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Modelling nationwide spatial variation of ultrafine particles based on mobile monitoring

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ABSTRACT

Background: Large nation- and region-wide epidemiological studies have provided important insights into the health effects of long-term exposure to outdoor air pollution. Evidence from these studies for the long-term effects of ultrafine particles (UFP), however is lacking. Reason for this is the shortage of empirical UFP land use regression models spanning large geographical areas including cities with varying topographies, *peri*-urban and rural areas. The aim of this paper is to combine targeted mobile monitoring and long-term regional background monitoring to develop national UFP models.

Method: We used an electric car to monitor UFP concentrations in selected cities and towns across the Netherlands over a 14-month period in 2016–2017. Routes were monitored 3 times and concentrations were averaged per road segment. In addition, we used kriging maps based on regional background monitoring (20 sites; 3 × 2 weeks) over the same period to assess annual average regional background concentrations. All road segments were used to model spatial variation of UFP with three different land-use (regression) approaches: supervised stepwise regression, LASSO and random forest. For each approach, we also tested a deconvolution method, which segregates the average concentration at each road segment into a local and background signal. Model performance was evaluated with short-term (400 sites across the Netherlands; 3 × 30 minutes) and external longer-term measurements (42 sites in two major cities; 3 × 24 hours). We also compared predictions of all six models at 1000 random addresses spread over the country.

Results: We found similar predictive performance for the six models, with validation R^2 values from 0.25 to 0.35 for short-term measurements and 0.52 to 0.60 for longer-term external measurements. Models with and without deconvolution had similar predictive performance. All models based on the deconvolution method included a regional background kriging map as important predictor. Correlations between predictions at random addresses were high with Pearson correlations from 0.84 to 0.99. Models overestimated exposure at the short-term and long-term sites by about 20–30% in all cases, with small differences between regions and road types.

Conclusion: We developed robust nation-wide models for long-term UFP exposure combining mobile monitoring with long-term regional background monitoring. Minor differences in predictive performance between different algorithms were found, but the deconvolution approach is considered more physically realistic. The models will be applied in Dutch nation-wide health studies.

1. Introduction

Most studies of the health effects of ultrafine particles (UFP) so far focused on short-term exposures in a relatively small area. Only a few studies looked at the long-term effects of UFP exposure (Downward et al. 2018; Simon et al. 2017) and only one study looked at long-term exposure to UFP over a wide area (Ostro et al. 2015). The Ostro study was conducted among participants across California, USA, while the former studies were restricted to a few cities. Nation-wide epidemiological studies including administrative cohort studies have been very informative to assess health effects of air pollutants such as $PM_{2.5}$ and NO_2, because of the large size of the population, avoidance of selection bias, and the increased contrast in exposure. These studies have become possible because of national exposure models, typically based on nation-

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wide routine monitoring and modelling, incorporating variation at the regional background, urban background, and local scale. More specifically, the inclusion of regional background concentrations in LUR models advanced exposure assessment and health-effect studies substantially by increasing exposure contrast (Beelen et al. 2008). This approach is not directly possible for UFP because of the lack of nationwide routine monitoring. Ongoing standardization work by CEN of particle counters may stimulate increased particle number monitoring in networks (Bustin et al. 2019).

In the past decade, exposure assessment of UFP has been revolutionized using mobile platforms. Together with advancements in air monitoring instrumentation, such as higher time resolution and greater portability, these platforms can capture the high variability of UFP in space and time. To assess the spatial variation of UFP, studies used cars (Larson, Henderson, and Brauer 2009; Patton et al. 2014; Weichenthal et al. 2014; Shairsingh et al. 2019; Kerckhoffs et al. 2016), bikes (Hankey and Marshall 2015; Farrell et al. 2016), backpacks (Sabaliauskas et al. 2015) and combinations thereof (Minet, Gehr, and Hatzopoulou 2017) to create land use regression (LUR) models. These campaigns were generally restricted to a single city's boundaries, limiting their ability to be used in wide-scale epidemiological studies including *peri*-urban and rural areas (Hoek 2017). We recently showed similarities in UFP models between cities in several European countries (van Nunen et al. 2017; Kerckhoffs et al. 2017). However, these campaigns focused only on cities and do not allow the inclusion of *peri*-urban and rural populations in health evaluations of UFP.

Researchers have used different algorithms to develop air pollution prediction algorithms, including stepwise linear regression, regularization methods and more recently machine learning methods (Kerckhoffs et al. 2019; Weichenthal et al. 2016; Hong et al. 2019). However, little is known about the relative performance of these algorithms. Furthermore, these models did not attempt to separate determinants at different spatial scales. Especially for larger study areas, models have been developed separately for different spatial scales such as the regional and urban background and local scale (Beelen et al. 2008), in order to better characterize spatial variation for the different spatial scales, which can be important in the development of predictive exposure maps. Recently, a deconvolution approach was proposed to separate local contributions from urban background contributions in mobile monitoring data (Shairsingh et al. 2018).

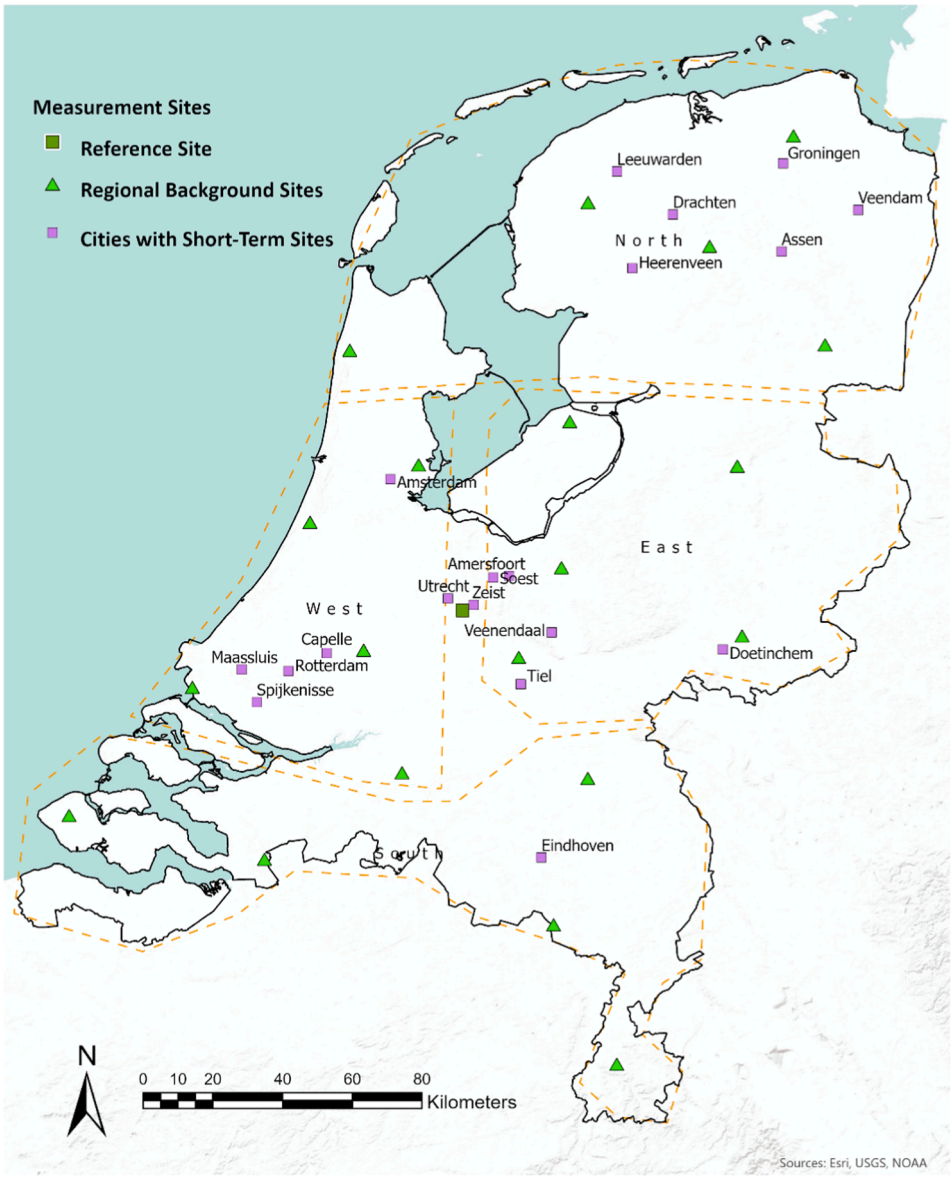

Fig. 1. *Map of towns with mobile and short-term UFP measurements in purple* (multiple sites per town) *and regional background sites in green. Green square is the reference site (Bunnik).* (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Here, we aimed to develop and assess the performance of a national UFP model by combining targeted mobile monitoring in multiple cities and regional background monitoring. A second aim was to assess differences in performance of three different prediction algorithms with and without deconvolution to develop spatial models.

2. Methods

2.1. Study design

This study combines nation-wide regional background measurements with short-term and true mobile measurements from a mobile platform. The regional background component has been described previously (van de Beek et al. 2020). Briefly, we selected 20 regional background sites across the country (green triangles in Fig. 1) and measured each site three times for periods of 14 days with a stationary setup. Individual regional background measurements ranged from 3,028 to 8,202 particles/cm^3. We used kriging to estimate annual average regional background concentrations across the Netherlands. The sampling periods, concentration statistics, and interpolated map used in this study can be found in the supporting information (figures A2 and A3).

Short-term and mobile monitoring was done alternately with the same mobile platform and measurements were done in towns that were selected to cover the study areas of selected cohort studies in the Netherlands, supplemented with towns to increase the national coverage. Most major cities (Amsterdam, Rotterdam, Utrecht, Eindhoven, Groningen) were included as well as several medium sized towns. Mobile measurements were performed when driving from one short-term site to the next and then aggregated over a road segment. We elaborate on the different data sets in the paragraphs describing the training (2.2) and test data (2.4). In brief, data for all road segments (n = 14,392) were used for model development (training data). The short-term and long-term measurements were used as test data. The short-term measurements consisted of 400 sites (traffic and urban background) spread across the Netherlands for repeated stationary short-term measurements (30 min). The long-term measurements were used from a previous study (van Nunen et al. 2017), consisting of 42 sites in the cities of Amsterdam and Utrecht with 3 times 24 h measurements. We furthermore selected 1000 random addresses in the Netherlands to compare models developed with different algorithms. Fig. 2 shows a schematic overview of the measurements (used for training and testing) and models used in this study.

Measurements started after 9:15 AM and stopped before 4:00 PM, to avoid rush hours and increase comparability between road segments, following previous campaigns (Klompmaker et al. 2015; van Nunen et al. 2017; Kerckhoffs et al. 2016). Avoiding rush hour resulted in lower spatial contrast but we assessed the lack of comparability to be more important than the loss of contrast. All measurements took place between June 2016 and November 2017. With this design, we captured the day-to-day, seasonal and a large part of the within-day variability of UFP concentration levels.

2.2. Training data

All measurements were performed using a condensation particle counter (TSI, CPC 3007), installed in the back of an electric car (REVA, Mahindra Reva Electric Vehicles Pvt. Ltd., Bangalore, India). The setup of the car and equipment has been extensively described by Klompmaker et al. (2015). In brief, the CPC was connected via conductive silicon tubes to the outside of the car and performed measurements every second. In previous analysis we also found that speed does not affect the measurements (Kerckhoffs et al. 2016). The geographical location of the electric car was recorded using a Global Positioning Unit (GPS, Garmin eTrex Vista) and linked to the instruments in the car based on date and time. Following our previous mobile monitoring measurement campaigns (Kerckhoffs et al. 2016), we corrected the GPS signal for small spatial errors by assigning all GPS points to the nearest road. We made sure that the GPS points were assigned to the correct road by creating a 30 m buffer around the route that was driven. Only GPS points within this buffer were assigned to roads that were completely in this buffer. Then we calculated average concentration levels of UFP per road segment. Road segments were on average 110 m long (SD: 68 m) and accumulated on average 43 s of UFP data (IQR: 9–44 s) over the study period.

UFP values were removed from the data set when the concentration was 500 particles/cm^3 or less or if the UFP number counts increased or decreased in one second by a factor of 10 or more, as these reflect malfunctioning of the instrument (Kerckhoffs et al. 2016). In total, less than 0.01% of the UFP data were removed due to these criteria.

As not all measurements were performed at the same time, we corrected all mobile measurements for temporal variation. This was done using a reference site with the same equipment (and tubing) as the electric vehicle, set up in the middle of the country (Bunnik; see Fig. 1). In our previous paper (van de Beek et al. 2020) we showed moderately high correlations between background concentrations of sites up to 200 km apart indicating that for UFP the Netherlands can be regarded as one airshed. We used the difference method to correct for temporal variation, following procedures in previous mobile monitoring campaigns

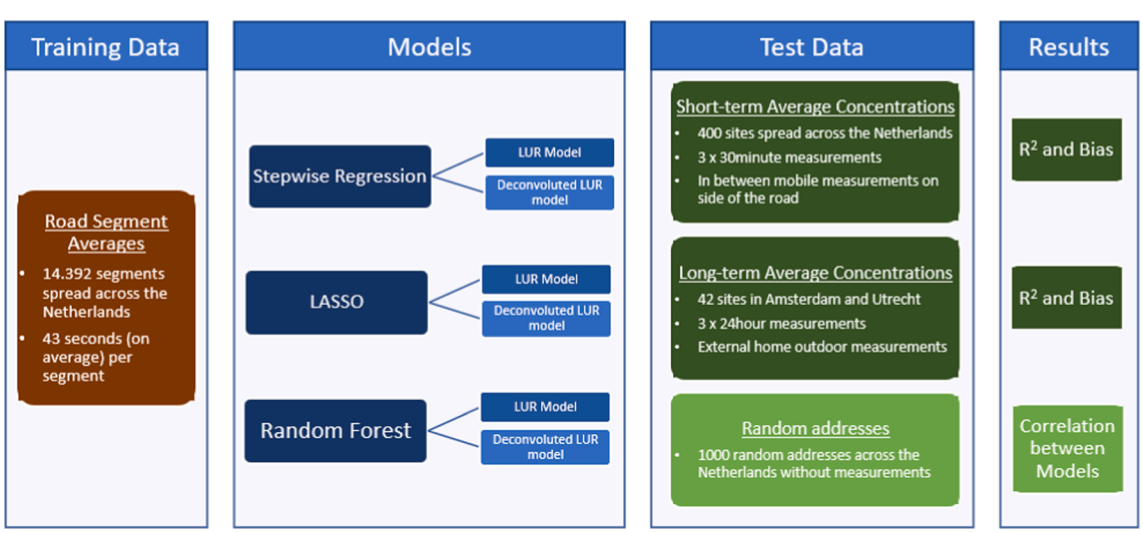

Fig. 2. Schematic overview of measurements and models.

(Kerckhoffs et al. 2016; 2017). First, the overall mean concentration for the entire campaign at the reference site was calculated. Next, we created a moving average of reference site measurements of 30 min (15 min before to 15 min after) for every timepoint of the mobile monitoring campaign. This value was then subtracted from the overall mean concentration at the reference site. Finally, this difference was added to the time-corresponding road segment average concentrations.

Every three weeks, the CPC used in the car and the CPC at the reference site were co-located in the laboratory to check comparability. This procedure is more elaborately described in the supplement information (table A1). We found a median ratio (averaged over 1 min) for the two CPCs of 0.88 (CI: 0.78–1.01). Because 14% of the reference site UFP measurements were missing, we imputed UFP using a DiscMini that was placed as backup at the same reference site. Pearson correlation of hourly concentrations between the two instruments was 0.81. The formula used for imputation was CPC = 1345 + 0.957*DiscMini. Both instruments have a lower cutoff value of 10 nm.

2.2.1. Deconvolution

The idea of deconvoluting is to separate local UFP contributions from background concentrations. To do this, we use an approach developed by Brantley et al. (2014) and Shairsingh et al. (2018) using a spline of minimums. First, measurements were averaged over different time scales (ranging from 60 s to 2000 s) and then correlated with a local traffic variable (Figure A1). The time scale for which the local traffic variable does not correlate with the average concentration anymore is considered the averaging period for the background concentration.

In figure A1, correlations are given for UFP measurements and traffic intensities in 50- and 1000-meter buffers, respectively. The local influence drops after an averaging period of 150 s. Taking a moving minimum value of ~ 200 s would represent a background signal in this case. With an average driving speed of about 30 km/h this would mean that background concentrations shift over a distance of about 1.6 km. Then, a regression spline is applied to interpolate between the minimum values from the time series. The minimum value in a time frame of 200 s around every measurement is thus the background concentration. The local signal is derived by subtracting the background concentration from the total concentration. We validated the urban background signal derived from deconvoluting by comparing it to short-term concentrations measured at urban background sites.

2.3. Models

Because there are inconclusive results as to which algorithms are best to be used in developing empirical air pollution models (Kerckhoffs et al. 2019; Chen et al. 2019), we used three different land-use (regression) approaches (stepwise regression, LASSO and random forest) to assess the spatial variation of UFP across the Netherlands. For each approach, we also tested the deconvolution method. All models were developed in R (R Core Team (2013), version 3.5.2).

2.3.1. Predictor variables

In accordance with our previous and most other mobile monitoring studies (Kerckhoffs et al. 2016; 2017), we identified the middle of each road segment and used this coordinate to acquire spatial GIS predictors for LUR modelling (for an overview of GIS predictors see table A2). Besides the regional background predictions from the kriging map, a range of traffic variables was defined, including traffic intensity and road length variables (in 50–1000 m buffers); land use (e.g. port, industry, urban green, airports) and population / household density in buffers from 100 to 5000 m.

2.3.2. Stepwise regression

We used a supervised forward stepwise regression approach that starts with an empty (intercept only) model and then adds variables based on goodness of fit determined by the adjusted R^2 value. This method has been used before for road segment averages (Kerckhoffs et al. 2016; 2017). Variables that i) lead to the largest increase in adjusted R^2, ii) for which the coefficient conforms with a predefined direction of effect and iii) for which the direction of effect of predictors already in the model does not change, are retained in the model. Predictor variables in the final model are removed from the model when they have a p-value larger than 0.1 or a variance inflation factor over 3.

2.3.3. Lasso

LASSO regression is optimized for prediction, because it deals with correlated predictors by imposing a penalty on the absolute size of the regression coefficients (attributing the full effect to only one of the variables, shrinking the other(s) towards zero). With LASSO regression the effects of some variables may be shrunken all the way to zero, and therefore it can be used for variable selection. We used the *cv.glmnet* with default parameters (alpha = 1, nfolds = 10 and "mse" as type.measure) from the *glmnet* package (version 2.0) for all LASSO models.

2.3.4. Random forest

Random forest is a machine learning method that combines many weak classifiers/trees into one strong classifier. A weak classifier is a classifier whose error rate is only slightly better than random guessing. Random forest restricts all regression trees to a limited number of variables, creating a large collection of 'weak' trees. Each tree selects a specified number of variables at random and chooses the best variable to split the data (Hastie, Tibshirani, and Friedman 2009). We set parameters to a 1000 trees and restricted the number of terminal nodes to 12 using the *randomForest* package.

2.4. Test data

The first test set is based on short-term measurements (400 sites measured for 3x30minutes). About 50% of these measurements were taken at urban background sites (no major roads within 1000 m) and the rest at sites with high intensity traffic (>10,000 vehicles/day). An advantage of this dataset is that sites are located across the Netherlands and sampled in the same time frame as the training data. A disadvantage is the short sampling time (average 90 min) that is expected to result in low explained variances because of relatively instable average concentrations. The second set of test data consists of long-term average outdoor UFP concentrations at 42 different sites (traffic and urban background) in Amsterdam and Utrecht that have been collected according to protocols described by van Nunen et al. (2017) and Eeftens et al. (2012). Measurements were made close to the home, at first floor balconies for major roads and either balconies or gardens in homes located in minor roads. These sites were sampled with a DiscMini (Testo AG, Lenzkirch, Germany) 3 times for 24 h. Previous studies have shown good agreement between CPCs and DiscMinis with limited differences in absolute values (Asbach et al. 2012; Meier, Clark, and Riediker 2013). Measurements at these sites are better representatives of average concentrations than short-term measurements at a specific site, but the sites are only in two cities. To compare the two test results directly, we also tested the models on short-term measurements restricted to Amsterdam and Utrecht (n = 80). Thirdly, we used a set of 1000 randomly selected home addresses to compare the predictions of the different models across the Netherlands.

3. Results

Table 1 shows the distribution of road segment average UFP concentrations, stratified by region and road type. Concentrations from mobile measurements were on average 15,861 particles/cm^3 over an average duration of 43 s per street segment. Concentrations measured in Amsterdam (population: 860,000) and Utrecht (population: 350,000) were on average higher than those measured in other regions of the country, mainly because these measurements were done in two of the

Table 1
Measured road segment average UFP concentrations in particles/cm3.

Region	Road Type	Number of Road Segments	Mean	10th Percentile	90th Percentile
AMS-UTR	Minor	1,221	15,557	7,177	26,289
	Major	2,094	21,831	8,887	40,799
East	Minor	1,118	11,495	6,847	16,902
	Major	1,150	17,061	8,110	29,176
North	Minor	2,144	11,044	3,103	18,288
	Major	614	16,290	5,082	28,851
South	Minor	1,490	11,621	5,378	18,609
	Major	1,105	17,697	6,919	32,581
West	Minor	363	12,179	6,253	20,021
	Major	3,016	18,694	7,069	33,132
All	Minor	6,561	12,080	5,061	20,250
	Major	8,316	18,618	7,453	33,907

biggest cities in the Netherlands, while measurements in other regions were also done in smaller cities and municipalities. Lowest concentrations can be found in the North, where population density is small opposed to other regions in the country. Measurements on major roads were on average 55% higher than on minor roads.

3.1. Deconvoluting

Table 2 shows the average background UFP concentrations obtained from the three different measurement strategies (short-term, mobile, and regional long-term), per region. Average concentrations at short-term urban background sites were very similar to the deconvoluted urban background signal calculated from the measurements with the mobile platform. The regional background kriging map was based on measurements outside of cities resulting in concentrations that were considerably lower than the urban background at short-term sites and deconvoluted background signals. However, the same regional pattern can be observed. In figure A4 we show the same comparison but divided by city.

3.2. External prediction

Table 3 shows the performance of all individual algorithms based on short-term and longer-term test datasets. In general, all algorithms perform equally well with the proportion of variance explained (R^2) in the external test sets ranging from 0.25 to 0.35 for the short-term test data and from 0.52 to 0.60 for the longer-term test data. Both random forest algorithms seem to perform slightly better on the short-term measurements, but deconvoluted stepwise regression and LASSO explain the longer-term measurements somewhat better. All models tend to overestimate our external test data, usually by between ~ 2000 and ~ 4000 particles/cm^3. Appendix B provides scatterplots of measured (short-term and longer-term) versus predicted concentrations and Bland Altman plots for all six algorithms. We did not find systematic differences in the extent of bias between different regions or road types

Table 2
Comparison of average background concentrations per region obtained from short-term monitoring, mobile monitoring, and regional background monitoring. Concentrations in particles/cm^3.

Region	Average concentration of short-term urban background sites	Average background concentration of deconvoluted mobile measurements	Average concentration of regional background long-term sites
AMS - UTR	11,348	11,420	/
East	7,583	8,493	5,543
North	6,100	6,130	4,359
South	8,732	8,787	5,856
West	10,318	10,356	6,053

Table 3
Comparison of predicted concentrations from different algorithms on external test data. Bias is expressed in particles/cm^3.

	Short-Term		Short-Term AMS-UTR		Long-Term AMS-UTR	
	R^2	Mean Bias	R^2	Mean Bias	R^2	Mean Bias
Stepwise Regression	**0.28**	2,883	**0.31**	3,422	**0.60**	2,156
Stepwise Regression Deconvoluted	**0.30**	3,470	**0.29**	3,904	**0.60**	2,690
LASSO	**0.29**	3,225	**0.31**	3,092	**0.60**	1,806
LASSO Deconvoluted	**0.25**	3,921	**0.35**	3,655	**0.55**	2,392
Random Forest	**0.34**	3,132	**0.32**	3,025	**0.52**	2,362
Random Forest Deconvoluted	**0.34**	3,390	**0.31**	3,146	**0.52**	1,818

(Table A3).

Variables selected in the stepwise regression and LASSO models can be found in table 4. As random forest does not perform variable selection, we show the top 7 variables that were most influential in random forest. Selected variables with regression coefficients and p-values can be found in the supplement (Tables B1-B4). For random forest we show the distribution of minimal depth for the best variables (figures B5 and B7). Figures B1-B4, B6, and B8 show scatterplots and Bland-Altman plots for the predictions of the mobile models on all 3 test data sets (i.e., short-term, short-term restructured to the Utrecht-Amsterdam region and long-term Utrecht Amsterdam Region) for all 6 algorithms. All models select (or give high importance to) very local traffic intensity variables, population density in a large buffer (5000 m) and the presence of a nearby port. The regional background map was selected as predictor in all three (deconvoluted) background models, but not in the standard models.

Applying the different algorithms to a set of random addresses (without measurements) across the Netherlands resulted in predictions that were highly correlated, with Pearson correlations ranging from 0.84 to 0.99 (Fig. 3). Correlations were higher between stepwise regression and LASSO models than between stepwise/LASSO models and the random forest models. Table A4 shows the distributions of the predicted concentrations for all algorithms.

4. Discussion

This is the first study showing that it is possible to develop predictive models for UFP concentrations on a national scale (41,543 km^2), while accounting for local variability of UFP concentrations in urban areas. We achieved this by combining mobile monitoring with targeted regional background monitoring. We found similar performance for all six algorithms. Models explained 25% to 35% of the variability in measured short-term average concentrations across the Netherlands and 52% to 60% of the variability in longer-term measurements. Correlations between predictions with different algorithms at random addresses were high with Pearson correlations between 0.84 and 0.99.

4.1. Development of a national model

While LUR models have been proven to be useful in assessing small scale variation of UFP (Hoek 2017) and are able to predict retrospective and prospective exposures (Montagne et al. 2015; Simon et al. 2020), few studies used LUR models to investigate the effect of long-term UFP exposure on adverse health effects (Simon et al. 2017; Downward et al. 2018). Concentrations of UFP vary significantly over space and time and it can therefore be difficult to assess exposure in a large area and over a long time. LUR models of UFP have therefore been restricted to a few cities. There is, however, a limited ability of LUR models measured in one city to be transferred to another city and even more so to rural areas (Shairsingh et al. 2019). We therefore had to take into account variation

Table 4
Predictor variables selected by the different models. Variables in the random forest column are the top 7 variables across several performance methods.[1]refers to buffer size; [2]near refers to nearest road.

	Stepwise	Stepwise deconvoluted		LASSO	LASSO deconvoluted		RF	RF deconvoluted	
		Background	Local		Background	Local		Background	Local
Background predictors									
Regional background		+			+			+	
Household 5000 m[1]	+			+	+		+	+	
Population 5000 m		+					+	+	
Port 5000 m	+	+		+	+			+	
Industry 5000 m									
Highways 1000 m					+				
Major roads 1000 m									
Heavy traffic intensity 1000 m					+				
Industry 5000 m					+				
Urban greenery 5000 m								+	
Agriculture 5000 m								+	
Y-Coordinate								+	
Local predictors									
Traffic intensity near				+					+
Heavy traffic intensity near[2]	+								
Traffic intensity 50 m	+		+	+		+	+		+
Traffic intensity major roads 50 m	+		+	+			+		+
Traffic intensity 100 m				+			+		+
Traffic intensity major roads 100 m							+		+
Traffic intensity 300 m			+	+			+		+
Household 300 m			+						
Restaurants near									+

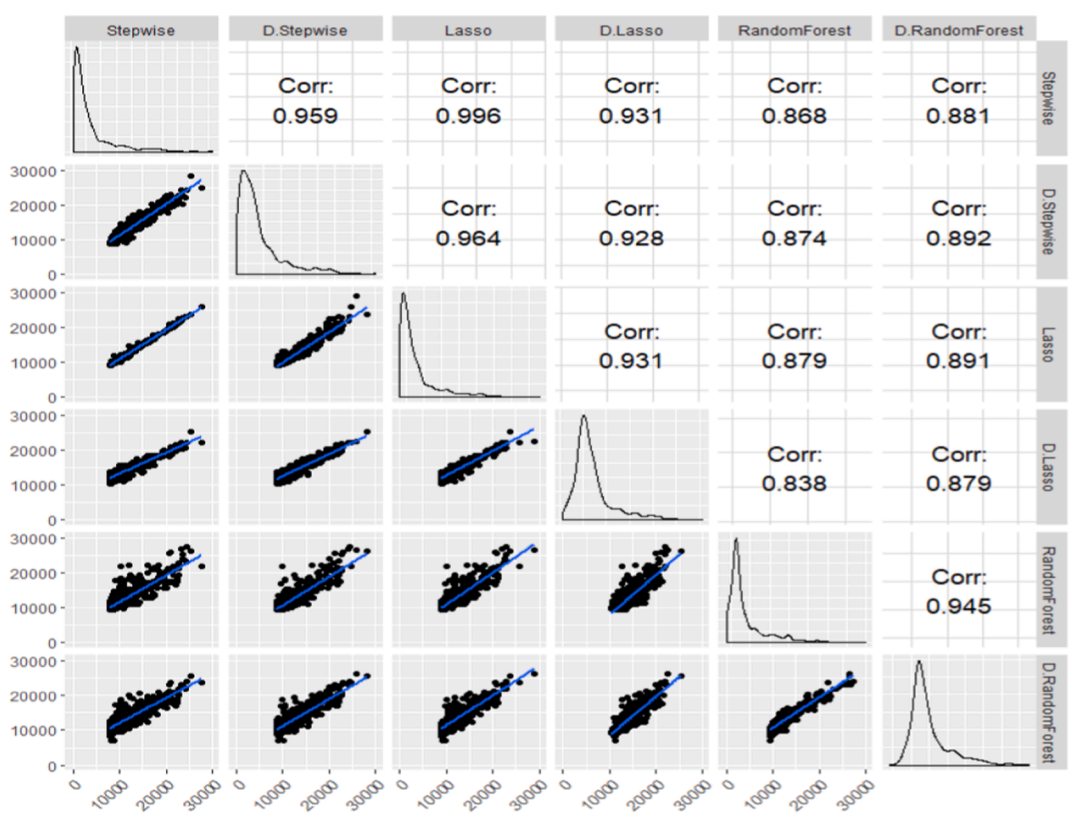

Fig. 3. Comparison of prediction algorithms at randomly selected addresses across the Netherlands (n = 1000). Concentrations in particles/cm^3. Abbreviation "D." refers to the deconvoluted method.

on larger spatial scales, including urban and regional background variation in addition to the more commonly included local scale variation within urban areas. Developing a national model for UFP is more challenging than for more commonly studied pollutants such as $PM_{2.5}$, NO_2 and O_3 for which more advanced modelling frameworks based on deterministic models and emission inventories are available. Such studies can often rely on routine monitoring, chemical transport modelling (CTM) and/or satellite data (Vienneau et al. 2013; Hystad et al. 2011; Novotny et al. 2011; de Hoogh et al. 2016). The only wide-scale model for UFP so far was used in the United States in the California Teachers Study, covering about 424,000 km^2 (about 10 times the size of The Netherlands). That study estimated UFP exposure at a 4 km grid

based upon chemical transport modelling (Ostro et al. 2015).

Routine monitoring, CTM, and satellite observations were not available for UFP, so we combined mobile monitoring in many towns across the country with specific long-term regional background monitoring. As mobile monitoring was not feasible in all towns of the Netherlands, our assumption was that the regional background component would account for differences in exposure related to region of the country.

4.2. Comparing prediction algorithms

We only found minor differences in explained variance and bias between different algorithms for developing land use regression models of spatial concentration variations of UFP. The low R^2 values of the short-term test set is primarily due to the influence of temporal variability in the short-term test data (3 × 30 min), which do not represent a stable average concentration. For the long-term test data compared to short-term test data we found substantially higher R^2, which can be explained by the longer measurement time of long-term measurements (Kerckhoffs et al. 2017; 2019; Montagne et al. 2015). We did not have long-term test data at the national scale, but we did observe that the R^2 values based upon short-monitoring tests data in Utrecht-Amsterdam (the cities with longer-term monitoring) and the entire Netherlands were similar. We suspect that on a national scale, the R^2 values for longer-term averages is similar to what we found in Amsterdam-Utrecht.

The minor differences between models (on the same test data) were consistent with findings of a previous study evaluating models based upon mobile and short-monitoring where we tested a much wider range of models and also found minor differences between approaches (Kerckhoffs et al. 2019). While no conclusion can be drawn regarding which algorithm performs best, it is encouraging that different approaches in this study generated robust predictions at randomly selected sites. Chen et al. (2019) compared modelling algorithms in a European setting with long-term measurements of NO_2 and $PM_{2,5}$ and also found minor differences between a wide range of algorithms, including the three algorithms included in the current paper.

Predictors that were selected or given the most weight in the different exposure models that we explored were similar. All models select very local traffic variables, population density in a large buffer and the presence of a port. These variables represent known sources of ultrafine particles and have been included in previous LUR models in the Netherlands (Kerckhoffs et al. 2016; van Nunen et al. 2017; Montagne et al. 2015; Hoek et al. 2011) and elsewhere (Sabaliauskas et al. 2015; Farrell et al. 2016; Rivera et al. 2012; Weichenthal et al. 2015; Jones et al. 2020). This persists in the deconvolution method, with local traffic variables selected in the local model and population density and port area in the background model.

All three background models in the deconvolution approach selected the regional UFP background map as a predictor into the model, while none of the standard algorithms (without deconvolution) selects the regional background concentration as important predictor. Conceptually, regional variation should be represented in a national model. However, all standard models select mainly local traffic source related variables into the model. Regional differences are apparently also represented by the difference in local traffic intensities or large-scale population density.

4.3. Bias

Predicted UFP concentrations from the different algorithms were on average 20% (~2,000 to ~ 4,000 particles/cm^3) higher than stationary measurements. This is less than the overestimation that we found in previous studies using mobile monitoring (Kerckhoffs et al. 2016; 2017; 2019). Previous mobile models were based on measurements performed in urban areas and overestimated measured concentrations on average by ~ 5,000 particles/cm^3, tested on the exact same test dataset with longer-term average concentrations that we used in the present study. In one of those previous studies, mobile stepwise, LASSO and random forest models overestimated exposure by 6,902, 3,374 and 5,688 particles/cm^3 respectively (Kerckhoffs et al. 2019). Other studies that used mobile models found similar differences. For example, a difference of 5,300 particles/cm^3 has been found between mobile monitoring and home outdoor sites in Chelsea (Massachusetts, USA) (Simon et al. 2017). Two other studies compared measurements of UFP on the sidewalk and at the façade of buildings and found a difference in concentration levels of about 15–28% (Kaur, Nieuwenhuijsen, and Colvile 2005; Ragettli et al. 2014; Sabaliauskas et al. 2015).

Overestimation comes from the fact that these models are based on data collected in the middle of the road, while we assign them to home outdoor residential locations. Of note, some over- and underprediction can also be expected by measuring only during daytime and not measuring during rush hours and night-time, respectively. In a previous study, we documented a high correlation ($R^2 > 0.95$) between UFP exposures at 50 fixed sites in Amsterdam for different times of the day including rush hours, daytime non-rush hours and 24-hour average (Downward et al. 2018). We also documented only small differences in the 24-hour average concentration and the average of the period used for mobile monitoring in that study (contrast between traffic and urban background sites was 16,000 and 21,000 particles/cm3 for 9–16 h and 24 h, respectively). While the absolute difference in concentrations between on-road driving and at the façade of buildings is higher on major roads, the relative difference is similar for minor and major roads. Therefore, we argue that the overestimation is modest and non-differential across road types. As the modest over-estimation did not differ much across road types, the exposure contrast in epidemiological studies will only be mildly affected. We suspect that the impact on effect estimates in epidemiological studies is likely limited. Errors from land use regression models are generally regarded to be Berkson-like. Berkson type measurement error affects the power of an epidemiological study, but does not bias regression coefficients (Armstrong 1998). A correction factor in the order of 15 to 30% seems to be the most appropriate for correcting on-road measurements if risk is to be expressed per unit of exposure.

4.4. Deconvolution

Adding a deconvolution step to stepwise regression, LASSO and random forest did not increase the predictive performance and did not reduce bias in our study. Shairsingh et al. (2018), who developed and applied this method, also found only minor differences in predictive performance between deconvoluted and standard models but found less bias when resolved models were used for external validation. A potential explanation for this difference could be the fact that Shairsingh and coauthors used spatiotemporal LUR models for their predictions, while we used purely spatial models. The temporal aspect also allowed them to split the background signal in an urban background and regional background signal, making it a 3-step deconvolution method.

All three deconvolution models included the regional background concentration in their final model, whereas the standard models did not. A national model including the background and local scale fits better with physical reality of spatial distribution of air pollution. Though bias is slightly higher on average in the deconvoluted models, bias is more evenly distributed along the concentration range (major vs minor roads). Separate models for different scales have been developed previously at the national and European scale in empirical LUR models (Beelen et al. 2008; 2009). In dispersion modelling the regional background, urban background and local scale are explicitly modelled with separate models as well (e.g Hvidtfeldt et al. 2019).

4.5. Implications for epidemiological studies

Applying models that are developed based on measurements from

one city is hampered by an unknown background concentration when they are transferred to another city. This study shows that variation in UFP concentrations is not only influenced by local variability within cities. Residences similar in terms of local (e.g. nearby traffic) and urban sources (e.g. population size) in different regions of the country still differ in exposure for two reasons. First, as previously described in van de Beek et al. (2020), substantial regional differences in background UFP concentrations were found within the Netherlands (figure A2). Second, background concentrations in Dutch cities ranged from about 3,300 to 13,100 particles/cm^3 (figure A4). Admittingly, these numbers reflect total background concentrations (urban background and regional background). Still, the differences in background concentrations between cities are generally bigger than the difference between traffic and urban background stationary sites within a city in this study (average of 3,500 particles/cm^3).

As standard LUR models are often limited to include large-scale background differences (Kerckhoffs et al. 2015; Hoek et al. 2015), adding a regional background component creates a better representation of the spatial variation of UFP in the Netherlands. While the regional background map was not selected in all standard models, it was selected in all deconvoluted models. Deconvolution of the mobile monitoring signal is therefore an alternative for specific regional background monitoring, especially when considering larger countries than the Netherlands, that can have even larger differences in regional background concentration. The available resources determine the feasibility of this approach.

Measuring with a mobile platform enabled us to measure spatially diverse environments in a limited amount of time, and with a limited number of monitoring devices. This is cost-effective and especially within city limits, it can capture the high variability of UFP concentrations in complex urban terrains. The goodness of fit of the UFP models was moderate to high when compared to long-term measurements. This performance is comparable to the performance of models successfully applied in epidemiological studies for $PM_{2.5}$, NO_2 and BC (de Hoogh et al. 2018). UFP models have also been developed based on longer term monitoring data in a few studies (Hoek et al. 2011; Cattani et al. 2017; Wolf et al. 2017). The number of sites in these studies was small however (between 20 and 50 sites). The cost of the UFP equipment and the need for regular technician supervision precludes long-term monitoring at a large number of locations at this time. Though we have not formally compared the performance of UFP models based upon mobile and long-term monitoring, we suspect that mobile monitoring is more attractive for UFP.

For developing a national model, separate modelling of background and local scale exposures should be considered. For larger countries, characterization of the regional background is likely best by a combination of fixed site monitoring and deconvolution of the mobile monitoring signal. To assess the potential overestimation by mobile monitoring models, longer-term measurements at a limited number of residential sites spread over the study domain should be part of the study design. Models used in this study include regional, urban background and local information on UFP concentrations, making them applicable to large nation-wide cohorts in the Netherlands.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2021.106569.

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