A Comparison of Strategies for Estimation of Ultrafine Particle Number Concentrations in Urban Air Pollution Monitoring Networks

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Abstract

We propose three estimation strategies (local, remote and mixed) for ultrafine particles (UFP) at three sites in an urban air pollution monitoring network. Estimates are obtained through Gaussian process regression based on concentrations of gaseous pollutants (NO_x, O₃, CO) and UFP. As local strategy, we use local measurements of gaseous pollutants (local covariates) to estimate UFP at the same site. As remote strategy, we use measurements of gaseous pollutants and UFP from two independent sites (remote covariates) to estimate UFP at a third site. As mixed strategy, we use local and remote covariates to estimate UFP. The results suggest: UFP can be estimated with good accuracy based on NO_x measurements at the same location; it is possible to estimate UFP at one location based on measurements of NO_x or UFP at two remote locations; the addition of remote UFP to local NO_x, O₃ or CO measurements improves models' performance.

Capsule abstract:

UFP can be estimated with good accuracy at one location based on NO_x measurements at the same location and based on measurements of NO_x or UFP at two remote locations.

Key words: Ultrafine particles estimation; urban air pollution; pollution monitoring network; Gaussian process regression; statistical modelling.

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1. Introduction

Exposure to traffic-related pollution, especially UFP and nitrogen oxides (NO_x), is of great concern in urban environments because of their adverse impact on human health (Hong et al., 2002; de Hartog et al., 2003; Atkinson et al., 2010; Jacobs et al., 2010; Bos et al., 2011; Kumar et al., 2011a; Kumar et al., 2014).

UFP are commonly defined as particles having a diameter of less than 100 nm (Morawska et al., 1998), and the consensus is that these particles contribute most (around 80%) of the total particle number concentration (PNC) (Heal et al., 2012; Kumar et al., 2011b; Morawska et al., 2008; Charron and Harrison, 2003), whereas their corresponding mass accounts for less than 20% of the total particle mass concentration (Kittelson, 1998). UFP can be classified into the “nucleation”, “Aitken” and “accumulation” modes. In terms of size ranges, the nucleation, Aitken and accumulation modes typically encompass 1–30, 20–100 and 30–300 nm, respectively. Particles with a diameter below 30 nm contain nearly 30% of total PNC (Morawska et al., 2008; Kumar et al., 2010).

Road vehicle emissions in polluted urban environments can contribute up to 90% of the total PNC (Kumar et al., 2010; Pey et al., 2009). The UFP along the roadside show an association with the vehicle flow characteristics. For instance, increasing vehicle speed increases the emissions of UFP (Kittelson et al., 2004). Among the road vehicles, diesel engines dominate road traffic emission of UFP, and heavy duty vehicles have an average factor of magnitude of two with respect to the light duty engine (Beddows and Harrison, 2008).

UFP vary spatially between the sources and the receptors living or travelling close to the roads (Kumar et al., 2014). This variation depends on many factors such as source type and strength, meteorological and dilution conditions, location geometry and transformation processes, among

others (Heal et al., 2012; Goel and Kumar, 2014).

Currently there is no limit value to control ambient UFP. Consequently, there are not many UFP monitors deployed as part of the governmental monitoring stations. On the other hand, NO_x, ozone (O₃) and carbon monoxide (CO) are regulated pollutants (Directive 2008/50/EC) and their monitors are spread all over Europe. Nitric oxide (NO) and nitrogen dioxide (NO₂) together make NO_x. Emissions of NO_x are associated with all types of high-temperature combustion, but similar to UFP, their most important sources in urban areas remain road vehicles (Westmoreland et al., 2007; Alvarez et al., 2008; Kumar and Imam, 2013).

The dispersion modelling of pollutants mostly fits into two categories: deterministic and statistical. Deterministic dispersion models provide a link between theory and measurements and account for source dynamics and physico-chemical processes explicitly (Holmes and Morawska, 2006). A drawback of these models is that they need detailed information (e.g. boundary conditions), which is not always available. Statistical models do not describe the actual physical processes, but they treat the input data as random variables to derive a statistical description of the target distribution using a set of measurements. A few studies have used a statistical approach in the past (Hussein et al. 2006; Clifford et al., 2011; Mølgaard et al., 2012; Sabaliauskas et al. 2012; Reggente et al., 2014).

We employ a statistical modelling approach – Gaussian process (GP) regression – to estimate UFP in an urban air pollution monitoring network based on local and remote concentrations of NO_x, O₃, CO and UFP.

2. Materials and Methods

2.1. Instrumentation

We recorded UFP and gaseous pollutants for one month at a sampling frequency of 5 min and

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then averaged on a half-hourly basis.

Measurements of UFP were obtained using the GRIMM Nano-Check model 1.320. The Nano-Check can count total PNCs between 25 and 300 nm, and provides the mean diameter of the measured size range.

Chemiluminescence (EN 14211), ultraviolet photometry (EN 14625) and non-dispersive infrared (EN14626) analysers (Air-pointer) were used to measure NO_x, CO and O₃, respectively. The lowest detectable concentration was 1 µg m⁻³ for NO_x and O₃, and 50 µg m⁻³ for CO.

Vehicle counts were recorded in four categories (cars, vans, small and big trucks/buses) using double inductive loop detectors at sites 1 and 3; video counting was performed to obtain traffic data at site 2 (Table 1).

Table 1: Description of the measurement sites.

	Distance from traffic (m)	Weekday traffic volume (veh/day)	Weekend traffic volume (veh/day)	Heavy duty vehicle on weekday (weekend) (% total)
Site 1	~ 3	5000	4000	5% (2%)
Site 2	~ 2	4000	3000	4% (2%)
Site 3	~ 20-30	37,000	25,000	7% (3%)

2.2 Description of the sampling locations

Measurements were carried out in the Borgerhout district (51° 13' N and 4 ° 26' E) of Antwerp, Belgium. Borgerhout is a typical urban commercial and residential area with busy traffic. Measurements were carried out simultaneously for one month (12/02/2010–12/03/2010) at three different sites (Figure 1). Sites 1 and 2 were located in two street canyons with two traffic lanes and moderate levels of traffic. The monitoring devices were deployed in parking lots (few metres far from the traffic). Site 3 was located in a parking area ~30 m far from a major access

road with busy traffic intersections and four lanes (two in each direction) and ~200 m far from a highway.

Figure 1: Map of the measurement sites (Antwerp, Belgium) and their distances from each other. The images show the deployed instrumentation at each site. The black arrow in the image of site 3 shows the location of the deployed monitors.

2.3. Description of the model

2.3.1 Gaussian process regression

We treat the estimation problem as a non-parametric regression problem, and solve it using Gaussian process (GP) regression.

Definition: A Gaussian process is a collection of random variables, any finite number of which have a joint Gaussian distribution (Rasmussen and Williams, 2006). We want to learn, from a set

of measurements ($\mathcal{D}$) a function $f(\cdot)$ of the relationship existing between the set of covariates $\mathbf{x}$

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(NO_x, CO, O₃, UFP) and the target variable, UFP ($\mathbf{y}$), assuming that the observed data $\mathbf{y}$ is generated with Gaussian noise around the underlying function f

$$\mathbf{y} = f(\mathbf{x}) + \epsilon \quad (1)$$

Because of the nature of the dataset used, we do not assume an independent noise, and the dependencies are modelled adding a noise term to the covariance function (k_{Noise}). This method has been suggested by Rasmussen and Williams (2006) and by Murray-Smith and Girard (2001). Prior beliefs about the properties of the latent function are included in the mean $m(\mathbf{x})$ and covariance $k(\mathbf{x}, \mathbf{x}')$ functions. In order to estimate UFP based on data, we consider the joint Gaussian prior of the training observations $\mathbf{y}$ and the test outputs $\mathbf{f}_*$. The posterior distribution is obtained by conditioning the prior on the observed training outputs, such that the conditional distribution of $\mathbf{f}_*$ only contains those functions from the prior that are consistent with the training data

$$p(\mathbf{f}_* | \mathbf{X}_*, \mathbf{X}, \mathbf{y}) = \mathcal{N}(\boldsymbol{\mu}_*, \boldsymbol{\Sigma}_*) \quad (2)$$

where

$$\boldsymbol{\mu}_* = \mathbf{K}(\mathbf{X}_*, \mathbf{X})[\mathbf{K}(\mathbf{X}, \mathbf{X}) + \sigma_n^2 \mathbf{I}]^{-1} \mathbf{y} \quad (3)$$

$$\boldsymbol{\Sigma}_* = \mathbf{K}(\mathbf{X}_*, \mathbf{X}_*) - \mathbf{K}(\mathbf{X}_*, \mathbf{X})[\mathbf{K}(\mathbf{X}, \mathbf{X}) + \sigma_n^2 \mathbf{I}]^{-1} \mathbf{K}(\mathbf{X}, \mathbf{X}_*) \quad (4)$$

are the posterior mean and the posterior variance, respectively. $\mathbf{K}$ is the covariance matrix, which is built from a covariance function (or kernel) $k(\mathbf{x}, \mathbf{x}')$. $\mathbf{X}$ and $\mathbf{X}_*$ are the matrices of the training and test inputs, respectively.

We have assumed that the mean of the GP prior is zero everywhere. At first glance, this could appear restrictive, but in practice it is not, because offsets and simple trends can be eliminated before modelling. The covariance function defines similarity between data points and it is chosen such that it reflects the prior beliefs about the function to be learned. Because UFP follow from a

dynamic process, we have used a non-stationary kernel based on the addition of a linear (k_{Lin}) and a rational quadratic kernel (k_{RQ}). Moreover, we also include a noise term (k_{Noise}) to take into account noise dependencies. The sum of kernels allows us to model the data as a superposition of independent functions representing different structures:

$$k(\mathbf{x}, \mathbf{x}') = k_{Lin}(\mathbf{x}, \mathbf{x}') + k_{RQ}(\mathbf{x}, \mathbf{x}') + k_{Noise}(\mathbf{x}, \mathbf{x}') \quad (5)$$

$$k_{Lin}(\mathbf{x}, \mathbf{x}') = \sigma_0^2 + (\mathbf{x}^\top \mathbf{x}') \quad (6)$$

$$k_{RQ}(\mathbf{x}, \mathbf{x}') = \sigma^2 \exp \left(1 + \frac{\|\mathbf{x} - \mathbf{x}'\|^2}{2\alpha\ell^2} \right)^{-\alpha} \quad (7)$$

For the noise model (k_{Noise}) we use the sum of a squared exponential (k_{SE}) contribution and an independent component

$$k_{Noise}(\mathbf{x}, \mathbf{x}') = \sigma_n^2 \exp \left(-\frac{\|\mathbf{x} - \mathbf{x}'\|^2}{2\ell_n^2} \right) + \sigma_l^2 \delta_{ii'} \quad (8)$$

The hyperparameters are σ_0 (offset of the model), σ , σ_n and σ_l (magnitudes), α (relative weighing), ℓ and ℓ_n (length-scales). The reliability of the regression is dependent on how well we select the covariance function and therefore the covariance hyperparameters θ . The hyperparameters are selected by minimising the negative log marginal likelihood with respect to θ . Since by assumption the distribution of the data is Gaussian, the log marginal likelihood is:

$$\mathcal{L} = \log p(\mathbf{y}|\mathbf{x}, \theta) = -\frac{1}{2} \mathbf{y}^\top k(\mathbf{X}, \mathbf{X})^{-1} \mathbf{y} - \frac{1}{2} \log |k(\mathbf{X}, \mathbf{X})| - \frac{n}{2} \log 2\pi \quad (9)$$

The values of the hyperparameters that optimize the marginal likelihood, are found using its partial derivative in conjunction with a numerical optimization routine based on conjugate gradients. We refer to Chapters 2 and 5 in Rasmussen and Williams (2006) for a detailed description of GP models.

The major limitation of GP regression is the computational complexity, since it requires matrix

inversion, which has a complexity of $\mathcal{O}(n^3)$, where n is the number of training data points.

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Different solutions have been proposed to cope with this problem (e.g. Higdon, 1998; Rasmussen and Ghahramani, 2002; Snelson and Ghahramani, 2006). In this work, in the case of 5 min data, we have used the FITC approximation (Snelson and Ghahramani, 2006).

2.3.2. Estimation strategies

Figure 2 depicts the three different strategies that we have employed to estimate UFP at each site in the monitoring network. For the sake of brevity, we show only the estimation of UFP number concentration (target of the model) at site 1 (black dot) in each panel of Figure 2.

Considering the high cost of the pollutant monitors (~10,000 Euro), we have evaluated models that use covariates from only one monitor at each site. The inclusion of additional covariates requires the inclusion of one monitor for each covariate, increasing the costs of the instrumentation and maintenance.

Local estimation: At each site, we use local measurements of NO_x, O₃ and CO (local covariates) to estimate UFP at the same site.

Remote estimation: In this strategy, we use either UFP or NO_x measurements from two sites to estimate UFP at a third site. For this strategy, we evaluate the models for the cases in which UFP measurements are either included or not in the set of covariates.

Mixed estimation: In this strategy, we use combinations of local gaseous pollutants measurements (local covariates) and remote UFP or gaseous pollutants measurements (remote covariates) to estimate UFP at the target site. Also for this strategy, we evaluate the models for the cases in which remote UFP number concentration measurements are either included or not in the set of covariates.

Figure 2: Estimation strategies and their set of covariates for: local estimation (left), remote estimation (middle), and mixed estimation (right). Black dots depict the site of the UFP estimation.

2.4. Model evaluation

In order to evaluate the model we have followed the steps suggested by Bennett et al. (2013). First of all, we have divided the dataset at each site into two disjoint datasets. The data collected during the first two weeks of the measurement campaign have been used as training set ($\mathcal{D}_{\text{train}}$). The data collected during the third and fourth week of the measurement campaign have been used as unseen data to evaluate the proposed model ($\mathcal{D}_{\text{eval}}$). At site 2, the evaluation dataset is limited to 9 days due to monitor malfunctioning. In the second step, we have used the highest marginal likelihood (ML) to select the models that have at the same time a good fit and a low complexity. At this stage, we have compared models (at half hour resolution) based on the maximum length (14 days) of training. In the third step, we have evaluated the selected models

in terms of their ability to estimate unseen measurements. We have used the R^2 and RMSE metrics because their wide usage aids communication of the model performance.

2.4.1. Marginal Likelihood (ML)

The log marginal likelihood is

$$\log p(\mathbf{y}|\mathbf{x}, \boldsymbol{\theta}) = -\frac{1}{2}\mathbf{y}^\top \mathbf{k}(\mathbf{X}, \mathbf{X})^{-1}\mathbf{y} - \frac{1}{2}\log|\mathbf{k}(\mathbf{X}, \mathbf{X})| - \frac{n}{2}\log 2\pi \quad (14)$$

The first term gives a measure of the quality of the model fit. It is the only term that involves observed targets. The second term is a complexity penalty term, which measures and penalizes the complexity of the model. The third term is a log normalization term. Models with a higher ML should be preferred to models with a lower ML.

2.4.2. Coefficient of determination (R^2)

The coefficient of determination R^2 indicates the fraction of variance of observations explained by the model:

$$R^2 = 1 - \frac{\sum_{m=1}^M (y_m - y_*)^2}{\sum_{m=1}^M (y_m - \bar{y})^2} \quad (15)$$

where y_m and y_* are the measured and estimated UFP; $\bar{y}$ is the mean of the observed UFP; M is the number of evaluation measurements.

2.4.3. Root Mean Square Error (RMSE)

The root mean square error (RMSE) is calculated as the difference between the measured UFP and the estimated ones:

$$\text{RMSE} = \sqrt{\frac{1}{M} \sum_{m=1}^M (y_m - y_*)^2} \quad (16)$$

where y_m and y_* are the measured and estimated UFP and M is the number of evaluation measurements.

3. Results and discussion

In this section, first we present a statistical summary of UFP concentrations, diameters and gaseous pollutant concentrations recorded over the entire sampling period. Second, for each model strategy, we show the model selection results (based on ML). Third, we evaluate and discuss the performance of the selected GP models by comparing the estimated UFP with the measured ones. We conclude by assessing the models on different amounts of training data at half hour resolution and their performance at 5 min resolution. All the results are based on log-transformed and standardized data with zero mean and unit variance.

3.1 Summary statistics

Table 2 shows that the UFP concentrations are within the same range at all three sites, although traffic density at site 3 is almost one order of magnitude higher than that at the other two sites (Table 1). Dilution effects are leading to the lower UFP concentration at site 3 compared to what would have been expected from the traffic counts. The summary of data in Table 2 confirms that the traffic volume is not the only decisive factor in the variations of UFP. The distance from the moving traffic, the site morphology and dispersion conditions specific to individual sampling locations are also contributing factors (Kumar et al., 2014). These observations are also in agreement with the findings of Kumar et al. (2008), Buonanno et al. (2009), Kumar et al. (2009), Buonanno et al. (2011), Fujitani et al. (2012) and Peters et al. (2013).

The mean UFP diameters at the sites are also similar and vary between 48 nm (site 3) and 52 nm (site1), with the maximum values ranging between 80 nm (sites 2 and 3) and 96 nm (site 1).

The NO_x, CO and O₃ concentrations (according to the medians and the inter-quartile ranges) are similar at all three sites. The higher traffic intensity at site 3 as compared to sites 1 and 2 is again probably offset by the larger distance to the traffic (Table 1) and resulting pollutant dilution.

Table 2: Summary statistics of UFP number concentrations measured at the three sites.

	Variable	Mean	Std	Median	Min	Max	Q1	Q3
Site 1	UFP (#cm ⁻³)	22,810	12,934	20,628	1768	88,004	13,190	29,316
	UFP diameter (nm)	52	9	50	32	96	46	57
	NO (µg m ⁻³)	57	69	36	0	571	14	70
	NO ₂ (µg m ⁻³)	56	26	54	5	150	36	73
	O ₃ (µg m ⁻³)	32	21	32	1	88	14	49
	CO (µg m ⁻³)	435	220	376	84	1658	286	515
Site 2	UFP (#cm ⁻³)	21,586	11,249	19,278	2168	80,355	13,866	27,486
	UFP diameter (nm)	51	8	49	29	80	44	56
	NO (µg m ⁻³)	78	80	53	1	716	29	97
	NO ₂ (µg m ⁻³)	72	30	70	11	170	50	89
	O ₃ (µg m ⁻³)	27	19	23	3	83	9	41
	CO (µg m ⁻³)	-	-	-	-	-	-	-
Site 3	UFP (#cm ⁻³)	23,219	14,129	19,518	2528	87,210	13,703	28,883
	UFP diameter (nm)	48	8	46	28	80	42	53
	NO (µg m ⁻³)	69	101	31	1	854	11	85
	NO ₂ (µg m ⁻³)	62	33	57	7	218	37	82
	O ₃ (µg m ⁻³)	30	21	26	1	91	10	47
	CO (µg m ⁻³)	322	221	270	25	1606	164	403

3.2 Model selection and evaluation

3.2.1 Local estimation

The ML metric (based on $\mathcal{D}_{\text{train}}$; Table 3) reveals that the models that use NO_x as covariates (GP_{S_n}(NO_x_{S_n}), $n = 1, \dots, 3$ in bold) outperform the models that use O₃ (GP_{S_n}(O₃_{S_n}), $n = 1, \dots, 3$) or CO as covariates (GP_{S_m}(CO_{S_m}), $m = 1, \dots, 2$). Moreover, the results based on the unseen measurements (R^2 and RMSE metrics in Table 3) confirm that the selected models

($GP_{Sn}(NO_{xSn})$) outperform the others, and they show a good correspondence between the modelled and the measured values. At all three sites, the models explain between 87% (site 2) and 90% (site 1) of the variance.

These results are probably due to the strong correlation of UFP with NO_x . More in detail, road vehicles are the major sources of UFP in urban environments (Kumar et al., 2014; Harrison et al., 2011; Pey et al., 2009). These vehicles also generate NO (from all types of combustion engines) and primary NO_2 (especially from diesel cars equipped with after treatment technologies including oxidation catalysts) at the same time. Moreover, the Belgian car fleet presents a high share of diesel vehicles (64.3%; Beckx et al., 2013), which have high emission of both UFP and NO_x (Beddows and Harrison, 2008).

From Figure 3, we can observe that the model tends to underestimate high and low values of UFP at site 2 as opposed to underestimation of low values at site 3. It should be emphasized that these deviations are not substantial, and the estimated distributions seem to describe the measurements well. In particular, the models do not tend to underestimate high concentrations.

In summary: the GP models that use NO_x as covariates outperform the models that use CO and O_3 as covariates.

Table 3: Local estimation: evaluation of the models at half hour resolution and 14 days of training in terms of ML, R^2 and RMSE. In bold are denoted the models with the highest ML.

Target	Model	Local Covariates	R^2	RMSE	ML	$\mathcal{D}_{eval}$ (days)
UFP Site 1	$GP_{S1}(NO_{xS1})$	NO/NO_2	0.90	0.35	-115	14
	$GP_{S1}(O_{3S1})$	O_3	0.57	0.67	-466	14
	$GP_{S1}(CO_{S1})$	CO	0.55	0.67	-511	14
UFP Site 2	$GP_{S2}(NO_{xS2})$	NO/NO_2	0.87	0.45	-168	9
	$GP_{S2}(O_{3S2})$	O_3	0.53	0.73	-392	9
UFP Site 3	$GP_{S3}(NO_{xS3})$	NO/NO_2	0.88	0.39	-403	14
	$GP_{S3}(O_{3S3})$	O_3	0.65	0.62	-520	14

Figure 3: Local estimation. The left column shows the time series plots of the estimated and the measured UFP number concentrations. The dashed grey line is the estimated UFP number concentration and the black line is the measured UFP number concentration relative to the evaluation period ($\mathcal{D}_{eval}$). The middle column shows the scatterplots, R^2 and RMSE between the estimated and measured UFP number concentrations. The grey lines have slope 1 and an intercept of 0 (ideal case, when the estimated and measured values are equal). The dashed grey lines delimit the FAC2 area. The right column shows the QQ plots between the estimated and measured UFP number concentrations. The top row refers to site 1, the middle row refers to site

2 and the bottom row refers to site 3.

3.2.2 Remote estimation

Tables 4 and 5 show the results obtained in the remote estimation configuration. In Table 4, the evaluation refers to those models that use UFP data recorded at any of the two sites (remote covariates) to estimate UFP at a third site. In Table 5, the evaluation refers to those models that use NO_x measurements as remote covariates to estimate UFP at a third site.

The models selected in the training phase (higher ML), at all three sites, are the ones that use both UFP measurements recorded at the other two sites (in bold in Table 4). Moreover, the results based on the unseen measurements (R^2 and RMSE metrics in Tables 4 and 5) confirm that the selected models outperform the others. Those models explain between 69% (site 1) and 87% (site 2) of the variance.

Comparison of these results with those obtained in the local estimation configuration (Tables 3) shows that the model performances at sites 1 and 3 are weaker compared with the local estimation and similar at site 2. The weaker performance at two sites can be explained by the absence of local covariates.

Table 4: Remote estimation based on UFP covariates: evaluation of the models at half hour resolution and 14 days of training in terms of ML, R^2 and RMSE. In bold are denoted the models with the highest ML.

Target	Model	Remote Covariates			R^2	RMSE	ML	$\mathcal{D}_{\text{eval}}$ (days)
		UFP Site 1	UFP Site 2	UFP Site 3				
UFP Site 1	GP_{S1}(UFP_{S2}, UFP_{S3})		X	X	0.69	0.58	-398	14
	GP _{S1} (UFP _{S2})		X		0.68	0.58	-433	14
	GP _{S1} (UFP _{S3})			X	0.58	0.65	-557	14
UFP Site 2	GP_{S2}(UFP_{S1}, UFP_{S3})	X		X	0.87	0.35	-190	14
	GP _{S2} (UFP _{S1})	X			0.65	0.59	-383	14
	GP _{S2} (UFP _{S3})			X	0.82	0.42	-345	14

UFP	GP_{S3}(UFP_{S1}, UFP_{S2})	X	X	0.81	0.42	-423	14
Site 3	GP _{S3} (UFP _{S1})	X		0.56	0.68	-526	14
	GP _{S3} (UFP _{S2})		X	0.80	0.45	-442	14

Table 5: Remote estimation based on NO/NO₂ covariates: evaluation of the models at half hour resolution and 14 days of training in terms of ML, R² and RMSE.

Target	Model	Remote Covariates			R ²	RMSE	ML	$\mathcal{D}_{\text{eval}}$ (days)
		NO/NO ₂ Site 1	NO/NO ₂ Site 2	NO/NO ₂ Site 3				
UFP Site 1	GP _{S1} (NOx _{S2} , NOx _{S3})		X	X	0.67	0.61	-405	9
	GP _{S1} (NOx _{S2})		X		0.67	0.63	-440	9
	GP _{S1} (NOx _{S3})			X	0.61	0.76	-556	14
UFP Site 2	GP _{S2} (NOx _{S1} , NOx _{S3})	X		X	0.80	0.47	-280	14
	GP _{S2} (NOx _{S1})	X			0.76	0.51	-408	14
	GP _{S2} (NOx _{S3})			X	0.74	0.52	-454	14
UFP Site 3	GP _{S3} (NOx _{S1} , NOx _{S2})	X	X		0.80	0.47	-443	9
	GP _{S3} (NOx _{S1})	X			0.79	0.49	-462	9
	GP _{S3} (NOx _{S2})		X		0.68	0.57	-489	14

In the case of models that use NOx measurements (Table 5) recorded at two sites (remote covariates) to estimate UFP at a third site, the best models are obtained using remote NOx measurements from two sites simultaneously. Those models have a similar performance, at sites 1 and 3, and worse, at site 2, than that of models that use UFP as covariates, and they explain between 67% (site 1) and 80% (sites 2 and 3) of the variance.

We would like to point out that caution has to be taken when comparing the model performances reported in Tables 4 and 5. At site 2, gaseous measurements are limited to 9 days due to monitor malfunctioning (Section 2.4). Therefore, the performance of the models, which use NOx covariates recorded at site 2, are computed using a shorter dataset ($\mathcal{D}_{\text{eval}}$) than the others (9 days instead of 14 days).

Figure 4: Remote estimation. The left column shows the time series plots of the estimated and the measured UFP number concentrations. The dashed grey line is the estimated UFP number concentration and the black line is the measured UFP number concentration relative to the evaluation period ($\mathcal{D}_{\text{eval}}$). The middle column shows the scatterplots, R^2 and RMSE between the estimated and measured UFP number concentrations. The grey lines have slope 1 and an intercept of 0 (ideal case, when the estimated and measured values are equal). The dashed grey lines delimit the FAC2 area. The right column shows the QQ plots between the estimated and measured UFP number concentrations. The top row refers to site 1, the middle row refers to site 2 and the bottom row refers to site 3.

Tables 4 and 5 also show that the models based on two remote locations are better performing than models based on covariates from one remote location. For example, at sites 1 and 3, the models that use the remote covariates from site 2 have a similar performance as the ones that use the covariates from the other two remote sites simultaneously. On the other hand, at site 1, the models that use the covariates from site 3, and at site 3, the models that use the covariates from site 1, have a weaker performance than the models that use the covariates from two remote sites simultaneously. At site 2 instead, all the models that use covariates from sites 1 or 3 have a weaker performance than the ones that use the covariates from two remote sites simultaneously. From Figure 4, we can observe that the model tends to overestimate low values of UFP at site 1 and underestimate low values at site 2.

In summary: (i) model results are comparable when using remote UFP only or when using remote NO_x only to estimate UFP at a distant location; (ii) models that use covariates from only one remote site have fair performance only if there is a priori knowledge of which of the two sites is more informative; (iii) models that use covariates from two remote sites do not need a priori knowledge of which of the two sites is more informative because the models learn at which covariate to give more importance during the training period, maximising the likelihood between the covariates and the target function.

3.2.3 *Mixed estimation*

Tables 6 and 7 show the performances of the models for the mixed estimation configuration.

In Table 6 the evaluation refers to models that use local gaseous covariates (NO_x, O₃ and CO recorded at the same site where the estimation are made) in addition to UFP concentrations recorded at the other two sites (remote covariates). Table 7 shows the results of cases where only remote NO_x (but not UFP) recorded at two sites are added to the local covariates (NO_x, O₃, CO).

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Comparison of Tables 3–6 shows that the performances of models are improved when the remote UFP are combined with the local gaseous covariates. The best performances (in bold in Table 6) are obtained using the local NO_x plus remote UFP; the models explain more than 90% of the variance at all sites.

The models that combine remote UFP with local O₃ or CO perform better either than the models that use only local O₃ and CO covariates (Table 3) or models based on remote UFP (Table 4).

Table 6: Mixed estimation: evaluation of the models at half hour resolution and 14 days of training in terms of ML, R^2 and RMSE. In bold are denoted the models with the highest ML.

Target	Model	Local Covariates	Remote Covariates			R^2	RMSE	ML	$\mathcal{D}_{eval}$ (days)
			UFP Site 1	UFP Site 2	UFP Site 3				
UFP Site 1	GP_{S1}(NO_x_{S1}, UFP_{S2}, UFP_{S3})	NO/NO₂		X	X	0.91	0.32	-114	14
	GP _{S1} (O ₃ _{S1} , UFP _{S2} , UFP _{S3})	O ₃		X	X	0.70	0.58	-282	14
	GP _{S1} (CO _{S1} , UFP _{S2} , UFP _{S3})	CO		X	X	0.77	0.50	-231	14
UFP Site 2	GP_{S2}(NO_x_{S2}, UFP_{S1}, UFP_{S3})	NO/NO₂	X		X	0.91	0.35	-69	9
	GP _{S2} (O ₃ _{S2} , UFP _{S1} , UFP _{S3})	O ₃	X		X	0.89	0.34	-177	9
UFP Site 3	GP_{S3}(NO_x_{S3}, UFP_{S1}, UFP_{S2})	NO/NO₂	X	X		0.92	0.32	-265	14
	GP _{S3} (O ₃ _{S3} , UFP _{S1} , UFP _{S2})	O ₃	X	X		0.82	0.42	-404	14
	GP _{S3} (CO _{S3} , UFP _{S1} , UFP _{S2})	CO	X	X		0.80	0.50	-367	14

Table 7: Mixed estimation: evaluation of the models at half hour resolution and 14 days of training in terms of ML, R^2 and RMSE.

Target	Model	Local Covariates	Remote Covariates			R^2	RMSE	ML	$\mathcal{D}_{eval}$ (days)
			NO/NO ₂ Site 1	NO/NO ₂ Site 2	NO/NO ₂ Site 3				
UFP Site 1	GP _{S1} (NO _x _{S1} , NO _x _{S2} , NO _x _{S3})	NO/NO ₂		X	X	0.91	0.35	-136	9
	GP _{S1} (O ₃ _{S1} , NO _x _{S2} , NO _x _{S3})	O ₃		X	X	0.74	0.62	-340	9
	GP _{S1} (CO _{S1} , NO _x _{S2} , NO _x _{S3})	CO		X	X	0.81	0.53	-299	9
UFP Site 2	GP _{S2} (NO _x _{S2} , NO _x _{S1} , NO _x _{S3})	NO/NO ₂	X		X	0.84	0.42	-188	9
	GP _{S2} (O ₃ _{S2} , NO _x _{S1} , NO _x _{S3})	O ₃	X		X	0.80	0.48	-275	9
UFP Site 3	GP _{S3} (NO _x _{S3} , NO _x _{S1} , NO _x _{S2})	NO/NO ₂	X	X		0.89	0.39	-332	9
	GP _{S3} (O ₃ _{S3} , NO _x _{S1} , NO _x _{S2})	O ₃	X	X		0.82	0.44	-411	9
	GP _{S3} (CO _{S3} , NO _x _{S1} , NO _x _{S2})	CO	X	X		0.80	0.50	-412	9

Comparison between Tables 3 and 7 shows that the models that use NO_x measurements from all

the sites have similar performances compared to the models that use only the local covariates. In

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other words, the remote NO_x measurements are not improving the estimations based on local gaseous components only. On the other hand, comparing Tables 3, 5 and 7, we note that models that combine remote NO_x with local O₃ and CO perform better either than models that use local O₃ and CO or models based on remote NO_x.

From Figure 5, we can observe that the model tends to underestimate low and high values of UFP at site 2. However, these deviations are not substantial, and the estimated distributions seem to describe the measurements well.

In summary: (i) the addition of remote UFP to local NO_x results in improved model performance; (ii) the addition of remote NO_x to local NO_x does not improve the estimation based on local NO_x measurements; (iii) the addition of remote UFP or NO_x to local O₃ or CO results in improved estimations compared to models that use only local O₃ or CO measurements.

3.3 *Training length*

In practical situations such as designing the measurement campaign and planning the facilities needed, it is useful to know how the model performs according to the amount of data used for training. In Figure 6, the model performance for each site and for each monitoring strategy is evaluated on different days of training at 30 min resolution (solid lines). One day of training refers to the day before the first day of evaluation, two days of training means two days before the first day of evaluation and so on up to 14 days.

The plots show that the performance of models increases with the training length. It seems that a training period of at least seven days (in which at least two days correspond to weekend days) is suitable (in terms of a trade-off between costs and model performance) to let the model learn the UFP dynamics in different typologies of traffic.

Figure 5: Mixed estimation. The left column shows the time series plots of the estimated and the measured UFP number concentrations. The dashed grey line is the estimated UFP number concentration and the black line is the measured UFP number concentration relative to the evaluation period ($\mathcal{D}_{eval}$). The middle column shows the scatterplots, R^2 and RMSE between the estimated and measured UFP number concentrations. The grey lines have slope 1 and an intercept of 0 (ideal case, when the estimated and measured values are equal). The dashed grey lines delimit the FAC2 area. The right column shows the QQ plots between the estimated and measured UFP number concentrations. The top row refers to site 1, the middle row refers to site 2 and the bottom row refers to site 3.

3.4 *Models at 5 min resolution*

All the above results are based on half hour resolution. Considering the high variability of UFP, it is also interesting to have models with a higher time resolution. In Figure 6, the performances of models for each site and for each monitoring strategy are evaluated on different days of training for models at 5 min resolution (dashed lines). The results of these models, as for the half hour models, show a good correspondence of the modelled UFP values with the measured values. Furthermore, the local and mixed estimation models explain up to 85% of the variance, and the remote estimation around 60%, at site 1. At site 2, the mixed estimation model explains up 85% of the variance, and the local and remote models up to 78% of the variance. At site 3, the mixed estimation model explains up to 90% of the variance, the local estimation model explains up to 86% of the variance and the remote estimation model explains up to 72% of the variance.

3.5 *Network complexity*

The three estimation strategies have different levels of complexity. In the local estimation, at the estimation site, this strategy requires the presence of the local covariate monitors or sensors (e.g. NO_x) for the whole period (training and estimation), plus the UFP monitor for the training period. The remote estimation strategy requires local UFP for the training, and remote NO_x or UFP for the training and estimation periods. The mixed estimation requires UFP data at the estimation site for the training period, plus local NO_x and remote UFP or NO_x data for the training and estimation periods. This is, however, a costly solution, compared with the local estimation case, given the number of monitoring devices needed and a rather limited increase in estimation accuracy.

Figure 6: Performances of the GP models at half hour (solid lines) and 5 min (dashed lines) resolution evaluated on different days of training. First row: coefficient of determination (R^2); second row: root mean square error (RMSE). First column refers to site 1, middle column refers to site 2 and the right column refers to site 3. One day of training refers to the day before the first day of evaluation, two days of training means two days before the first day of evaluation and so on up to 14 days.

3.6 Limitations

The applied modelling approach also has its limitations. For instance, there is no guarantee that the proposed model structure is optimal. However, different covariates (e.g. traffic and

meteorological data) could be easily added to the proposed structure. For instance, considering that the rain influences the concentrations of gaseous pollutants and UFP differently, models that include weather conditions may have better performances.

The models are developed and trained in the first place for use in traffic locations within city boundaries. All three locations in this study are urban traffic locations, and their pollution profile is dominated by traffic emissions. The three locations are distinct from each other in terms of traffic intensity, distance to traffic and surrounding street pattern. We have tested the method simultaneously at these three different traffic locations, and results were found to be encouraging. Therefore, we assume that the proposed method could be applied to other traffic locations to address part of the spatial inhomogeneity of UFP between sites within a city reported in literature (Mejia et al. 2008; Buonanno et al., 2011; Mishra et al., 2012; Birmili et al., 2013; Kumar et al. 2014). However, this assumption could not be tested with the available data set. Moreover, this study cannot assess how models trained at one area/city perform in other areas/cities with different fleet composition, traffic dynamics and meteorological circumstances. The transferability of these models to other areas is probably limited when circumstances differ substantially. In that case, a new data collection period should be carried out for model training.

A further limitation of the used data set is that it is only one month long, and considering that half of it has been used for training, only half a month was left for the evaluation. This restricted the possibility to assess questions such as how long the proposed model will perform satisfactorily, and how often the training has to be performed.

The measurements used in this study were performed during a winter when the influence of photochemical reactions is rather limited. Considering that ratios of NO-NO₂-O₃ are strongly influenced by photochemistry, and secondary formation of UFP is partially driven by

atmospheric photochemical reactions and conversion (Westerdahl et al., 2005; Seinfeld and Pandis, 2006), it could be interesting to study their long-term performances by applying the proposed models on data sets that cover various seasons.

Finally, the lower cut-off limit of UFP used here does not account for the nucleation mode particles that are volatile and much more dynamic. It would be interesting to use the model on such data set to evaluate its performance.

4. Summary and conclusions

In this work, we investigated strategies to estimate UFP at specific locations based on concentrations of gaseous components at the same and remote locations, and/or UFP at remote locations. We have used Gaussian process regression to estimate UFP at three sites in an air pollution monitoring network.

In the local estimation, we found that the models that use NO_x have the best performances. This strategy would be especially interesting in case a dense network of low-cost gas sensors can be deployed: novel low-cost gas sensors are being developed with increasing level of performance (Mead et al., 2013, Kumar et al., 2015).

The case of the remote estimation reflects the situation where one tries to estimate UFP in locations where no local measurements are available. We used the measurements from two locations to estimate UFP at a third location. On a practical level this corresponds to the installation of permanent monitoring devices at two locations, and training the models at all similar locations of interest. The results also suggest that it is possible to estimate UFP at one location based on measurements of NO_x at two remote locations. This would give rise to the possibility to install a limited number of NO_x monitors at specific locations to estimate UFP at all similar locations in the same city.

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The case of the mixed estimation examines combinations of remote and local measurements to improve the model performance. This strategy requires the highest number of monitoring devices, and thus presents a trade-off between higher accuracy and increased costs. In practical terms we can conclude that estimations based on remote UFP are improved by adding local covariates to take into account local variability.

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