COMBINED USE OF SATELLITE OBSERVATIONS WITH URBAN SURFACE CHARACTERISTICS TO ESTIMATE PM CONCENTRATIONS BY EMPLOYING MIXED-EFFECTS MODELS

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ABSTRACT

Linear mixed effects models were developed for the estimation of the average daily Particulate Matter (PM) concentration spatial distribution over the area of Greater London (UK). Both fine (PM2.5) and coarse (PM10) concentrations were predicted for the 2002-2012 time period, based on satellite data. The latter included Aerosol Optical Thickness (AOT) at 3×3 km spatial resolution, as well as the Surface Relative Humidity, Surface Temperature and K-Index derived from MODIS (Moderate Resolution Imaging Spectroradiometer) sensor. For a meaningful interpretation of the association among these variables, all data were homogenized with regard to spatial support and geographic projection, thus addressing the change of support problem and leading to a valid statistical inference. To this end, spatial (2D) and spatio-temporal (3D) kriging techniques were applied to in-situ particulate matter concentrations and the leave-one-station-out cross-validation was performed on a daily level to gauge the quality of the predictions. Satellite-derived covariates displayed clear seasonal patterns; in order to work with data which is stationary in mean, for each covariate, deviations from its estimated annual profiles were computed using nonlinear least squares and nonlinear absolute deviations. High-resolution land-cover and morphology static datasets were additionally incorporated in the analysis in order to catch the effects of nearby emission sources and sequestration sites. For pairwise comparisons of the particulate matter concentration means at distinct land-cover classes, the pairwise comparisons method for unequal sample sizes, known as *Tukey'*s method, was performed. The use of satellite-derived products allowed better assessment of space-time interactions of PM, since these daily spatial measurements were able to capture differences in PM concentrations between grid cells, while the use of high-resolution land-cover and morphology static datasets allowed accounting for local industrial, domestic and traffic related air pollution. The developed methods are expected to fully exploit ESA's new Sentinel-3 observations to estimate spatial distributions of both PM10 and PM2.5 concentrations in arbitrary cities.

1. INTRODUCTION

The importance of Particulate Matter (PM) regarding urban air quality and corresponding human health problems has been studied and highlighted extensively in the past. Both fine (PM2.5) and coarse (PM10) particles high concentrations have been correlated with high rates of morbidity and mortality, caused primarily by respiratory and cardiovascular diseases (e.g. [1], [2]).

While in large Metropolitan areas, including the Greater Area of London, dense PM monitoring station networks exist and operate, large areas inside the city remain unmonitored, or highly dependent on the functionality and operational period of specific instruments. Satellite-based PM monitoring is the main approach to overcome these disadvantages of in situ measurements and provide regular assessments of the PM spatial distributions across large urban areas. In fact, the combined use of satellite observations, ground measurements and numerical modelling is among the most efficient approaches towards this goal [3].

The Aerosol Optical Thickness (AOT), which is a measure of the amount of airborne particles in the atmosphere, is the most widely used parameter in studies relating PM measurements with satellite products. Several different techniques have been developed for the association of surface PM with satellite-derived AOT (see [4] and references therein). The two main difficulties in this approach are the correlation of surface PM with columnar AOT and the association between in situ PM and spatially coarse AOT products.

In the present study, mixed-effect models with day-specific and site-specific random effects were applied for the estimation of PM concentrations over the area of Greater London. Satellite products of AOT, Surface Temperature (STMP), Relative Humidity (RHUM) and the K-Index (KIND) derived from the MODIS (Moderate Resolution Imaging Spectroradiometer) sensor were used. These products and their pre-processing are described in the next Section, along with surface parameters, including Land Use/Cover (LUC) data and the Sky View Factor (SVF). In Section 3 the developed methodology is presented, comprising the AOT validation, block kriging (used to address the change of support problem [5]) and the estimation of mixed-effects models, before the Results (Section 4) and Conclusions (Section 5).

2. STUDY AREA AND DATA

Fig. 1 shows the study area (black polygon), namely the Greater Area of London. The map also shows the spatial distribution of the PM10 monitoring stations and the London UCL-UAO AERONET station, used for validating the MODIS AOT (Section 3.1).

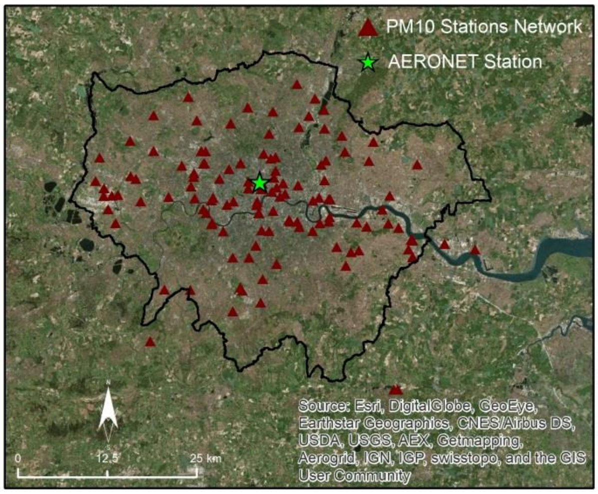

Figure 1. The Greater Area of London, along with the locations of the PM10 monitoring stations and the London UCL-UAO AERONET station.

LUC categorical data from the Urban Atlas [6] is used in this study to capture the emission sources of PM concentrations within the study domain. Fig. 2 shows 20 distinct classes of Urban Atlas for the study area, available in polygons and at 4 m × 4 m spatial resolution.

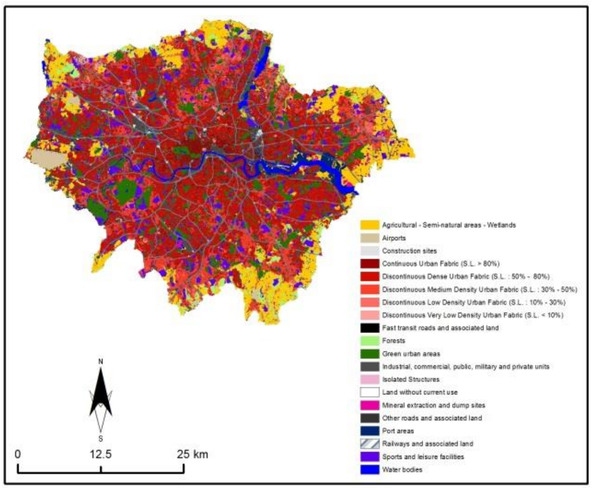

Figure 2. Urban Atlas LUC categories at the study area

The SVF (Fig. 3), used here as an indicator of the PM degree of dispersion, was computed as in [7], using a high spatial resolution Digital Surface Model [8].

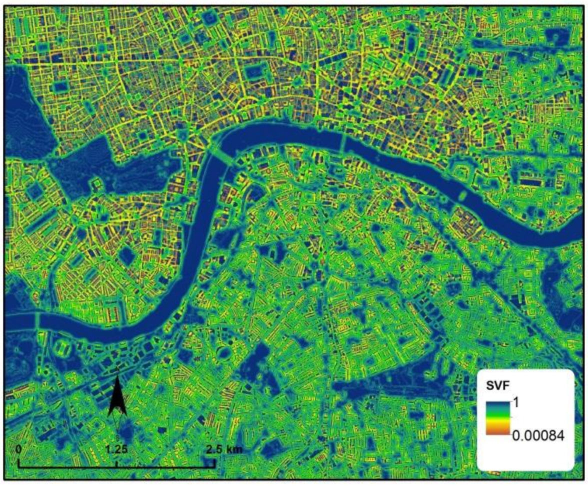

Figure 3. Sky View Factor spatial distribution at the central area of London.

For the estimation of STMP and RHUM, atmospheric temperature and relative humidity profiles were retrieved from the MODIS Level 2 Collection 6 Atmospheric Profile product, available during the entire period examined (2002-2012) at 20 pressure levels. Corresponding surface pressure data were used for the interpolation of these profiles at the surface level. The KIND, available from the same product, was used as an indicator of the aerosol vertical mixing height [9].

3. METHODOLOGY

3.1. AOT validation

The MODIS AOT was retrieved from the Level 2 Collection 6 Aerosol Product, available at 3 km × 3 km spatial resolution separately from Terra and Aqua satellites. While this product has been validated [10] results are reported using MODIS AOT values collocated with sun photometer AOT in both space and time. Here, however, PM is estimated on a daily average basis; hence, validation of the MODIS AOD against daily average AERONET measurements in the urban area of London is required to ensure the validity of corresponding PM results. For this purpose, average MODIS AOT from Terra and Aqua was validated against daily average AOT from the London-UCL-UAO AERONET station, available during the 2-year period 2009-2010.

A protocol similar to the one used in [11] was also used here. Specifically, after identification of the 3 km × 3 km pixel where the AERONET station is located, several grid sizes, centered on this pixel, for MODIS AOT spatial average computations were tested, ranging from single pixel (3 km × 3 km) to 13 × 13 pixels, which correspond to roughly 40 km × 40 km. In all cases, at least 20% of pixels with valid MODIS AOTs were required for the spatial average to be used in the validation. Similarly, only AERONET daily average AOTs computed from at least 10 measurements were regarded representative of the daily average and used in the process.

3.2. Spatial (2-D) and Spatio-Temporal (3-D) kriging

The average daily PM concentrations are expected to differ for specific LUC classes. In order to perform pairwise comparisons of PM concentrations means between distinct LUC categories, the pairwise comparisons method for unequal sample sizes, known as *Tukey*'s method, was applied. Thus, an adjusted to PM spatial variability post-classification of LUC product was developed based on statistically significant differences in means of PM between LUC classes. After merging post-classified land cover categories, a multiple linear regression was applied to log-transformed daily averaged PM concentrations with LUC and SVF as explanatory variables (Eq. 1).

$$\log\left(PM_{ij}\right) = a_0 \cdot SVF_i + a_i \cdot LUC_i + \varepsilon_{ij} \qquad (1)$$

The residuals obtained from Eq.1 were interpolated using 2-D ordinary kriging and 3-D metric kriging [12] techniques. In the case of 2-D kriging the spatial dependence patterns are allowed to change on a daily basis using separate daily variograms; 3-D kriging extends the geographical space to a 3-dimensional space-time environment: interpolation is based on a chosen number of nearest neighbours in the space-time cube.

Interpolation was followed by the back-transformation of the estimates combining regression predictions and kriged residuals. The sequential leave-one-station out cross-validation was performed on a daily-level and the Root Mean Square Error (RMSE), Mean Error (ME), Mean Absolute Error (MAE) and Pearson's Correlation Coefficient (R) metrics were computed. Rather than using predictions at the points where the stations were located, the geostatistical method known as block kriging [13] was used to predict average concentrations per 100 m x 100 m block. This approach has been used in previous works to link data with different spatial supports [14]. Finally, to match with MODIS AOT, the kriged 100 m × 100 m PM surface was upscaled at 3 km × 3 km resolution by computing the averages of every 30 × 30 pixels cell.

3.3. Linear Mixed-Effects Models

Homogenization of data with respect to their spatial support was followed by estimation of Linear Mixed Effects Models (LMM) with (a) day-specific (LMM1) and (b) site-specific (LMM2) random intercepts and slopes. The first model class (LMM1) was proposed in [15]; it allows day-to-day variability in daily PM ~ AOT, STMP, RHUM, KIND relationship using data from all days to stabilize the results. In this way, the satellite-derived products are calibrated on a daily basis with PM measurements at stations. In the second model (LMM2), the random effects represent site-specific deviations from the mean value of the parameters.

A clear seasonal pattern was observed in each satellite-derived product. The deviations from annual profiles (estimated by fitting a sinusoid to the time series) were computed to be used as covariates in the mixed-effects models. The model-building procedure proposed in [16] and [17] was adopted to decide which coefficients in the LMM1 and LMM2 models need random effects and which can be considered solely as fixed effects. Comparisons between models with different random effects structures were based on Maximum Likelihood ration tests, as well as on the Akaike and the Bayesian Information Criteria. The quality of both mixed-effects models was gauged using 5-fold cross-validation.

For the detailed information on the model development methodology applied to AOT derived from MERIS/AATSR synergy at 1 km × 1 km spatial resolution see [4].

4. RESULTS

Tab. 1 shows the results of the MODIS AOT validation for the grid sizes tested, which include correlation coefficients, intercepts, slopes and the corresponding confidence intervals. It is apparent from Tab. 1 that MODIS tends to overestimate AOT compared to AERONET daily averages. This tendency, however, is reduced as the test area size increases, while correlation between the data sets also improves significantly.

Table 1. Validation results of the MODIS AOT against daily average AOT from London AERONET station.

Grid size	R	Intercept	Slope
1×1 (3km)	0.81	-0.05±0.07	0.61±0.22
3×3 (9km)	0.78	-0.03±0.06	0.57±0.18
5×5 (15km)	0.94	-0.07±0.04	0.80±0.13
9×9 (27km)	0.95	-0.05±0.03	0.83±0.11
13×13 (39km)	0.95	-0.02±0.03	0.83±0.11

As expected, statistically significant differences between means of PM concentrations at some groups of land cover classes were discovered. Based on the resulting *p*-values, six land-cover classes were formed. Since the initial set of classes provided by the Urban Atlas LUC Map was too large (20 classes), in order to get estimates for the whole area, there was a need to empirically merge the rest of the classes (with no in situ observations) with these six groups. The final categorization is presented in Tab.2 and the merged land cover raster map is shown in Fig.4.

Table 2. Merged Land Cover Classes

Category	Land Cover Class
I	Agricultural – Semi-natural areas – Wetlands | Forests | Land without current use | Water bodies
II	Sports and leisure facilities | Discontinuous Very Low density Urban Fabric | Discontinuous Low Density Urban Fabric | Green urban areas
III	Airports | Discontinuous Medium Density Urban Fabric | Discontinuous Dense Urban Fabric
IV	Construction sites | Industrial, commercial, public, military and private units | Isolated Structures | Mineral extraction and dump sites
V	Fast transit roads and associated land | Other roads and associated land | Port areas | Railways and associated land
VI	Continuous Urban Fabric

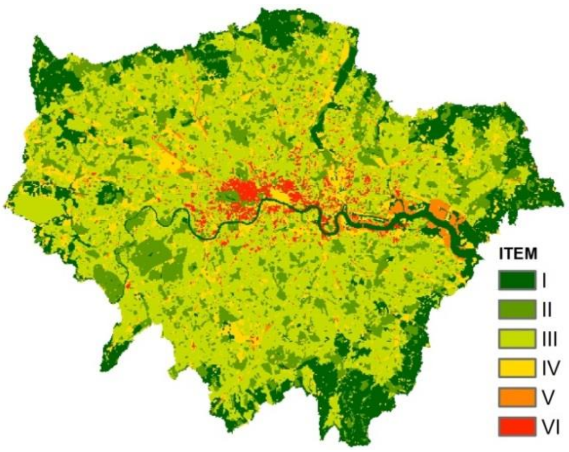

Figure 4. Post-classified merged LUC map

Interpolation of the residuals obtained from the Eq.1 has shown improved cross-validation performance, in comparison to kriging of log-transformed data. Furthermore, the large local variability observed in daily mean PM concentrations and the small number of in situ stations measuring PM2.5, resulted in singular variogram models in the 2-D kriging. Therefore, in the estimation as well as validation of the mixed-effects models, the interpolated values using 3-D kriging were used. The corresponding performance metrics are presented in Tab. 3.

Table 3. Cross-validation results of 3-D kriging

Metrics	RMSE	ME	MAE	R
PM10	9.00	-0.04	5.58	0.78
PM2.5	4.87	-0.05	3.38	0.81

In the left part of Fig. 5 and Fig.6 the effect of LUC and SVF on the spatial distribution of PM concentrations is presented: higher particle concentrations are observed in areas that generate emissions (e.g. continuous urban fabric areas) with low SVF values leading to constrained dispersion.

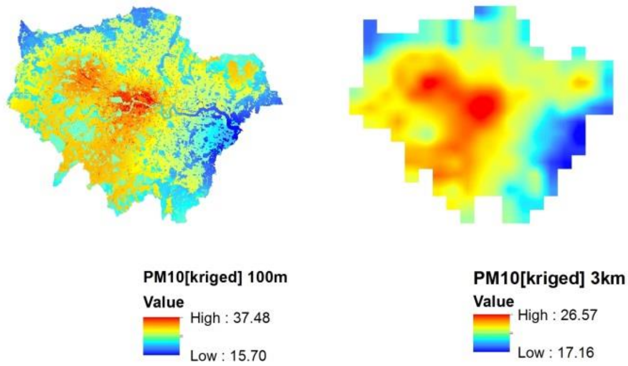

Figure 5. Example of interpolated daily PM10 (using 3D kriging)

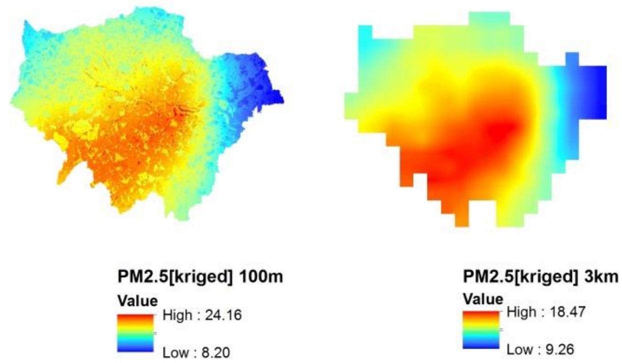

Figure 6. Example of interpolated daily PM2.5 (using 3D kriging)

Fig. 7 depicts estimated seasonal profiles using Nonlinear Least Squares and Nonlinear Absolute Deviations. The performance of median regressions was similar to the one achieved by conventional mean regression, suggesting the absence of outlying measurements. Moreover, the use of deviations from annual profiles as covariates in the LMM1 and LMM2 models resulted in improved out-of-sample performance, better explaining the variability of PM.

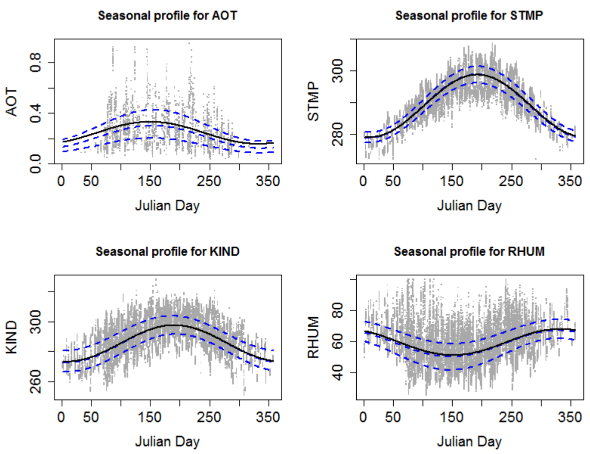

Figure 7.Estimated Seasonal Profiles

The (out-of-sample) predictive performance of LMM1 models was satisfactory: 5-fold cross-validated MAE=2.12 for PM10 and MAE=2.53 for PM2.5 concentrations with MAPE and MDAPE values considerably small in both cases (Tab. 4). Furthermore, the cross-validation results suggest that predictions were unbiased (ME = -0.02 for PM10 and ME=0.07 for PM2.5); this is vital when producing estimates to support risk assessment and epidemiological studies.

Table 4. Cross-validation results of LMM1 model

Metrics	ME	MAE	1-MAPE	1-MDAPE
PM10	-0.02	2.12	93.78%	95.21%
PM2.5	0.07	2.53	86.82%	89.58%

The predictive performance of LMM2 models is evidently inferior relative to LMM1, as shown in Tab. 5. However, use of such models is promising in the perspective of the fast evolving satellite remote sensing products. Application to temporally denser satellite observations, with measurements of higher quality, is expected to provide considerably improved results.

Table 5. Cross-validation results of LMM2 model

Metrics	ME	MAE	1-MAPE	1-MDAPE
PM10	0.01	7.58	76.03%	81.83%
PM2.5	0.08	4.96	72.43%	78.50%

Figure 8 shows the spatial pattern of estimated average PM10 concentrations derived from the satellite observations for the entire study period (2002-2012), using mixed-effects models with day-specific random effects at 3 km × 3 km resolution. Figure 9 shows the corresponding map of PM2.5. Both maps are presented partitioning the distribution of particulate matter concentration into ten equally sized bins. Predicted PM10 concentrations lie between 32.08 and 59.58 $\mu g m^{-3}$, whereas predicted PM2.5 values range from 8.43 to 25.49 $\mu g m^{-3}$.

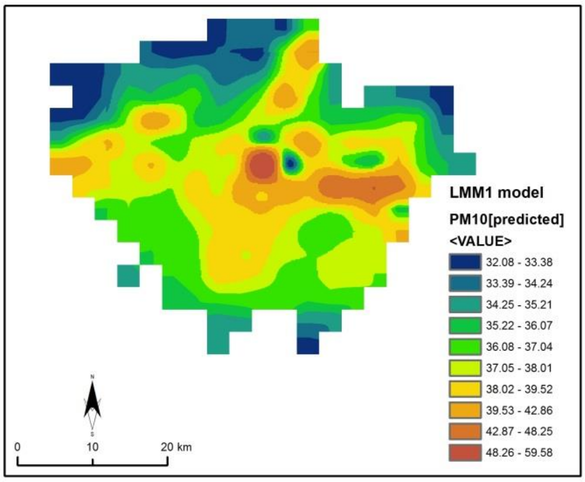

Figure 8.Predicted PM10

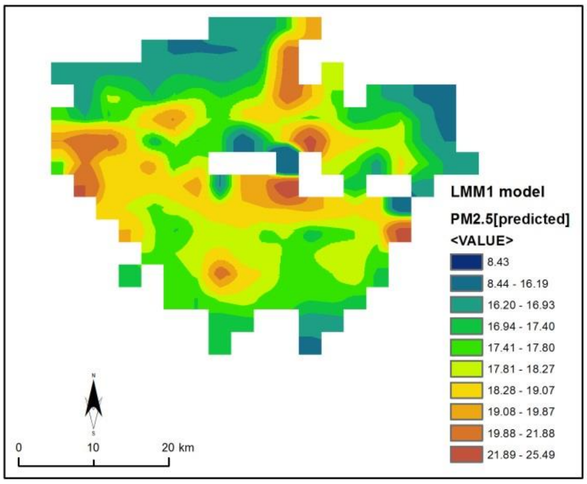

Figure 9.Predicted PM2.5

5. CONCLUSIONS

Surface PM10 and PM2.5 concentration patterns over the area of Greater London were estimated based on satellite-derived parameters and static data sets. The former comprised AOT, TEMP, RHUM and KIND from MODIS Level 2 Collection 6 products, while the latter included LUC and SVF, to account for local scale emission, sequestration and diffusion of particles in the city.

The results presented here are very promising, especially in view of the forthcoming Copernicus Sentinel-3 missions, scheduled to launch in 2015; observations from sensors on board these platforms will allow the derivation of the parameters used here at higher spatial and temporal resolution. Hence, the methodology developed here is expected to fully exploit these new satellite-derived products, while it can also be transferred to other urban areas of interest.

6. ACKNOWLEDGEMENTS

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