_2	34.17	39.33	0.80	34.67	31.42	33.00 [a]
Essen-Vogelheim (BG) (grid cell: 942)						
$PM_{2.5}$	22.08	16.21	0.74	20.18	19.31	18.50 [b]
PM_{10}	27.66	23.79	0.81	27.32	26.64	26.40 [b]
NO_2	35.17	41.56	0.76	35.70	28.75	53.30 [c]
Essen-Ost city (TRAFFIC) (grid cell: 690)						
$PM_{2.5}$	20.08	14.72	0.69	20.51	21.05	20.90 [d]
PM_{10}	26.61	23.77	0.81	26.64	33.38	32.70 [d]
NO_2	46.36	44.97	0.87	47.65	42.01	43.50 [d]

[a] 6.7 m; [b] 2665.0 m; [c] 4060.1 m, [d] 2.2 m.

3.3. Temporal Resolution of Air Pollution Concentrations

Regarding different years (2006–2008) we saw a weak time-dependent decline in PM concentrations (Table 2), in line with the observed overall decline in PM concentrations from the year 2001 to 2008 within the HNR study area [29]. To examine the temporal resolution on a monthly basis, Figure 3 and Figure S3 present monthly distributions of EURAD-CTM estimated air pollution concentrations of PM_{10}, $PM_{2.5}$, and NO_2 respectively, in two grid cells, including one background grid cell (679) and one traffic routine monitoring site grid cell (690), presenting spatial variation. For the purpose of comparison, yearly mean air pollution concentrations estimated with EURAD-CTM for the two grid cells as well as the temporally-stable ESCAPE-LUR air pollution concentrations estimated at the locations of the monitoring sites, and monthly-based measured air pollution concentration at routine monitoring sites are presented as lines. Overall, we observed strong seasonal variation (high in winter and low in summer) for estimated EURAD-CTM air pollution concentrations and measured air pollution concentrations, which cannot be detected when using the temporally stable ESCAPE-LUR estimates. While ESCAPE-LUR estimates are primarily designed to yield long-term exposure estimates without temporal resolution, the integration of other measurements (*i.e.*, from routine monitoring sites), or other measurement periods (e.g., three month instead of one year), can be used to derive LUR-data for the analysis of medium-term health effects [41], although not covered in this manuscript.

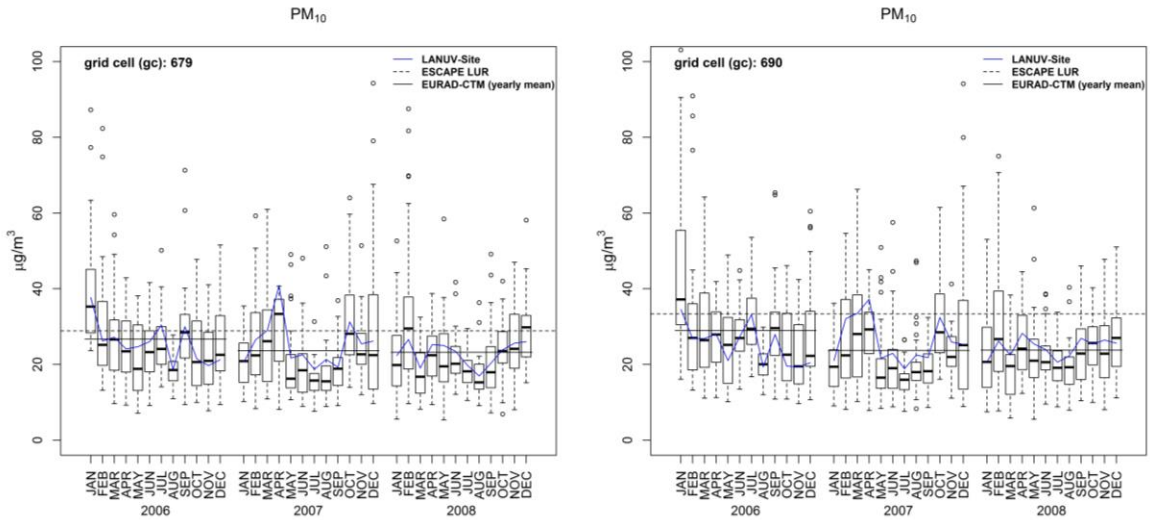

Figure 3. Box plots of air pollution concentrations of PM_{10} over time for two grid cells (gc), representing background (gc: 679) and traffic (gc: 690), estimated by EURAD-CTM on a monthly and yearly basis, long-term ESCAPE-LUR estimation and measured at monitoring sites on a monthly basis (median per month).

The seasonal patterns differed slightly across years and air pollutants (Figure 3 and Figure S3). Reasons for such differences might be specific meteorological conditions during the observation period as well as different chemical processes differentially influencing the concentration of the examined air pollutants, e.g., regarding transport, deposition or physical and chemical aging. These observed seasonal changes underscore the importance of time-dependent air pollution models for the analysis of short- and medium-term health effects. When using a LUR for short- and medium-term exposures, a finer temporal resolution can be achieved using back-extrapolation based on routine monitoring sites, as has been applied for birth outcomes in the framework of ESCAPE [41]. Furthermore, estimated $PM_{2.5}$ by EURAD-CTM, although following the seasonal pattern of measured $PM_{2.5}$, was considerably under-estimated, reflecting the lack of data assimilation within this modeling procedure. In contrast to the temporal variation over the considered time period, the spatial variation, presented by the two locations of a background and traffic site, is considerably smaller. This finding is in line with earlier findings, indicating a slightly higher temporal, than spatial, variation of particle number concentrations within the Ruhr area [42].

3.4. Source-Specific EURAD-CTM

Estimated local traffic-specific (TRA) and local industry-specific (IND) air pollution concentrations take up only a small amount of all sources: for $PM_{2.5}$ local traffic takes up 3.4% and local industry 9.6%; for PM_{10} it is 2.7% and 10.5%, respectively, and for NO_2 it is 21.4% and 2.4%, respectively. Correlation coefficients between PM concentrations, including all sources and including only local traffic, were weak (0.34–0.43), while all-sources PM and industry-specific PM correlated well (0.73–0.96) (Figure 4). Correlation coefficients for NO_2 were, in contrast to PM, higher between all sources and local-traffic (0.63) and lower for industry-specific (0.44). The rather small amount of local traffic-and industry-specific concentrations is not surprising considering that long-range transport and formation of secondary particles in the atmosphere can contribute considerably to the particle mass concentration in North-Rhine-Westphalia and the Ruhr area, sometimes more than 50% depending on the meteorological situation [28]. The spatial distribution within the study area, represented by quintiles of respective PM_{10} distributions (Figure 4), illustrates that the agreement between all sources and industry-specific sources is better than between all sources and traffic-specific PM. Due to substantial industrial emissions from the Duisburg inland harbor and the adjacent industrial area west of the study region, a strong west-east gradient can be observed for industry-specific PM and for all sources PM. The spatial distribution traffic-specific PM follows closely the population-density in the study area, with a strong north-to-south gradient.

The associations between residence-based exposure estimates derived from $EURAD\text{-}CTM_{TRA}$ and ESCAPE-LUR are relatively high ($PM_{2.5}$: 0.69, PM_{10}: 0.58, and NO_2: 0.45), while they are expectedly considerably lower for $EURAD\text{-}CTM_{IND}$ and ESCAPE-LUR ($PM_{2.5}$: 0.16, PM_{10}: 0.0, and NO_2: 0.25) (Table 5). Such patterns are displayed for PM_{10} in the spatial distribution of traffic-specific EURAD-CTM and ESCAPE-LUR and industry-specific EURAD-CTM and ESCAPE-LUR, respectively (Figure 4). A similar pattern is observed taking into account correlations for 14-day mean measurements at ESCAPE monitoring stations (background and traffic) and estimated 14-day mean $EURAD\text{-}CTM_{TRA}$ within respective grid cells (Table 5).

Table 5. Spearman correlation coefficients between 14-day series of measurements at ESCAPE-LUR-monitoring stations and 14-day mean estimations of $EURAD\text{-}CTM_{TRA}$ in respective grid cells.

$EURAD\text{-}CTM_{TRA}$ (Traffic-Specific)	ESCAPE Background Sites	ESCAPE Traffic Sites	All ESCAPE Sites
$PM_{2.5}$	0.69 (n = 9)	0.88 (n = 6)	0.77 (n = 15)
PM_{10}	0.02 (n = 9)	0.83 (n = 6)	0.32 (n = 15)
NO_2	0.57 (n = 16)	0.79 (n = 13)	0.63 (n = 29)

These observations indicate that EURAD-CTM and ESCAPE-LUR do not represent identical aspects of air pollution: while EURAD-CTM represents an area average similar to urban background concentrations, the ESCAPE-LUR was designed to predominantly estimate variability in local traffic-related air pollution, leading to a comparatively high correlation with local traffic-specific air pollution concentrations modeled by EURAD-CTM. The very low correlation with local industry-specific air pollution concentration at the residences indicates, that ESCAPE-LUR represents industry rather poorly compared to EURAD-CTM, where the overall spatial distribution (Figure 3) is mainly driven by industrial sources as has been observed in a previous study [32].

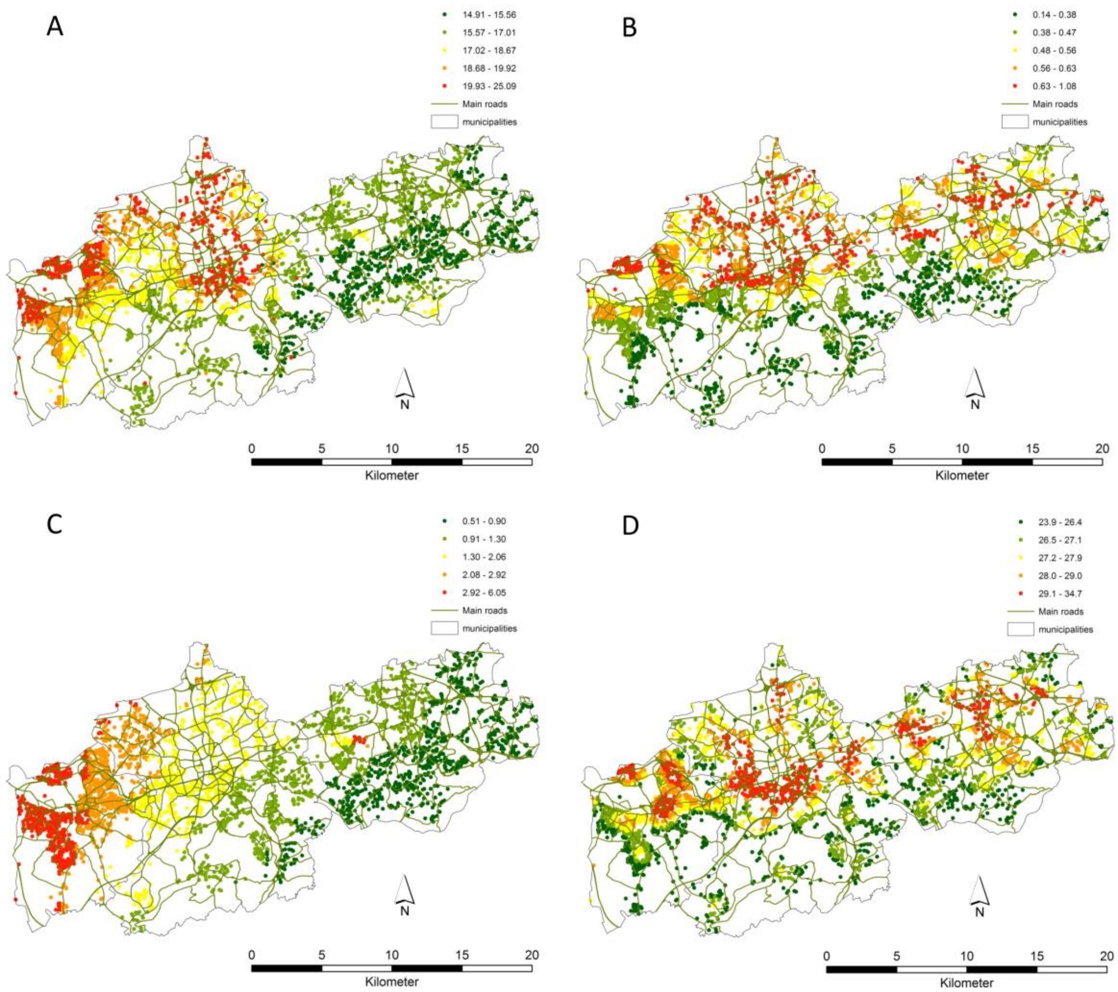

Figure 4. Residence-based spatial distribution of PM_{10} concentrations from EURAD-CTM: all-sources (**A**); local traffic (**B**) and local industry (**C**); and ESPCAPE-LUR (**D**).

4. Conclusions

Based on the comparison between air pollution concentrations modeled by ESCAPE-LUR and EURAD-CTM within the HNR study area, we showed that both model types have different input data as well as different temporal and spatial resolutions, driven by their different aims and application. While the point-specific ESCAPE-LUR primarily aims to estimate temporally stable and spatial variable long-term exposure to locally-emitted (mostly traffic-related) air pollution with a very high spatial resolution, the EURAD-CTM aims to estimate a spatio-temporal average air pollutant concentration in a small area (*i.e.*, 1 km^2), taking into account a range of major sources, e.g., traffic, industry, meteorological condition, and transport. While the observed weak to moderate overall agreement between the ESCAPE-LUR and the EURAD-CTM supports earlier findings [12], our analysis showed that the agreement between the two models improved considerably after restricting the EURAD-CTM to local traffic only. This finding was further supported by results comparing 14-day mean concentrations estimated by EURAD-CTM and measured at purpose-specific ESCAPE monitoring sites, yielding the highest correlations for traffic-specific EURAD-CTM estimates and measurements at traffic sites.

One of the principal strengths of the point-specific ESCAPE-LUR is to capture very small-scale variations in air pollution. Yet, this accuracy may be more error-prone than the coarser spatial resolution of 1 km^2 used by EURAD-CTM, regarding exposure assignment in cases of high personal mobility within small distances, like daily chores around the residence. The biggest strength of an LUR approach in general is the wide-ranging applicability, like the relatively small requirements on measurement sites (low cost), the individual location of measurement sites, the easy assessment of land use data, and the straight forward model building procedure, based on linear regression modeling. In contrast, the EURAD-CTM, or chemical transport and dispersion modeling approaches in general, are less accessible to changes by the user due to the highly complex underlying mathematical, physical, and chemical modelling procedures. These complex procedures are, however, accompanied with benefit of including chemical transport actions, which allow modeling air pollution components that have not been measured. The LUR, on the other hand, is limited to modelling measured air pollutants. Moreover, CTMs enable the investigation of the role of meteorology and the prediction of air pollutant concentrations under hypothetical emission situations.

The comparatively easy applicability of LUR modeling and statistical model building procedure may come along with potential costs of wrong decisions: the initial choice of locations of the measurement sites limits the specificity of the model to capture those emission sources, whose concentration gradients are well captured by the chosen sites and may fail to capture all important source-specific concentration gradients across a study area, especially if important sources change over time. Restricting predictors to land use data might neglect important predictors of air pollution concentrations from other sources and processes, like chemical interaction and transport. Similarly, CTMs are only valid if based on a comprehensive and detailed emission database. To overcome limitations of each of the models and optimally make use of the respective strengths, we propose to combine the two approaches into a hybrid model [43,44]. These hybrid models are usually based on the LUR model since LURs are by design much easier to modify.

To conclude, our results show that ESCAPE-LUR and the EURAD-CTM are constructed to estimate complementary aspects of air pollution and both approaches have respective strengths and limitations, which need to be considered especially when investigating health effects. The possibility of combining the strengths of both, e.g., using hybrid models will be the next step to enhance exposure assessment.

Supplementary Materials: The following are available online at www.mdpi.com/2073-4433/7/3/48/s1. Figure S1: Flowchart of the EURAD model system containing the meteorological driver MM5, the pre-processors ECP and PREP, the emission model EEM and the chemistry transport model EURAD (input parameters are shaded in blue, output parameters are shaded in yellow and procedural parts are shaded in green or magenta), Figure S2: Spatial distribution of EURAD-CTM (1 km^2, yearly mean 2008) and ESCAPE-LUR (point-specific yearly mean October 2008–October 2009) at 4809 residences within the HNR study area for PM_{10} (A+C) and NO_2 (B+D), Figure S3: Boxplots of air pollution concentrations of monthly-mean PM_{10} and NO_2 concentrations over three year for a traffic-specific (grid cell: 690), and a background-specific location (grid cell: 679) with annual mean ESCAPE-LUR estimates and annual measurements at LANUV monitoring sites, Table S1: Time and locations of the ESCAPE-measurement campaign, Table S2: ESCAPE-LUR for $PM_{2.5}$, PM_{10} and NO_2, Table S3: Time and Location of routine monitoring sites, provided by LANUV, within the HNR study area.

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