In the main text, "suburban" refers to low population areas, while "rural" refers to far from road areas.

Figure S1 voronoi map - the spatial distribution of NO2 measurement stations (global)

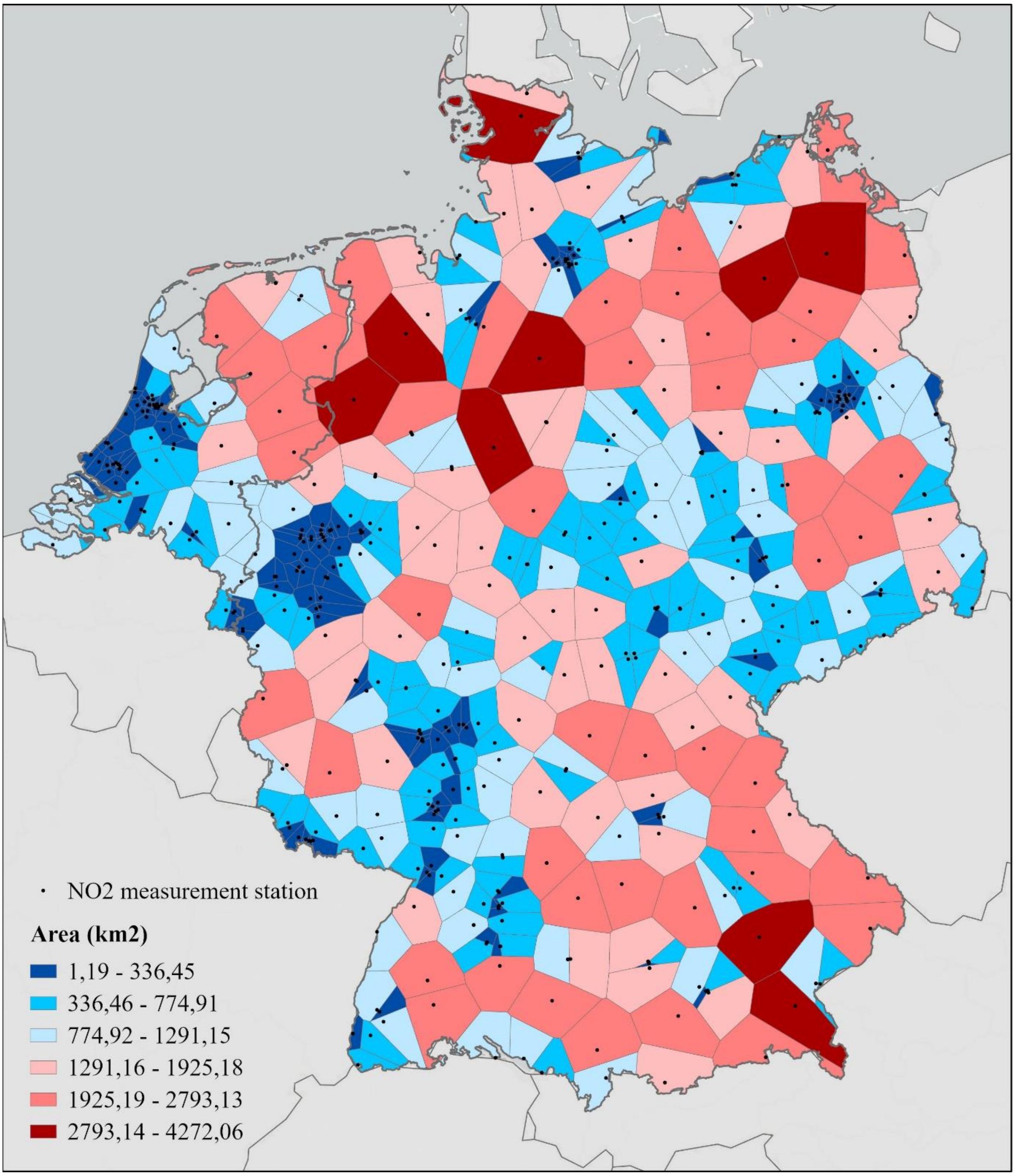


Figure S2 voronoi map - the spatial distribution of NO2 measurement stations (local)

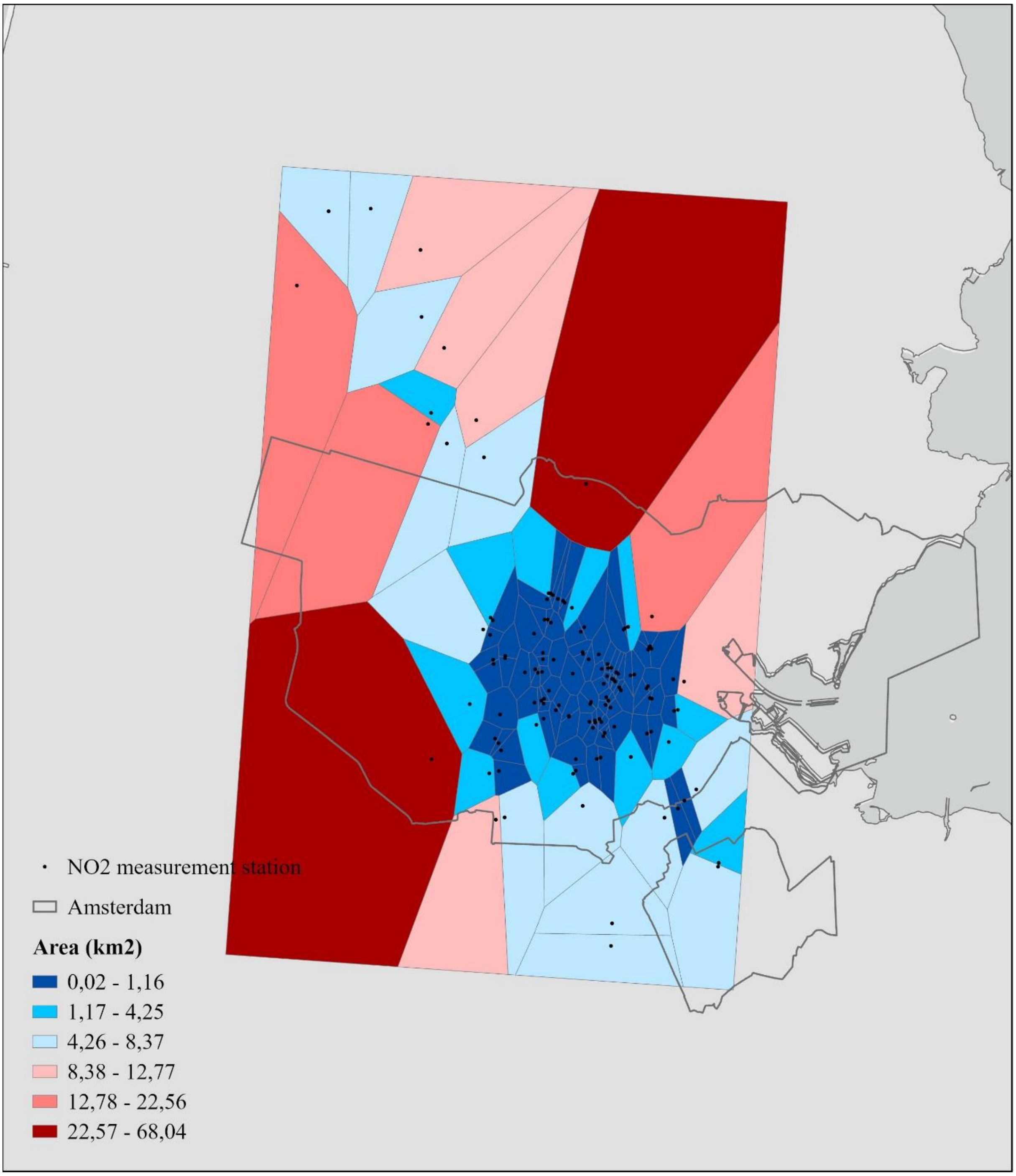
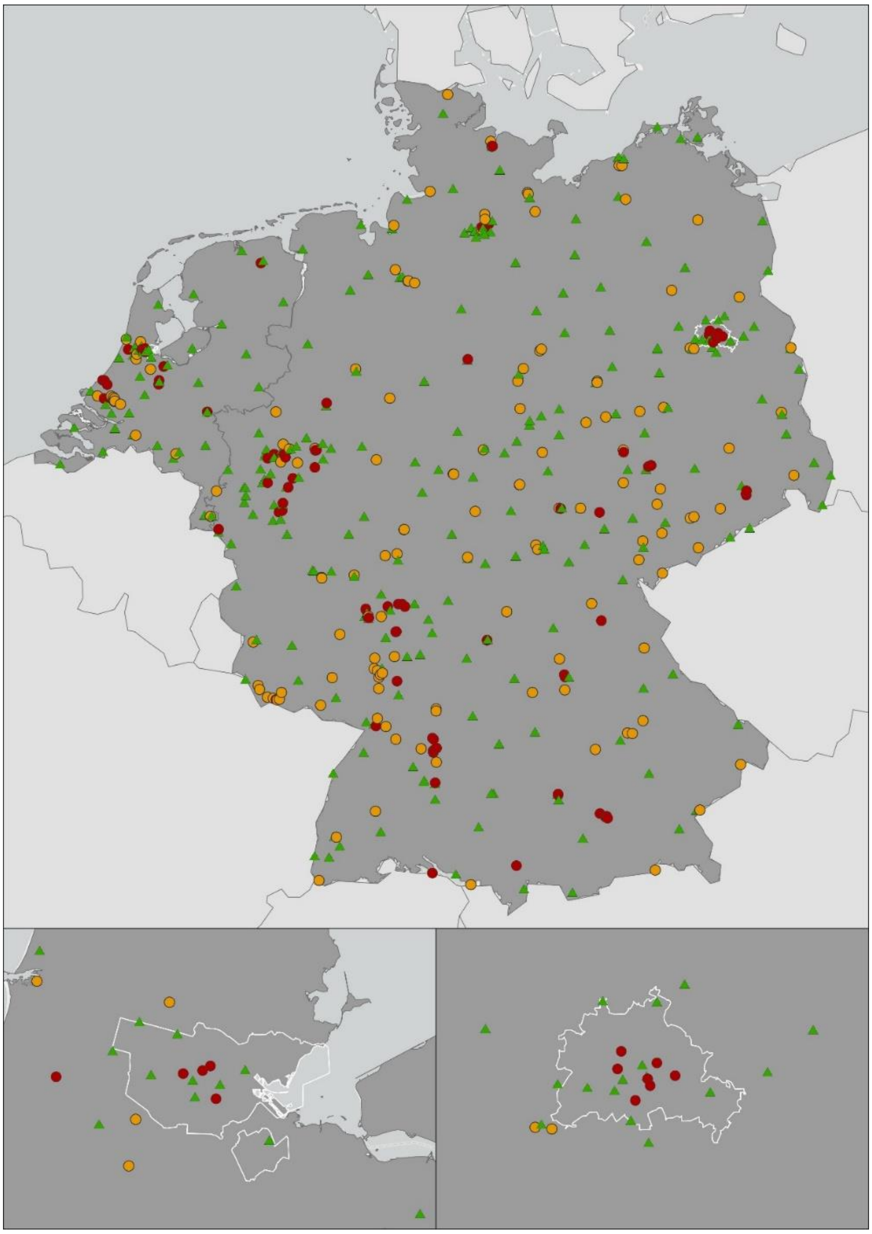


Figure S3 Spatial distribution per spatial group (global)



Spatial group

- Urban (close to road (<100m) ; in 25% highest populated areas)
- Suburban (close to road (<100m); in 75% lowest populated areas)
- Rural (>100m)

Figure S4 Spatial distribution per spatial group (local)

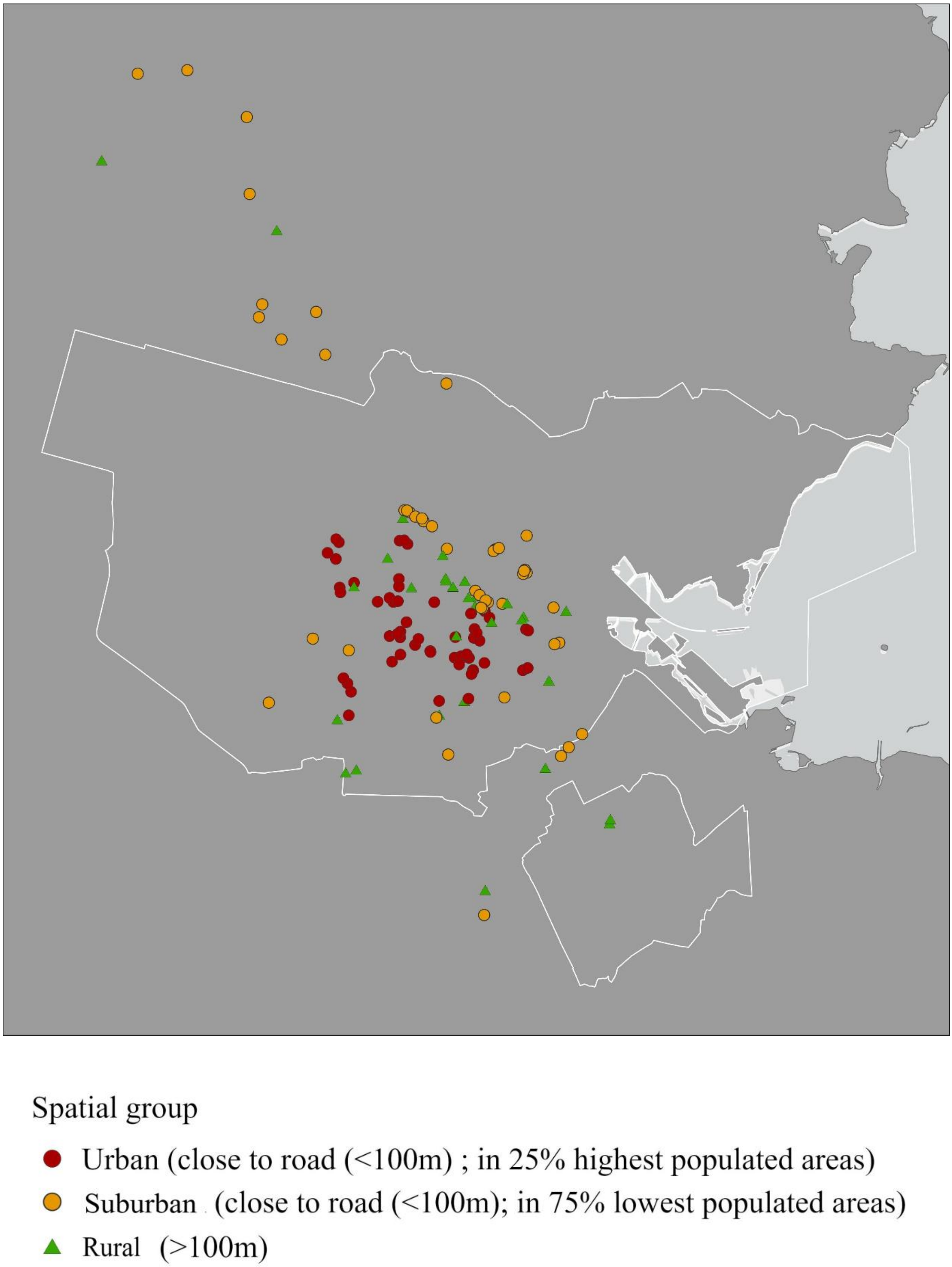


Figure S5 Spatial distribution of NO2 concentration values (global)

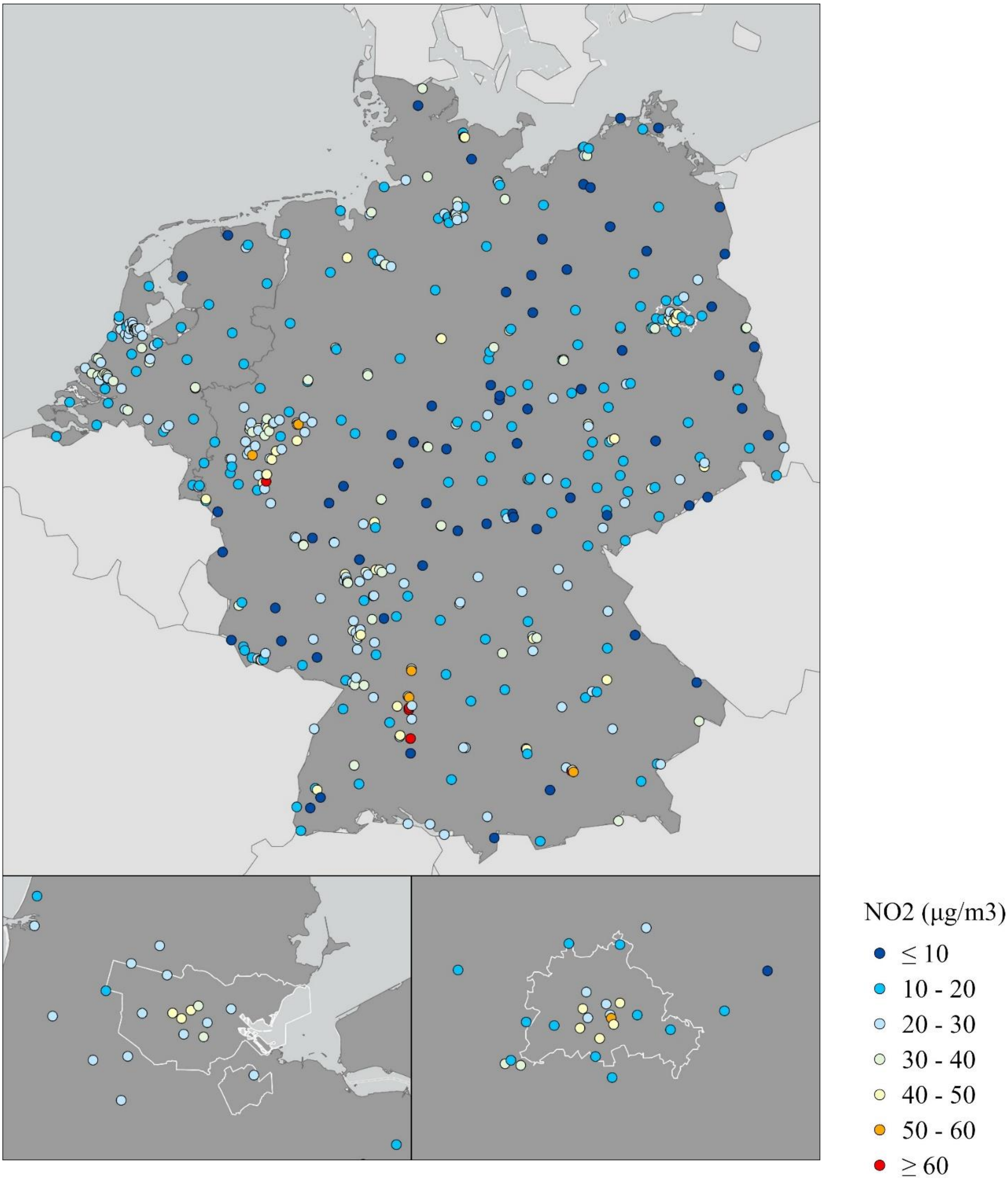
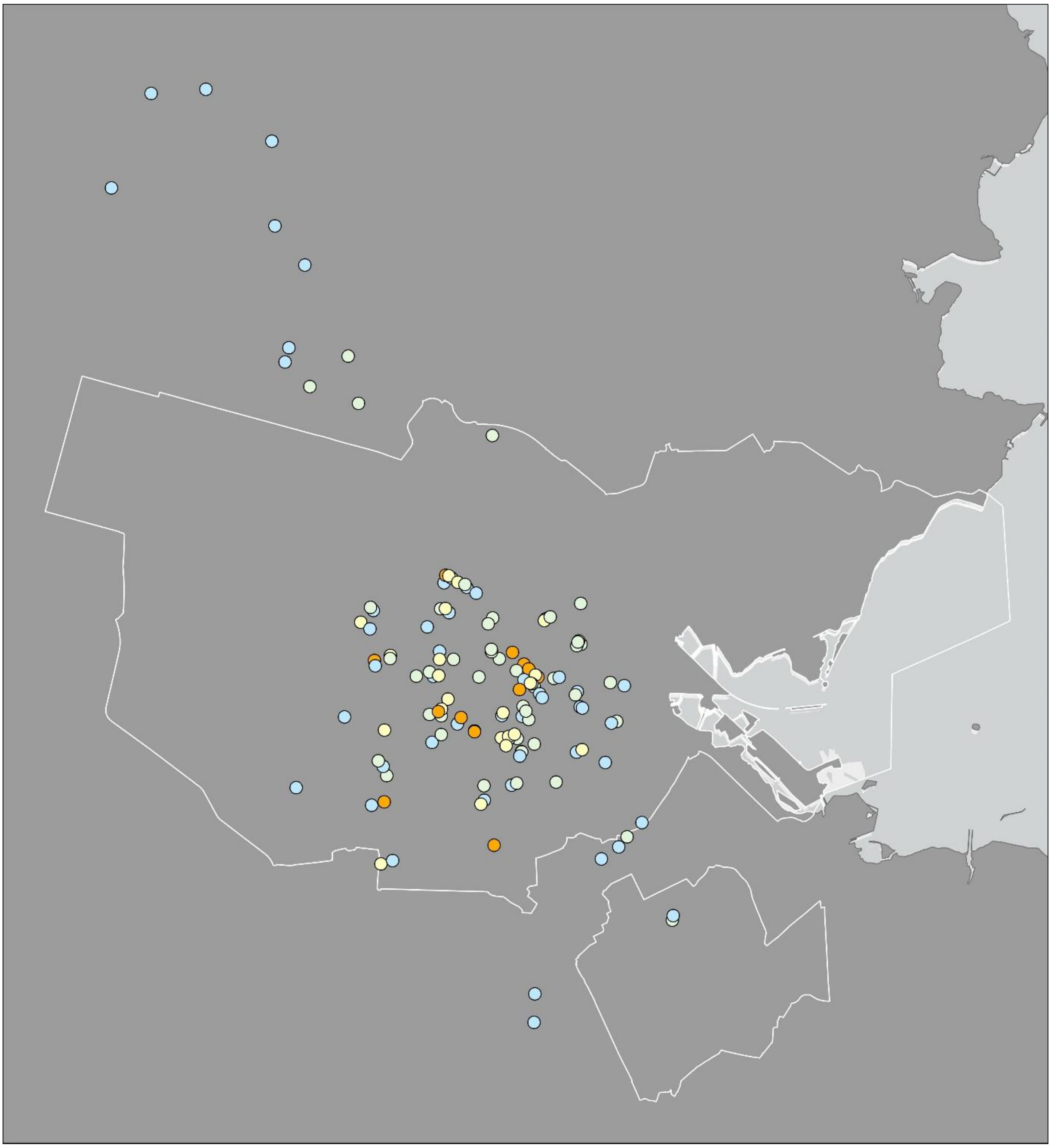


Figure S6 Spatial distribution of NO2 concentration values (local)

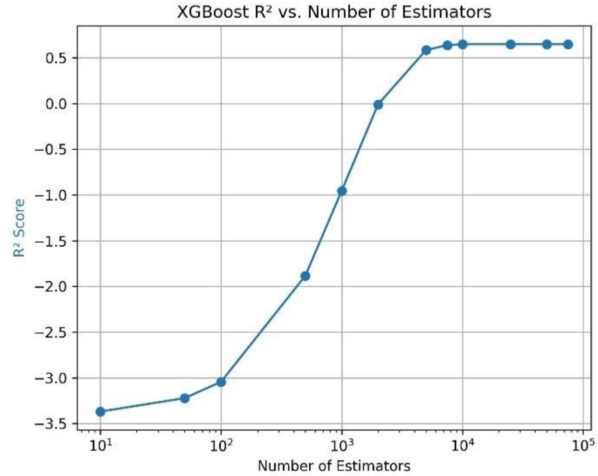


NO2 (µg/m3)

- ≤ 10
- 10 - 20
- 20 - 30
- 30 - 40
- 40 - 50
- 50 - 60
- ≥ 60

Figure S7 performance (r2) of the xgboost model as a function of number of estimators

Figure S8 performance (RMSE) of the xgboost model as a function of number of estimators



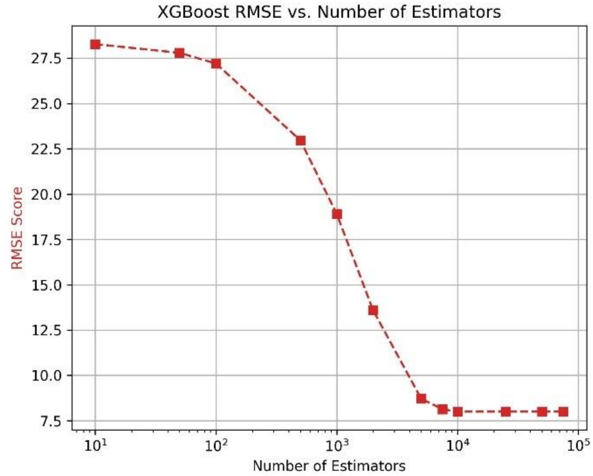


Figure S9 Out-of-sample performances evaluated using 20-fold repeated random sampling validation including LightGBM results – (global) - metric variability in R2

Rf = random forest, lgb = LightGBM, xgb = XGBoost

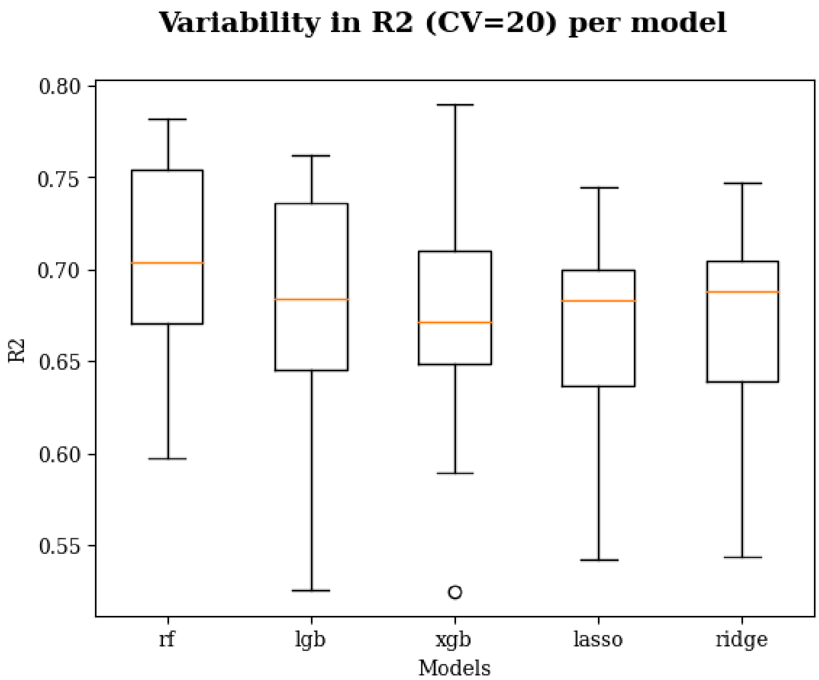


Figure S10 Out-of-sample performances evaluated using 20-fold repeated random sampling validation including LightGBM results – (global) - Figure S11 metric variability in RMSE

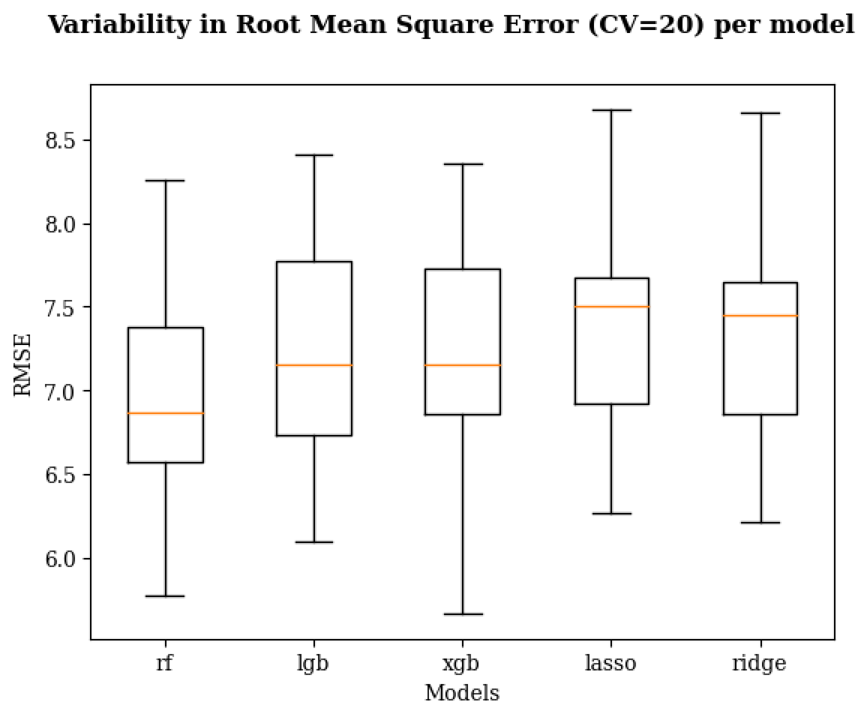


Figure S11 Out-of-sample performances evaluated using 20-fold repeated random sampling validation including LightGBM results – (global) - Figure S12 metric variability in MAE

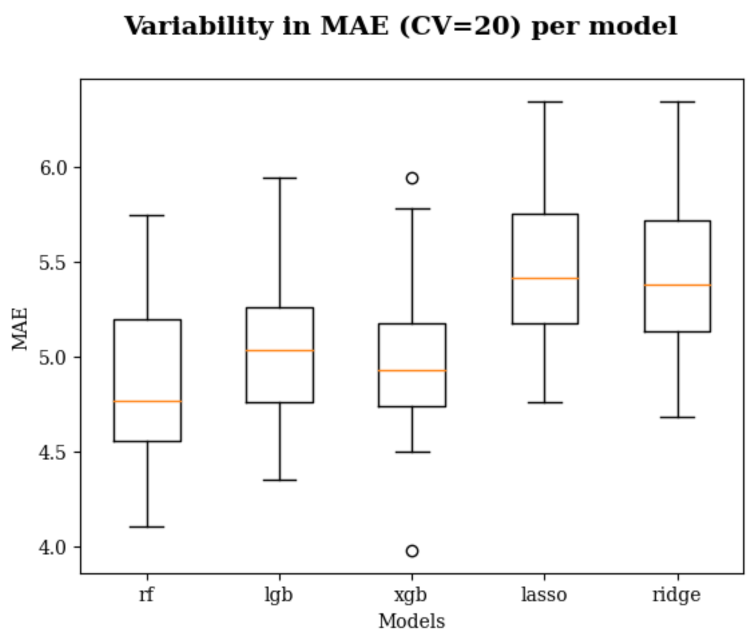


Figure S12 Spatial patterns of predicted NO2 (100m), measured in µg/m3, LightGBM for Amsterdam area

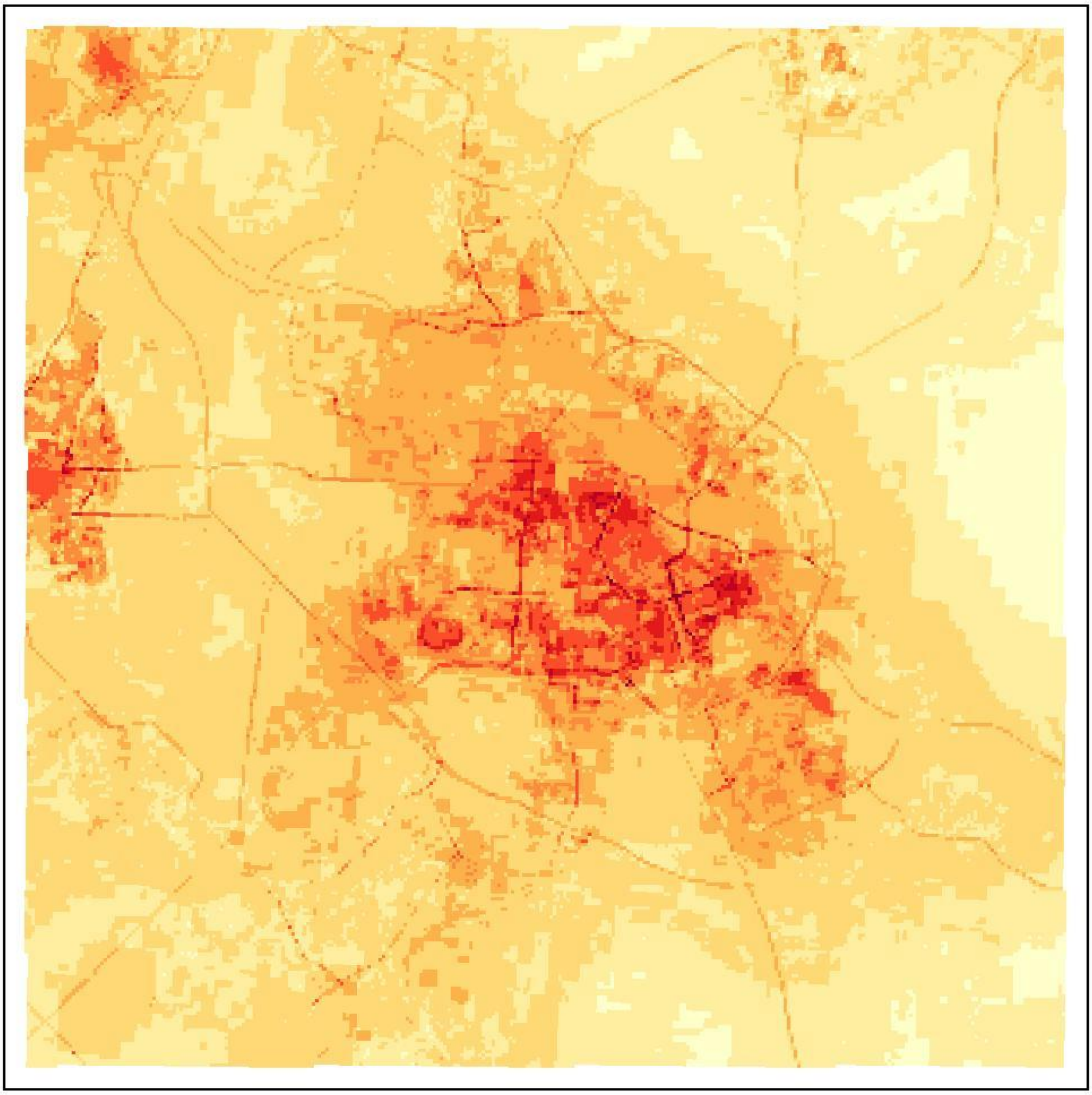


Figure S13 mean Shapley ranking against median Shapley ranking (global)

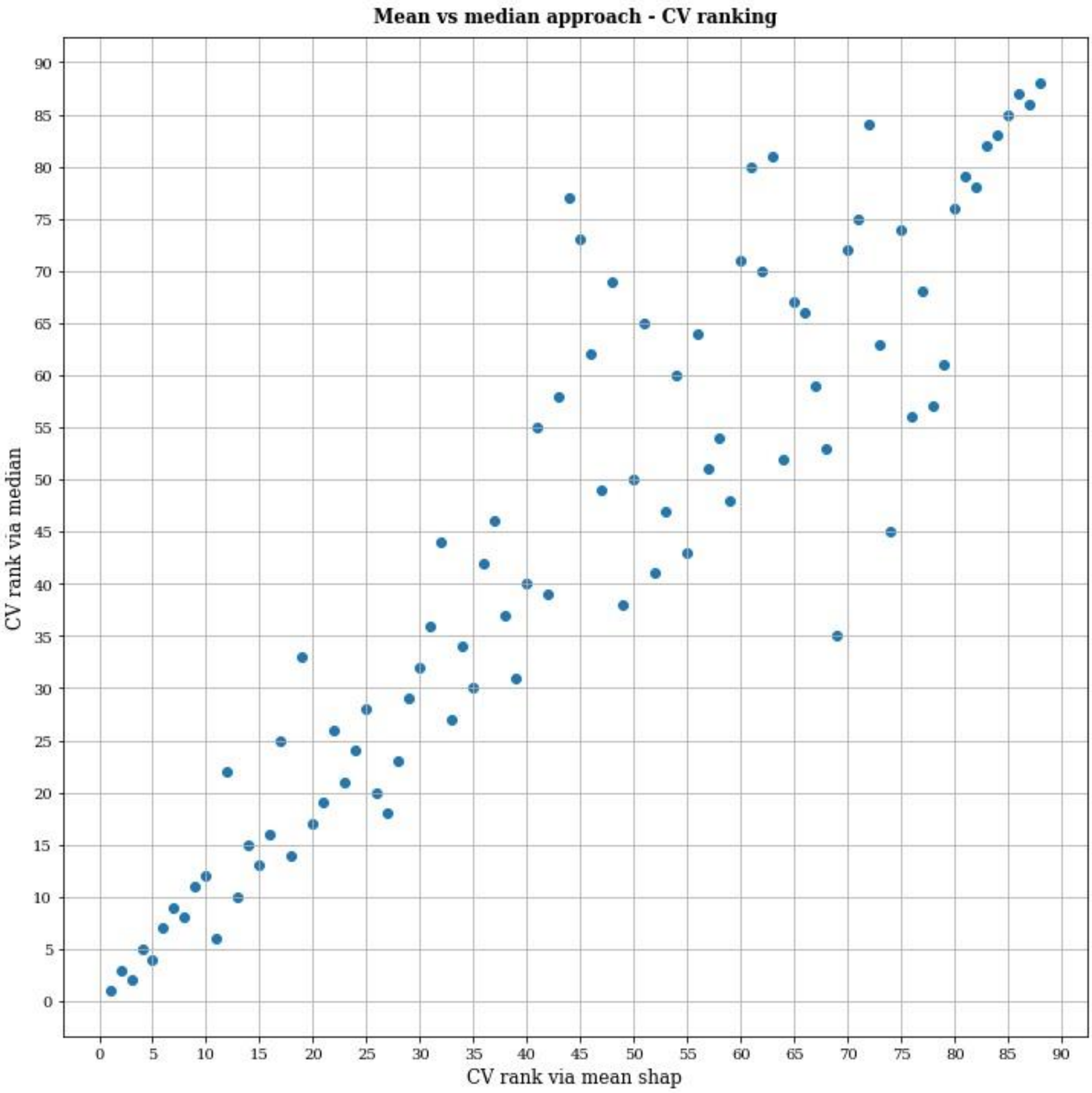
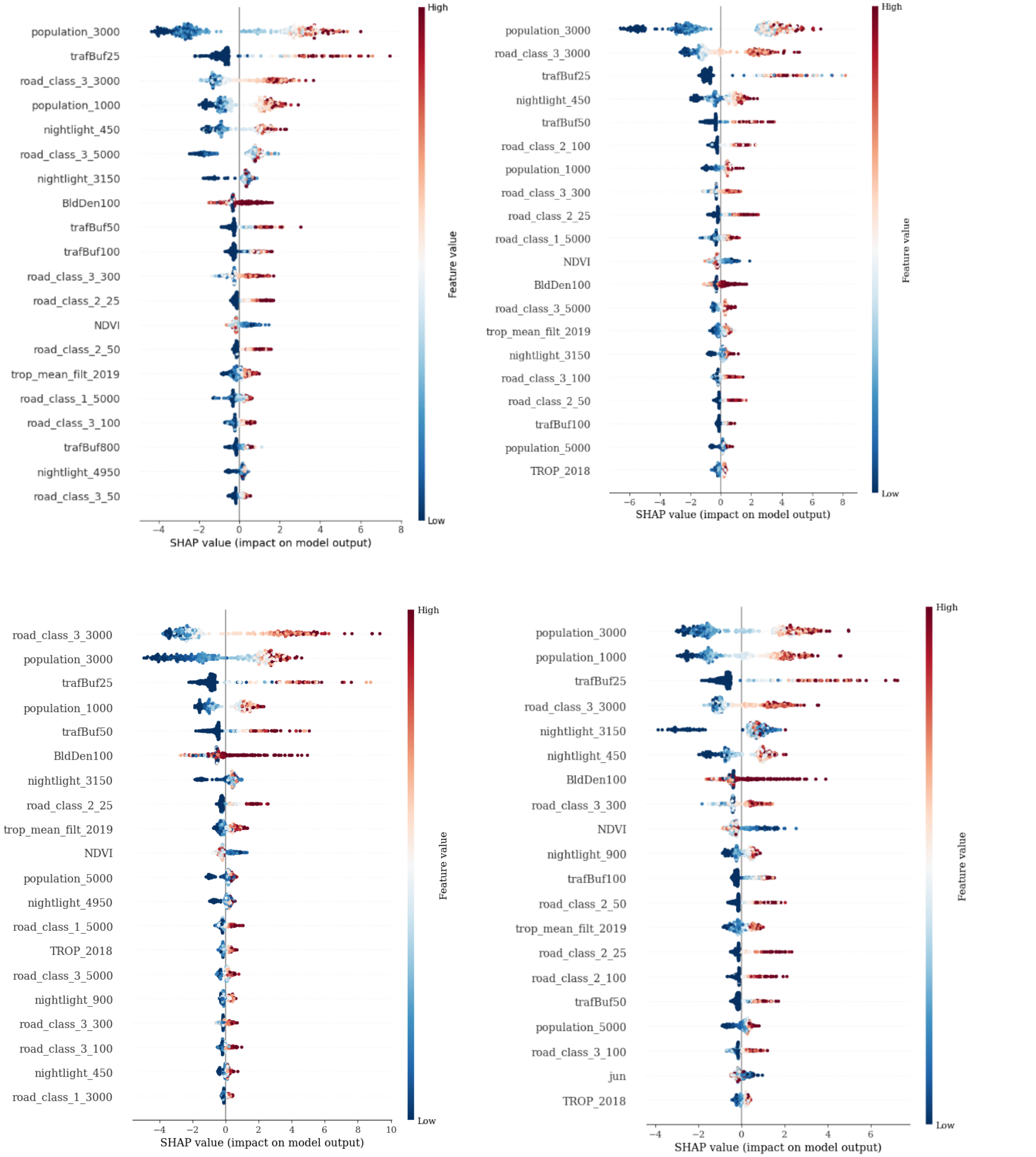
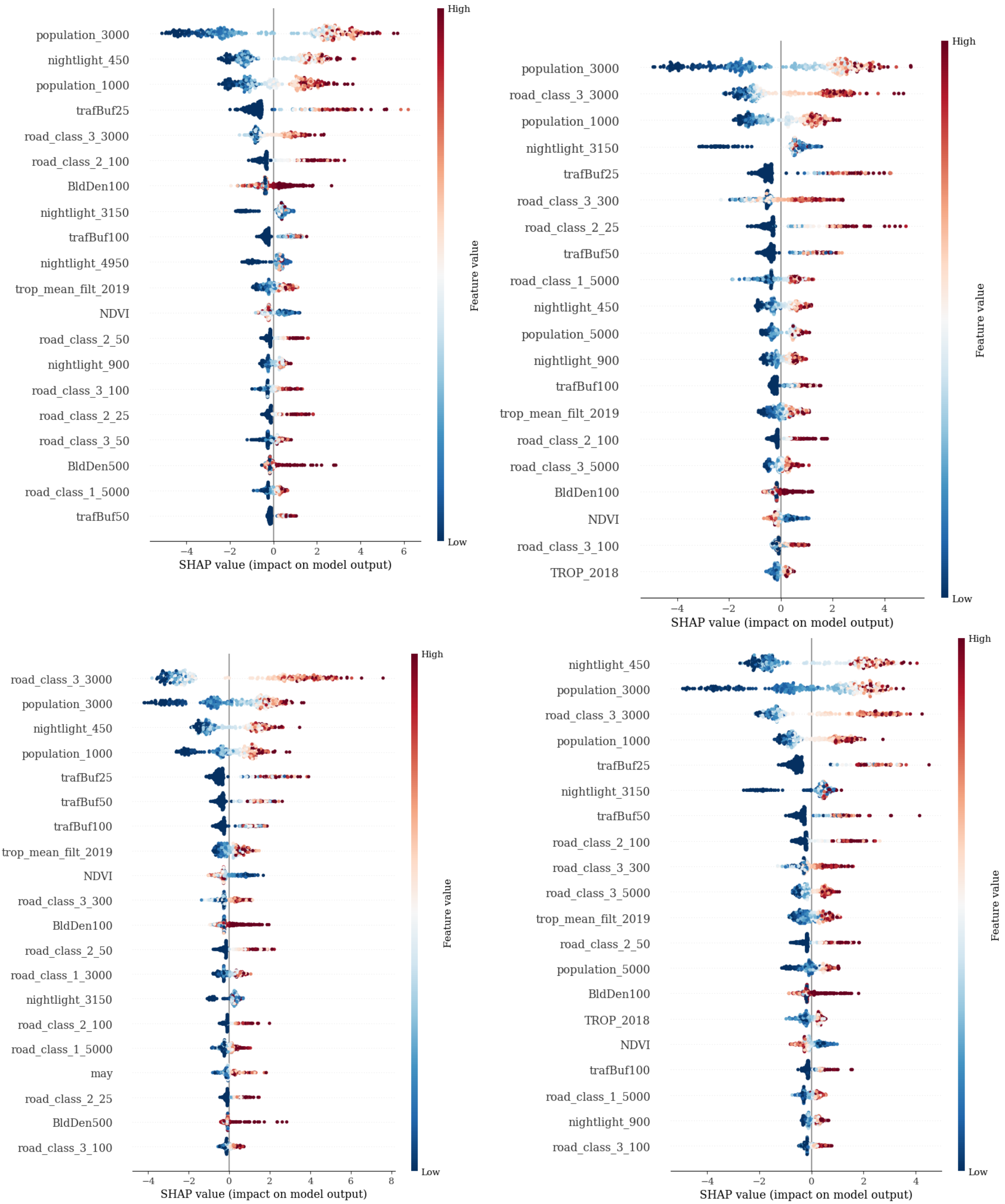


Figure S14 – Shapley seismic (global)





population_3000
nightlight_450
population_1000
trafBuf25
road_class_3_3000
road_class_2_100
BldDen100
nightlight_3150
trafBuf100
nightlight_4950
trop_mean_filt_2019
NDVI
road_class_2_50
nightlight_900
road_class_3_100
road_class_2_25
road_class_3_50
BldDen500
road_class_1_5000
trafBuf50
−4 −2 0 2 4 6
SHAP value (impact on model output)
High
Feature value
Low
population_3000
road_class_3_3000
population_1000
nightlight_3150
trafBuf25
road_class_3_300
road_class_2_25
trafBuf50
road_class_1_5000
nightlight_450
population_5000
nightlight_900
trafBuf100
trop_mean_filt_2019
road_class_2_100
road_class_3_5000
BldDen100
NDVI
road_class_3_100
TROP_2018
−4 −2 0 2 4
SHAP value (impact on model output)
High
Feature value
Low
road_class_3_3000
population_3000
nightlight_450
population_1000
trafBuf25
trafBuf50
trafBuf100
trop_mean_filt_2019
NDVI
road_class_3_300
BldDen100
road_class_2_50
road_class_1_3000
nightlight_3150
road_class_2_100
road_class_1_5000
may
road_class_2_25
BldDen500
road_class_3_100
−4 −2 0 2 4 6 8
SHAP value (impact on model output)
High
Feature value
Low
nightlight_450
population_3000
road_class_3_3000
population_1000
trafBuf25
nightlight_3150
trafBuf50
road_class_2_100
road_class_3_300
road_class_3_5000
trop_mean_filt_2019
road_class_2_50
population_5000
BldDen100
TROP_2018
NDVI
trafBuf100
road_class_1_5000
nightlight_900
road_class_3_100
−4 −2 0 2 4
SHAP value (impact on model output)
High
Feature value
Low

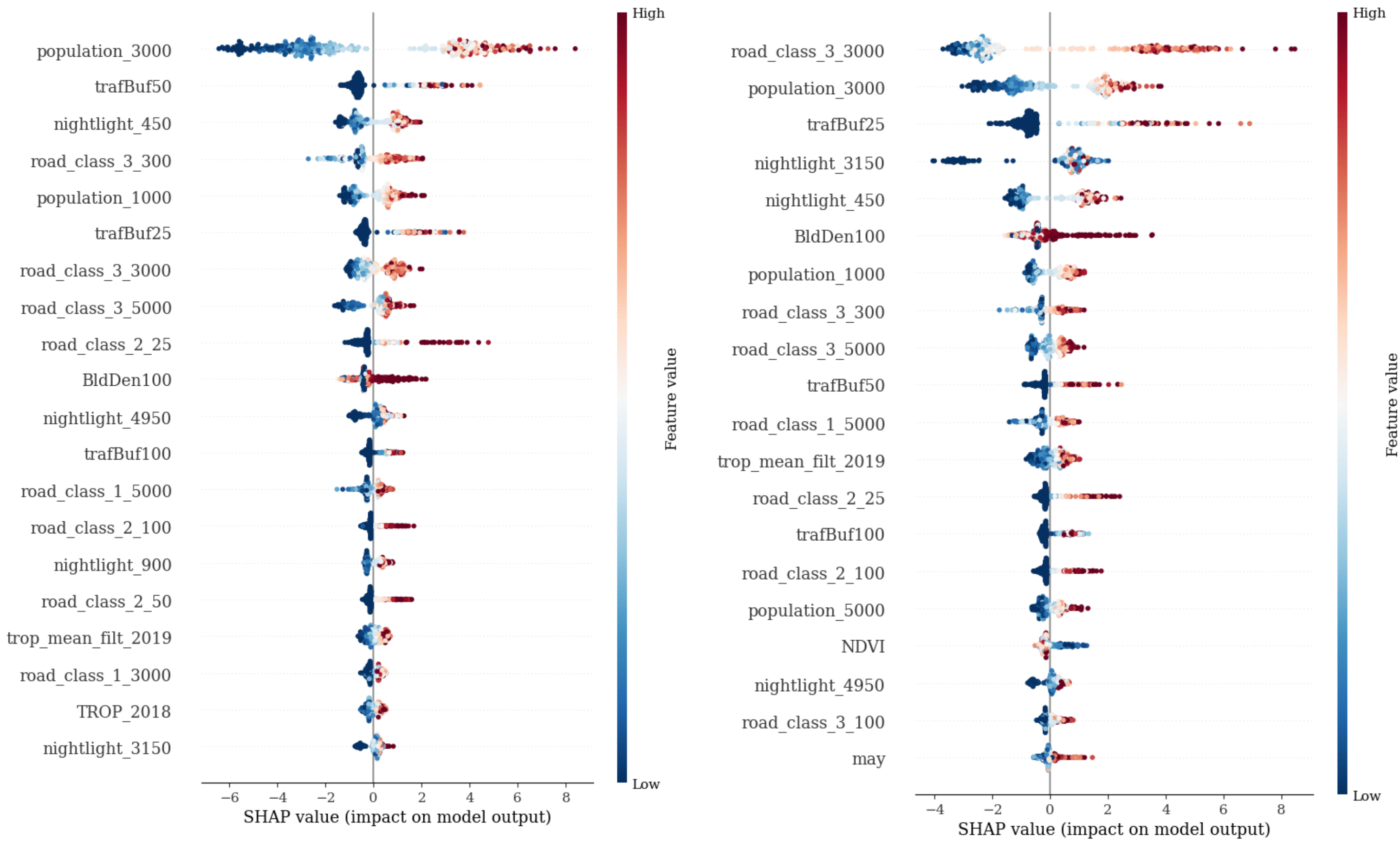


Figure S15 – R2 – Out-of-sample performances evaluated using 20-fold repeated random sampling validation – (local)

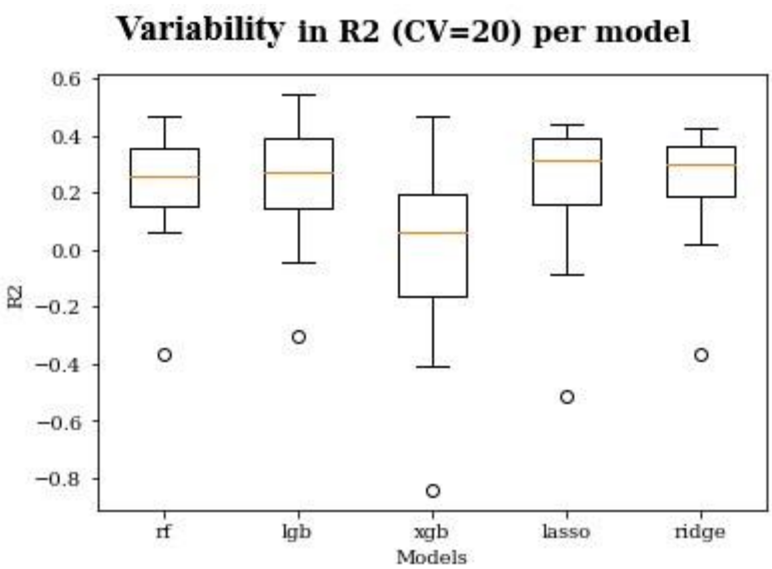


Figure S16 – RMSE – Out-of-sample performances evaluated using 20-fold repeated random sampling validation – (local)

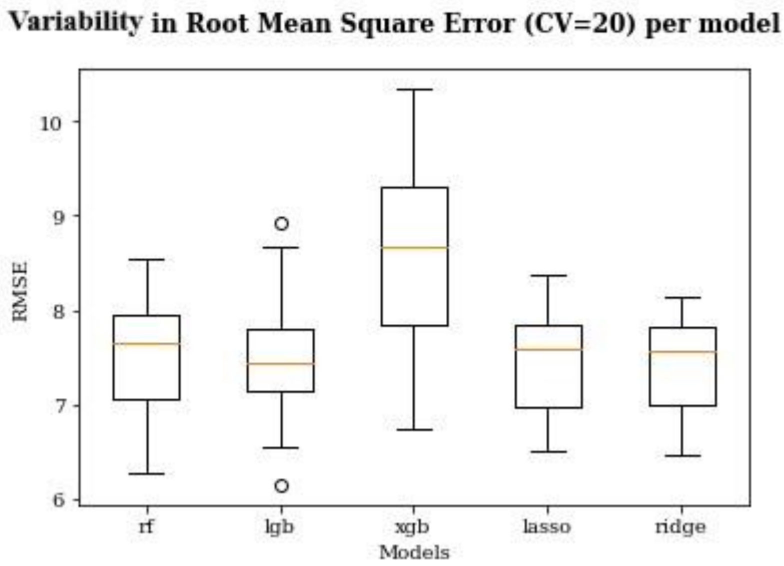


Figure S17 – MAE – Out-of-sample performances evaluated using 20-fold repeated random sampling validation – (local)

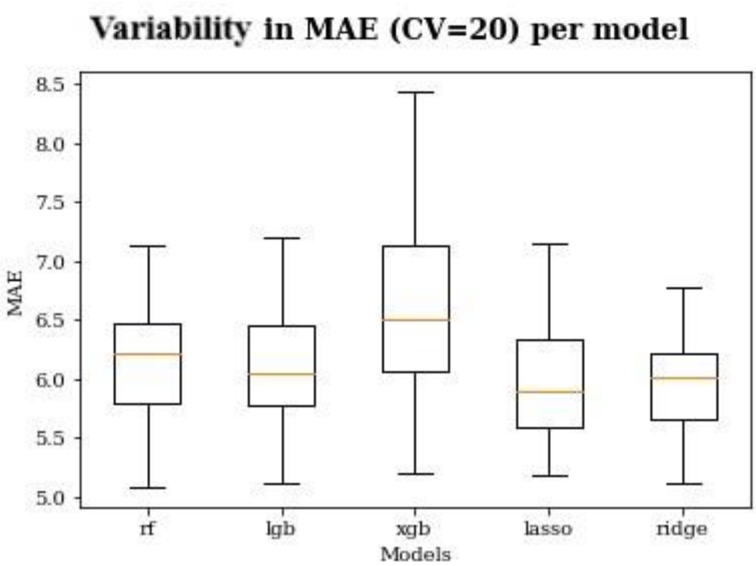


Figure S18 Spatial references Amsterdam area

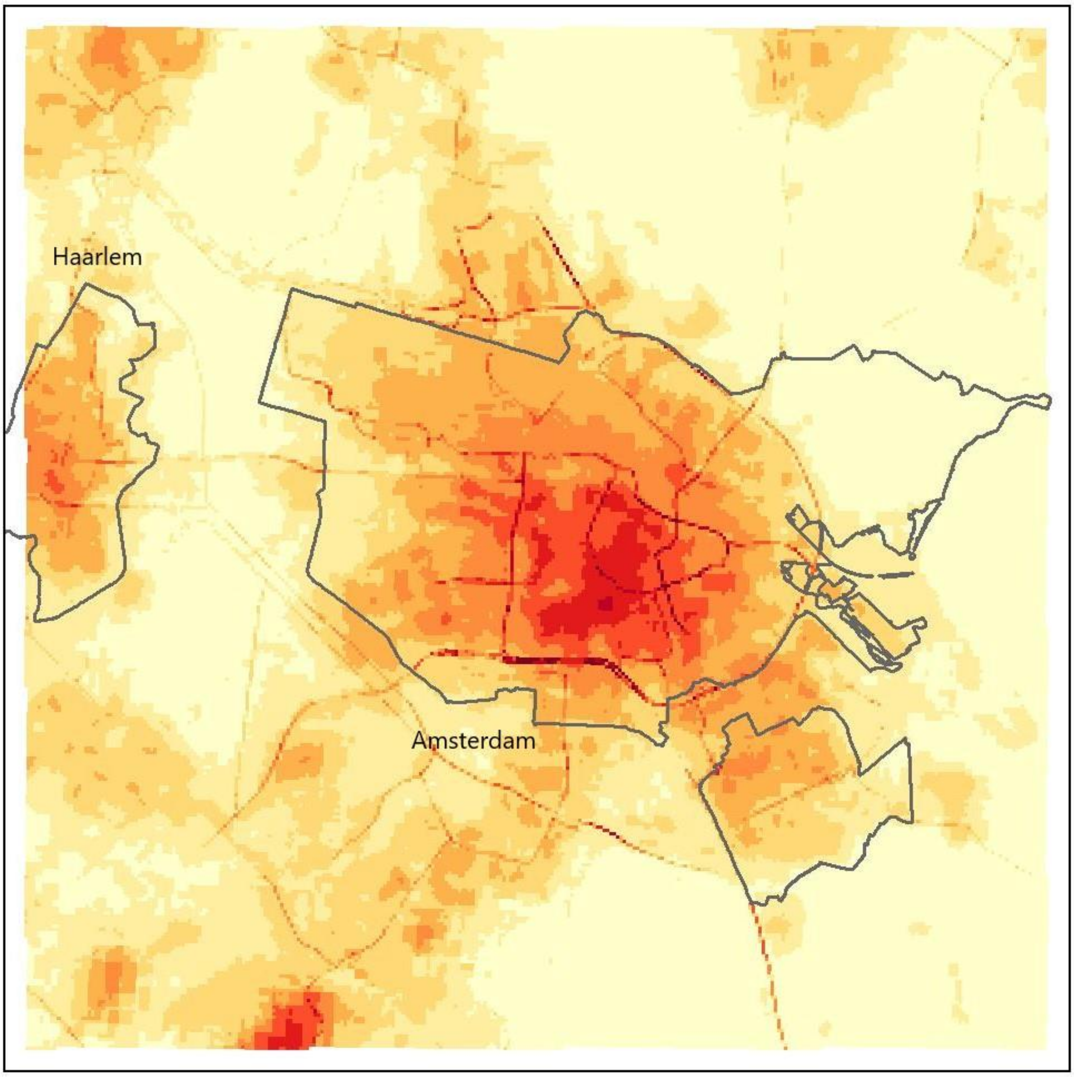


Figure S19 Hamburg predicted NO2 LASSO (colorblindfriendly)

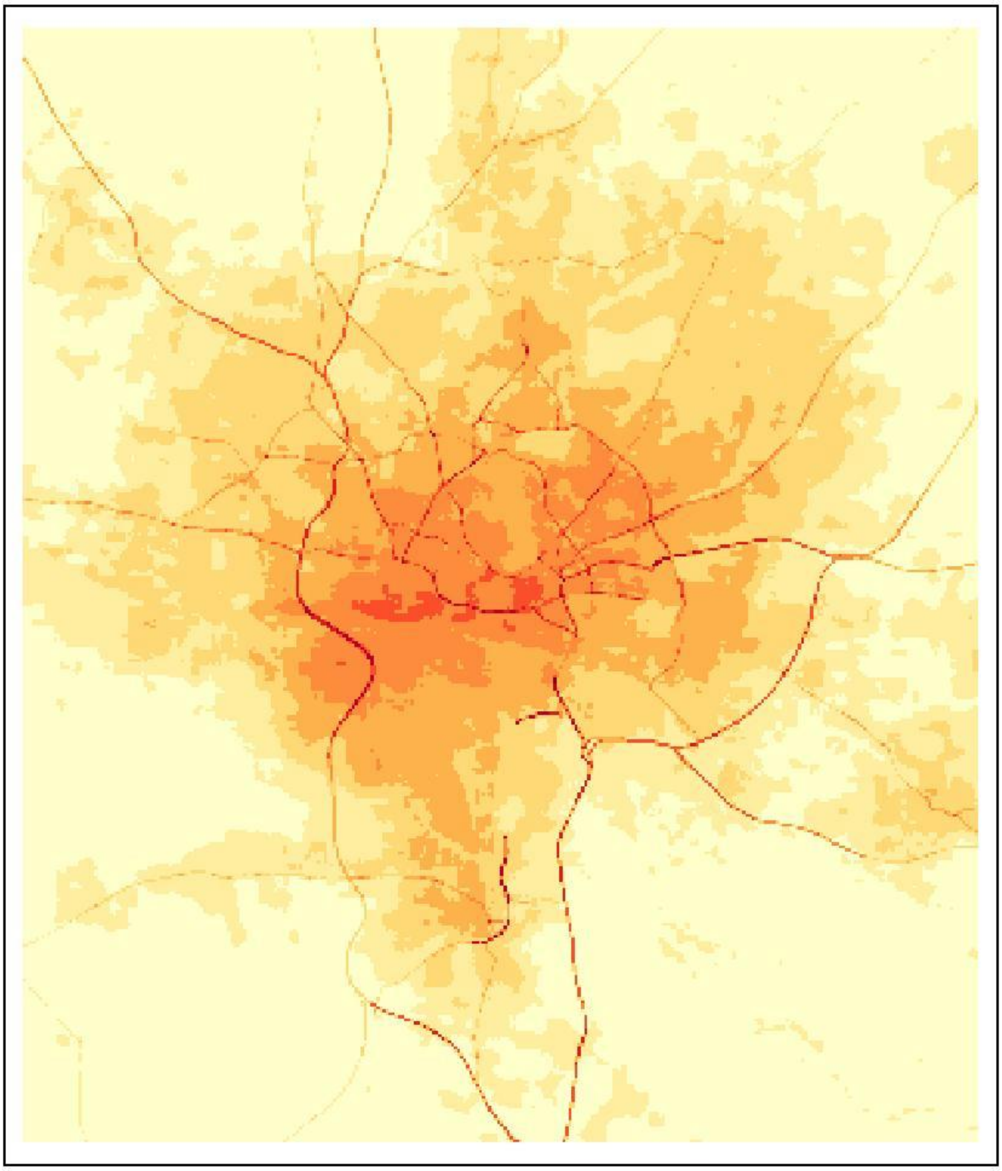


Figure S20 Hamburg predicted NO2 LightGBM (colorblindfriendly)

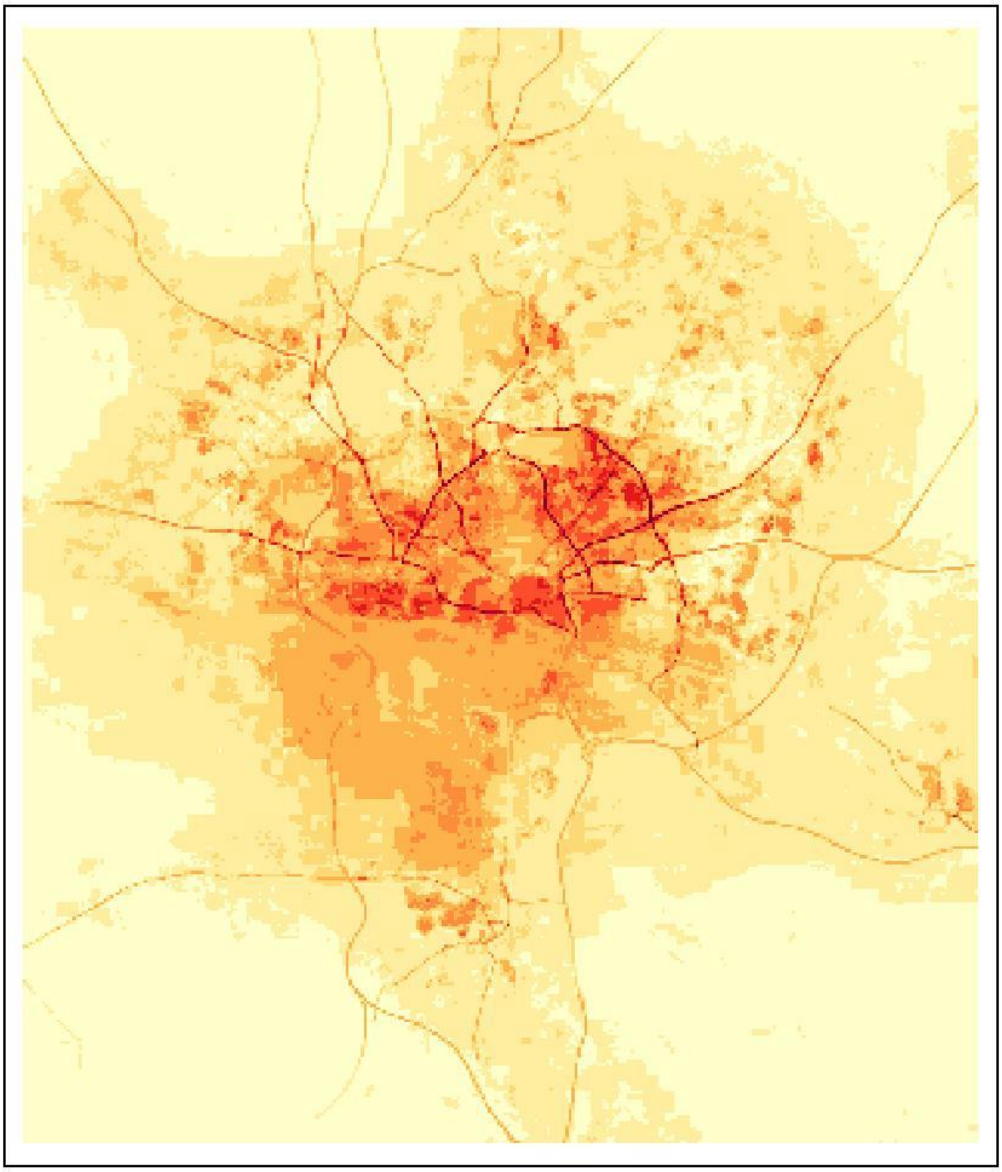


Figure S21 Hamburg predicted NO2 XGBoost (colorblindfriendly)

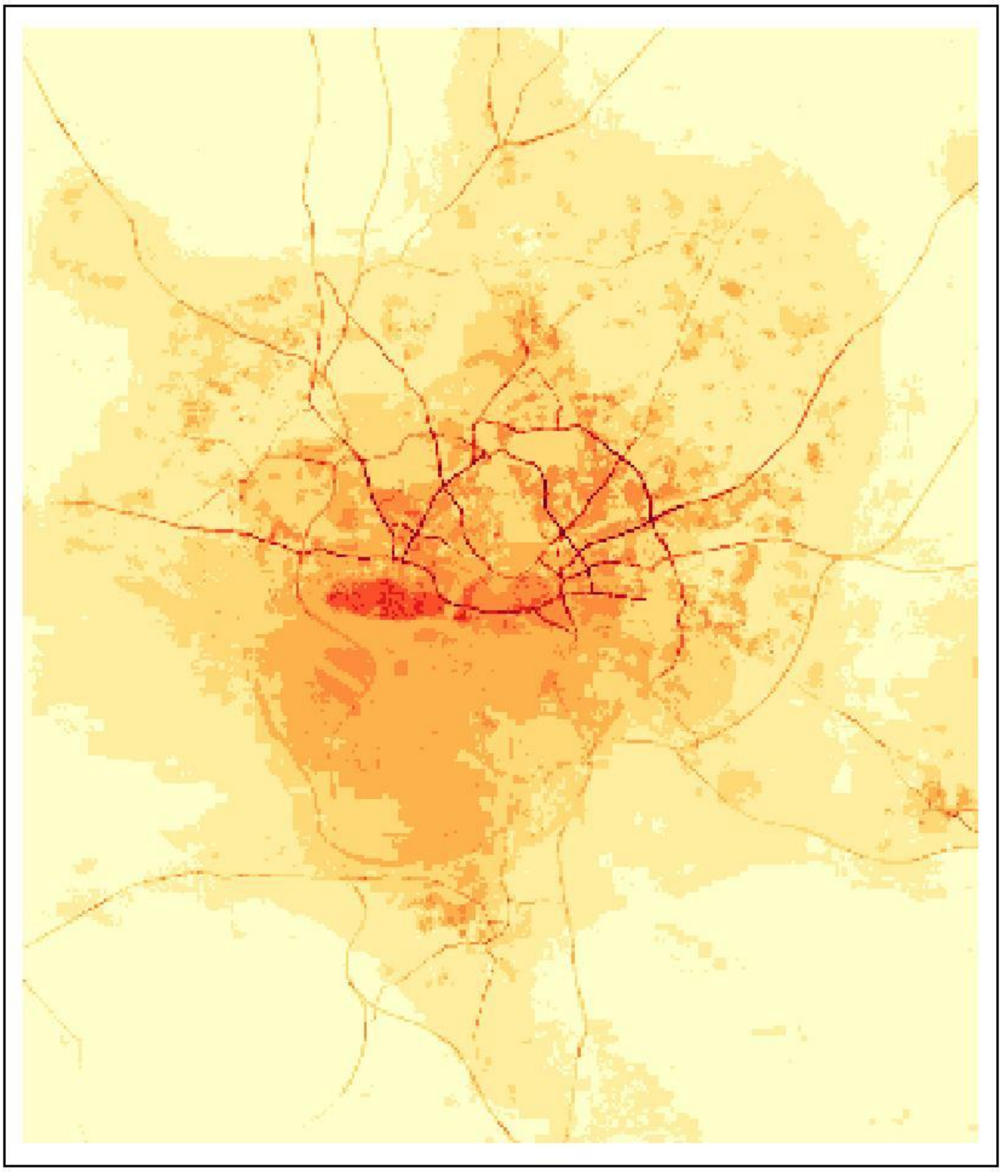


Figure S22 Utrecht predicted NO2 LASSO (colorblindfriendly)

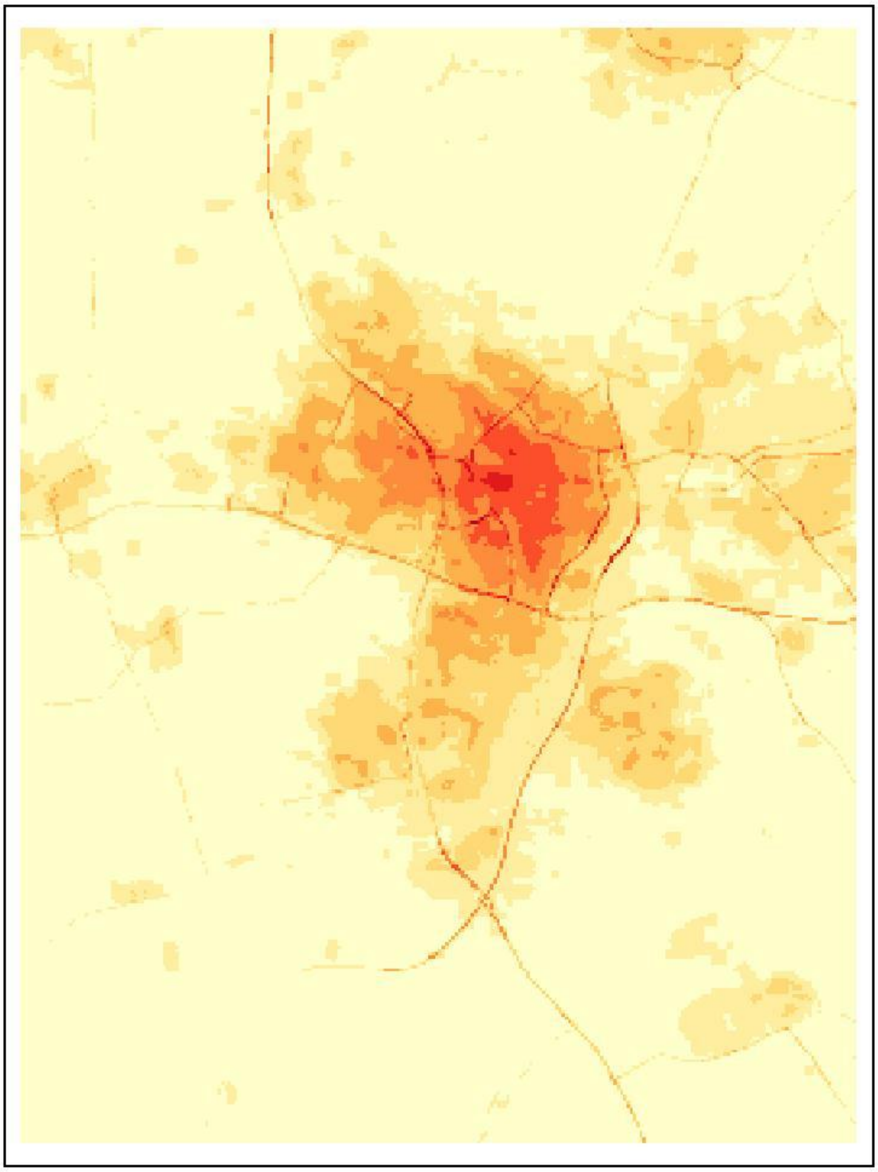


Figure S23 Utrecht predicted NO2 LightGBM (colorblindfriendly)

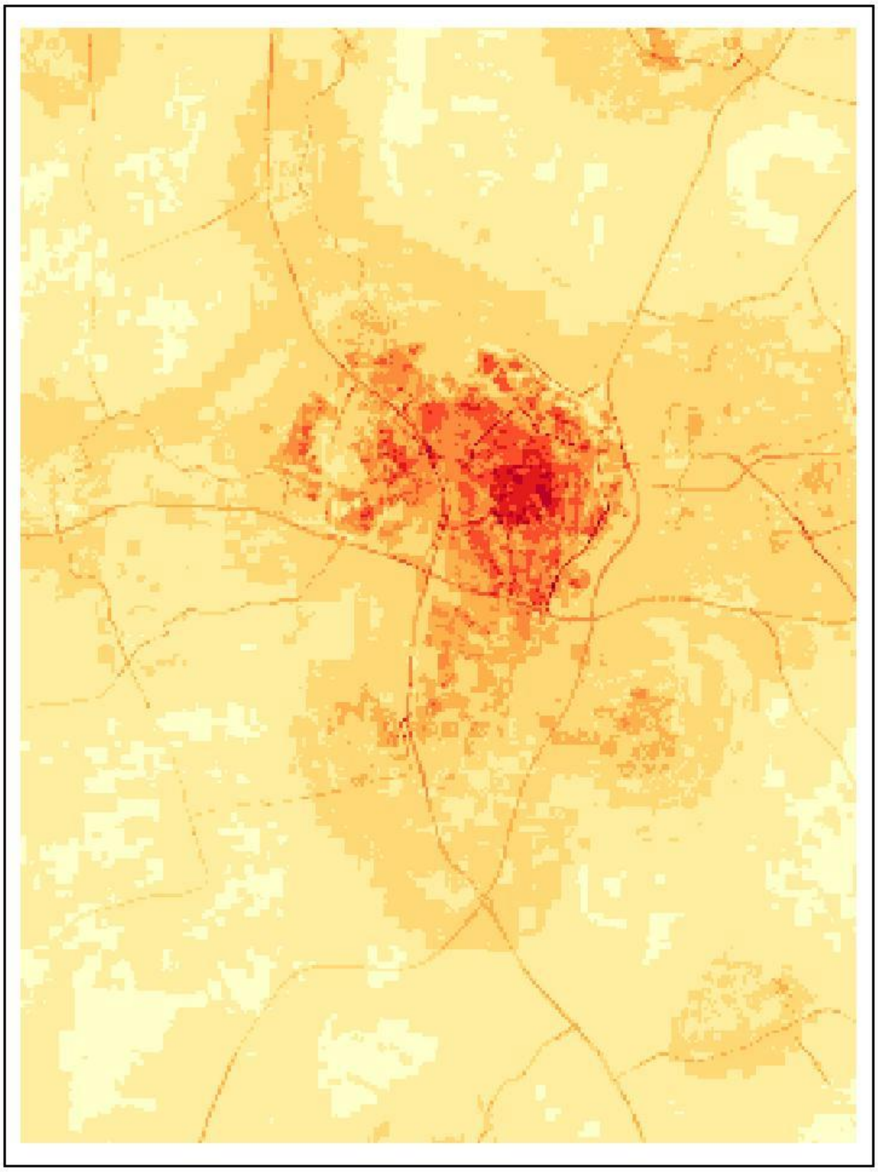


Figure S24 Utrecht predicted NO2 XGBoost (colorblindfriendly)

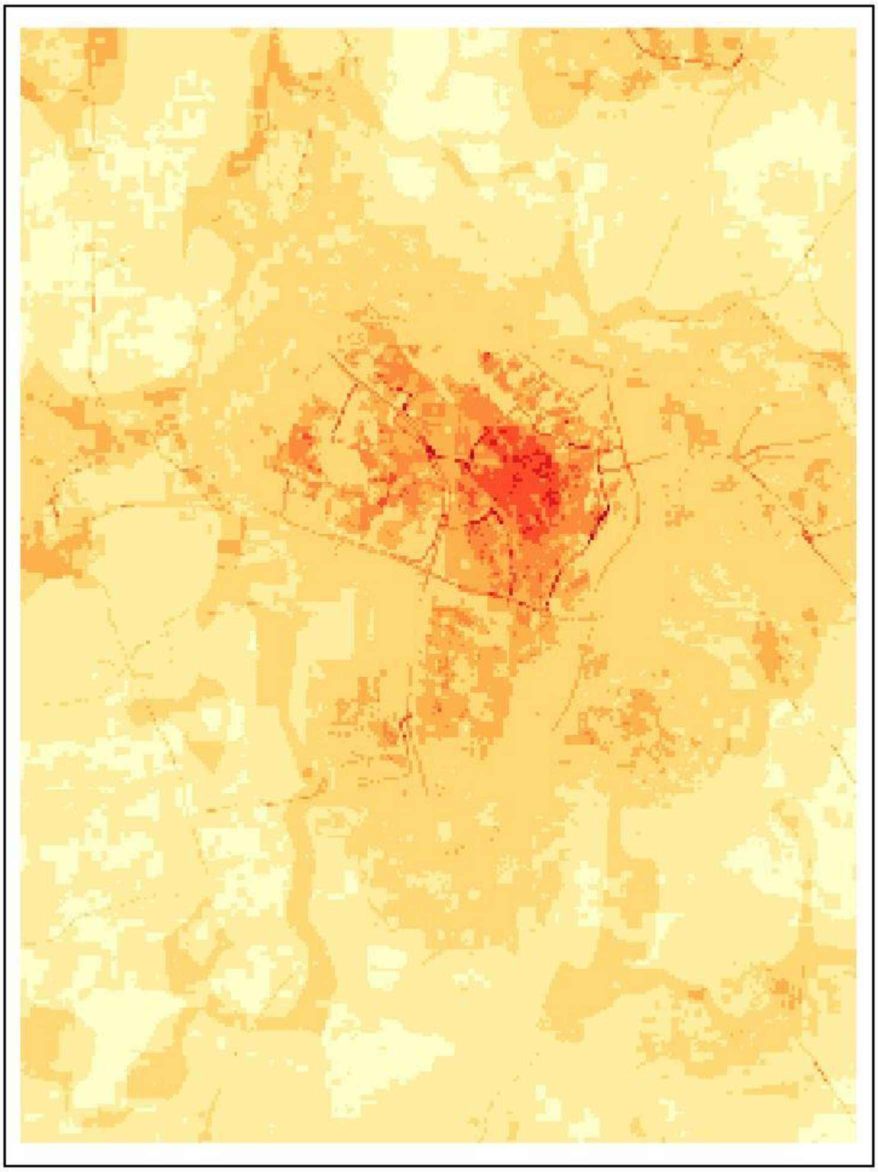


Figure S25 Bayreuth predicted NO2 LASSO (colorblindfriendly)

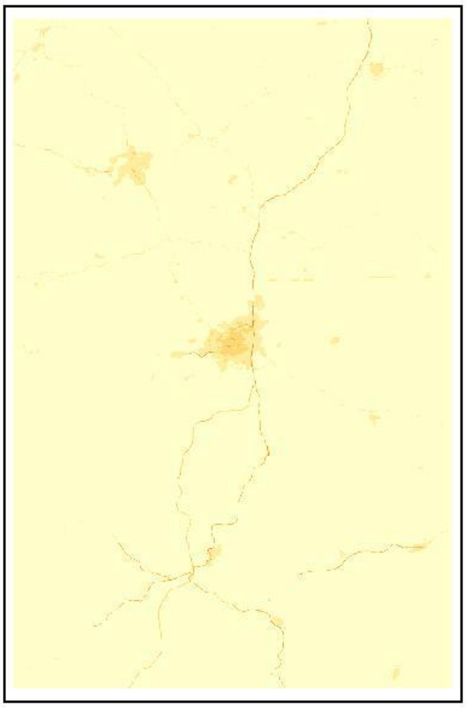


Figure S26 Bayreuth predicted NO2 LightGBM (colorblindfriendly)

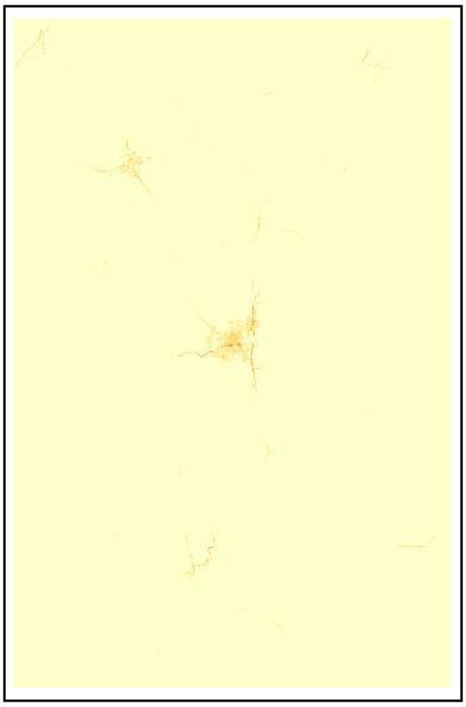


Figure S27 Bayreuth predicted NO2 XGBoost (colorblindfriendly)

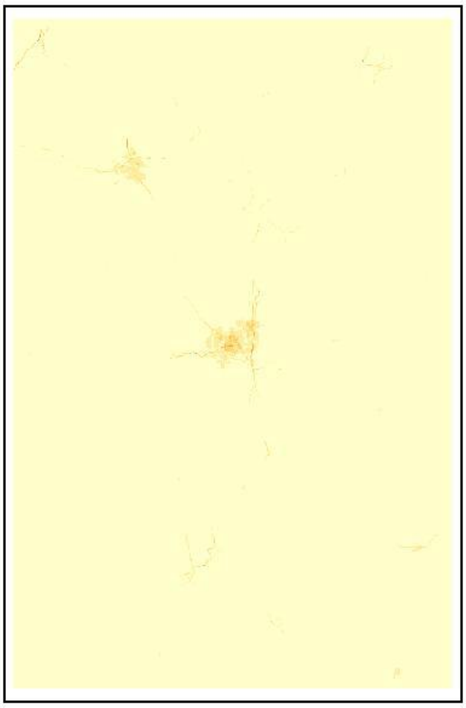


Figure S28 Bayreuth zoomed in predicted NO2 LASSO (colorblindfriendly)

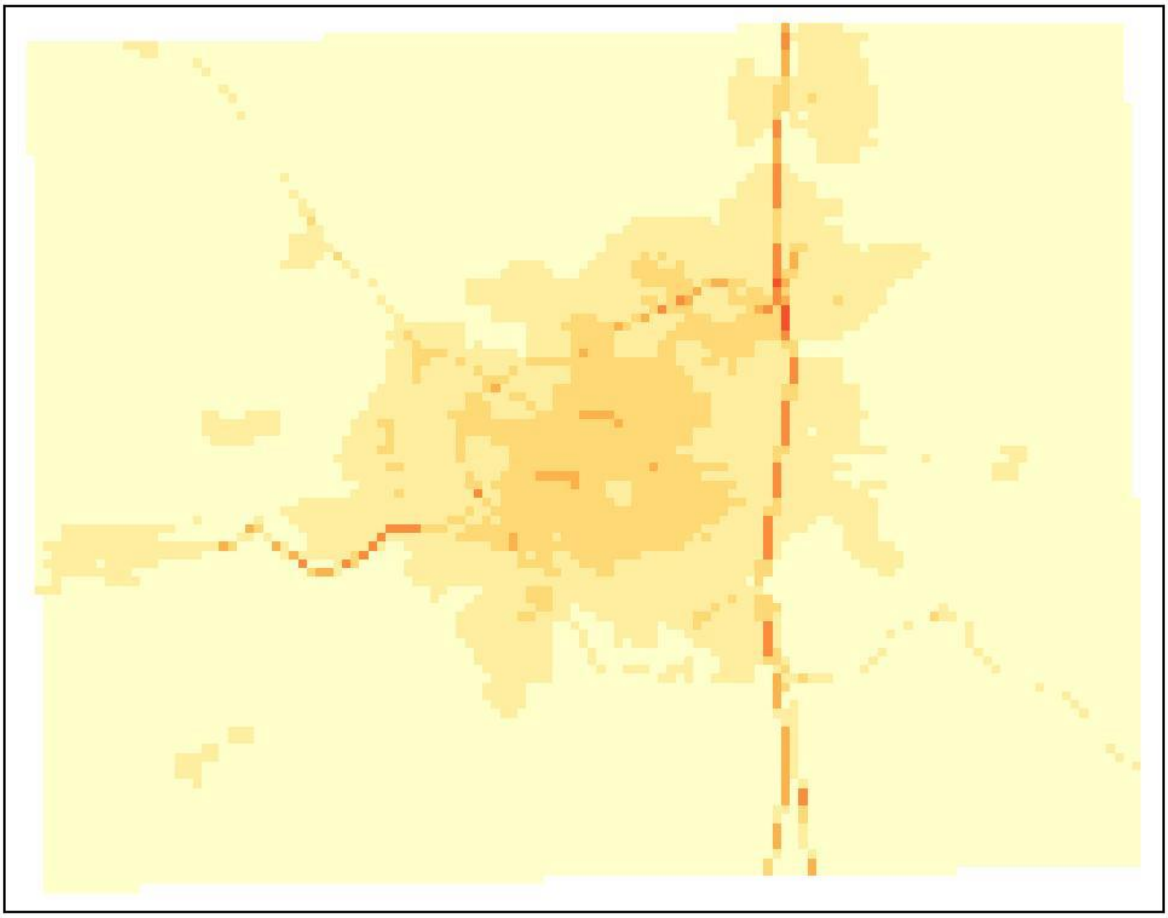


Figure S29 Bayreuth zoomed in predicted NO2 LightGBM

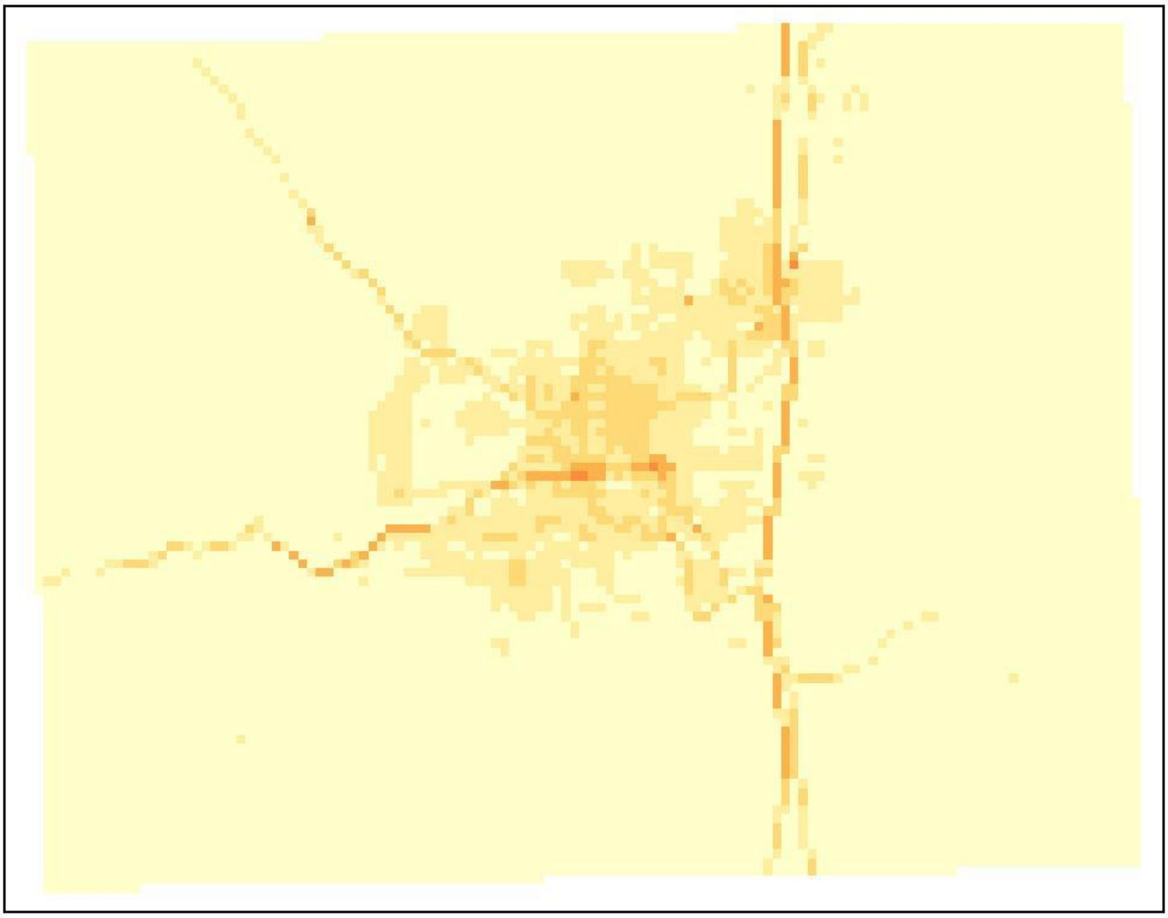


Figure S30 Bayreuth zoomed in predicted NO2 XGBoost (colorblindfriendly)

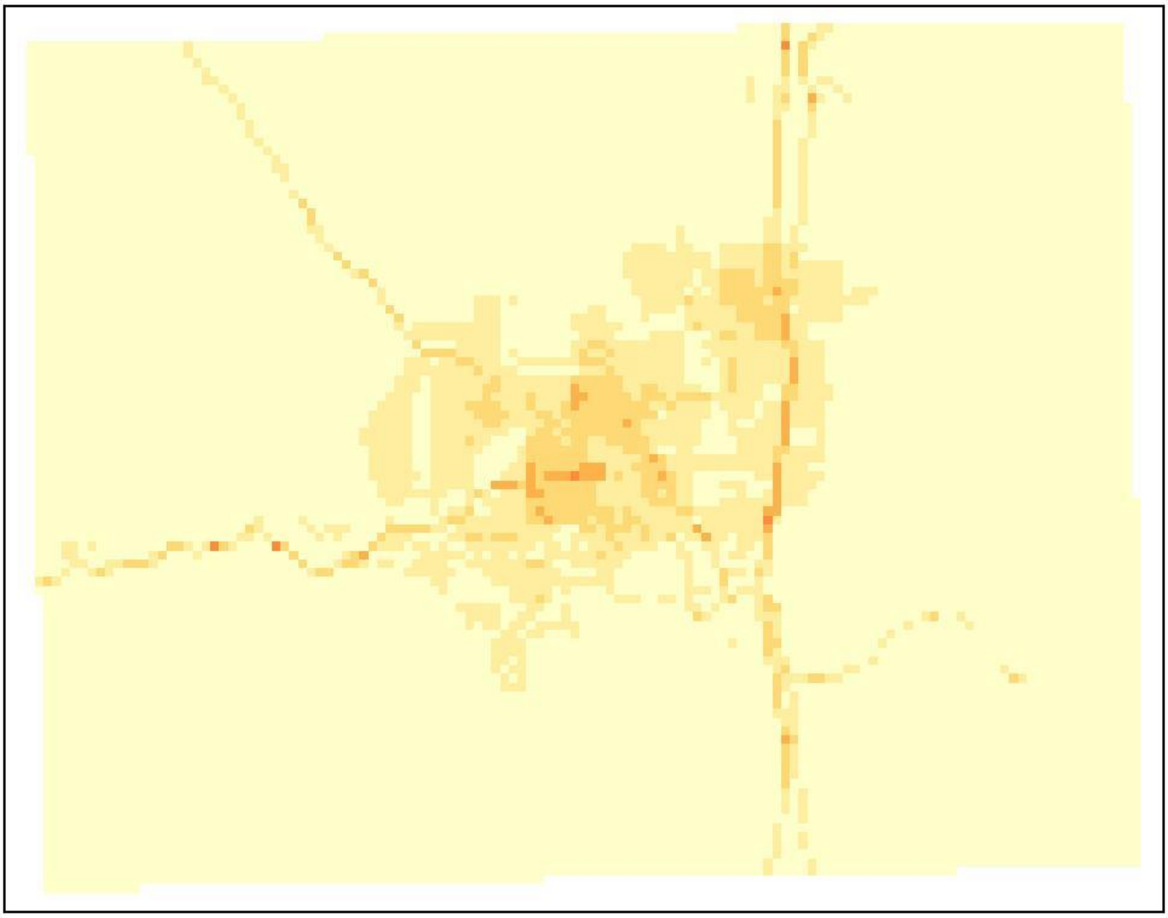


Figure S31 - Distribution predicted NO2 per global model per area of interest (global)

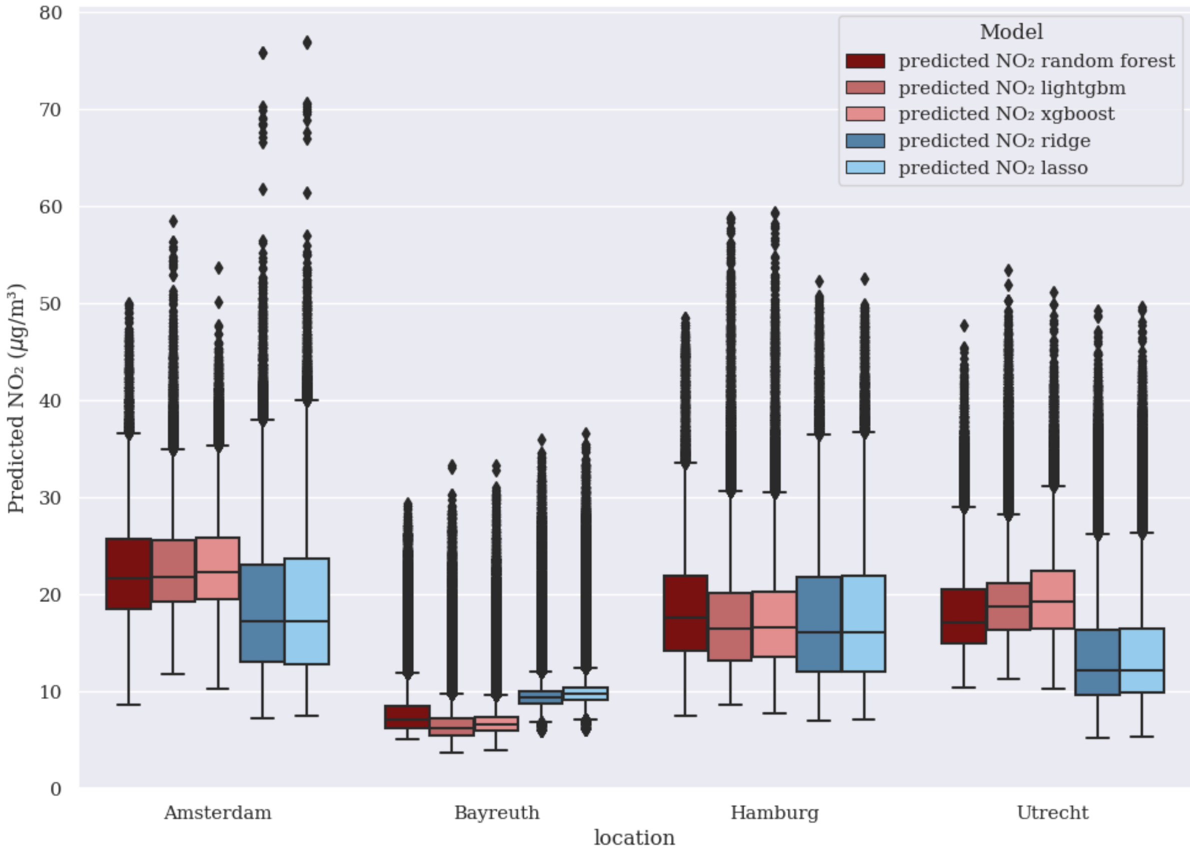


Figure S32 – Spatial distribution of local road class 2 5000 in Amsterdam Area

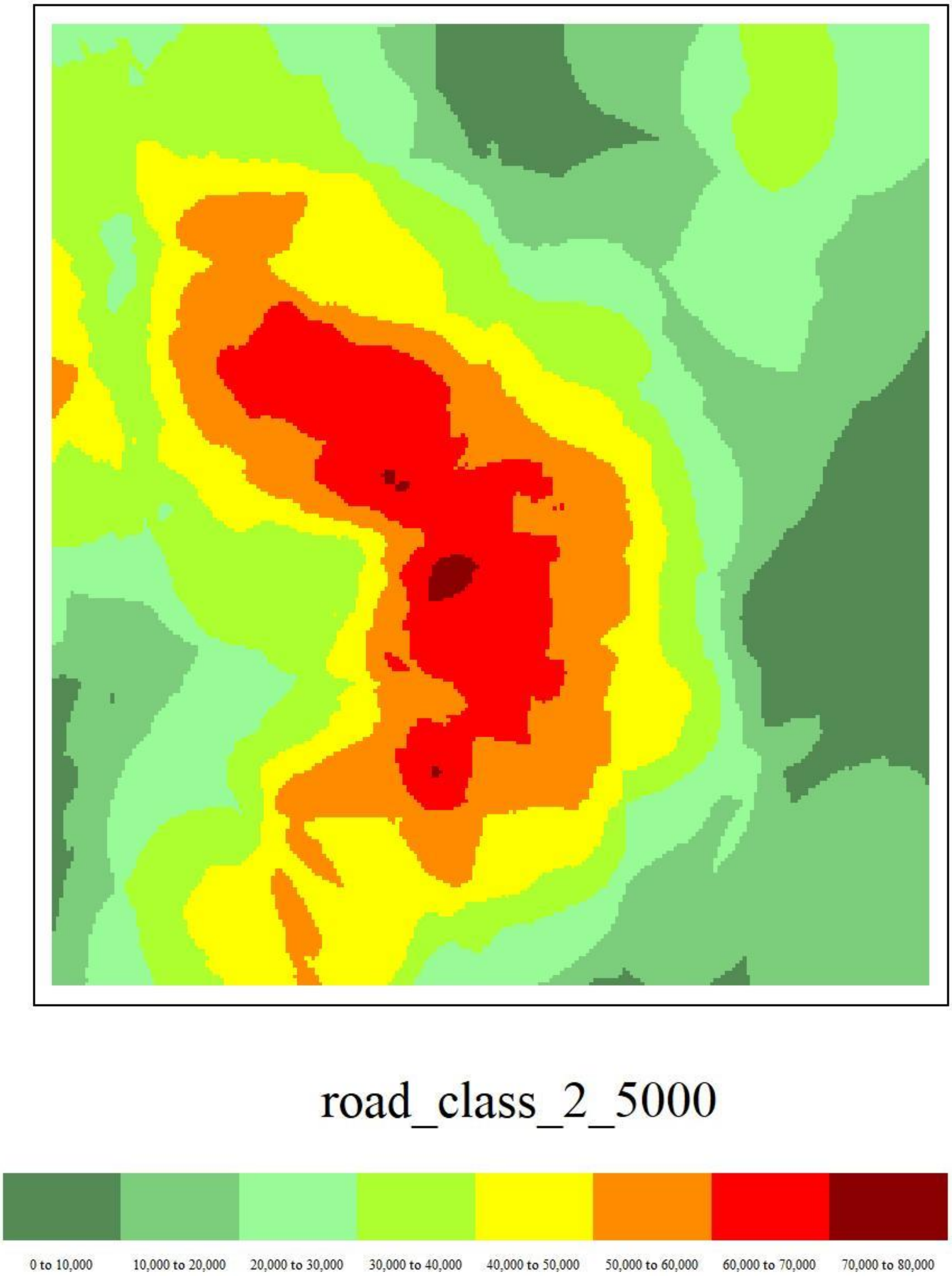


Figure S33 – local predictor nightlight 450

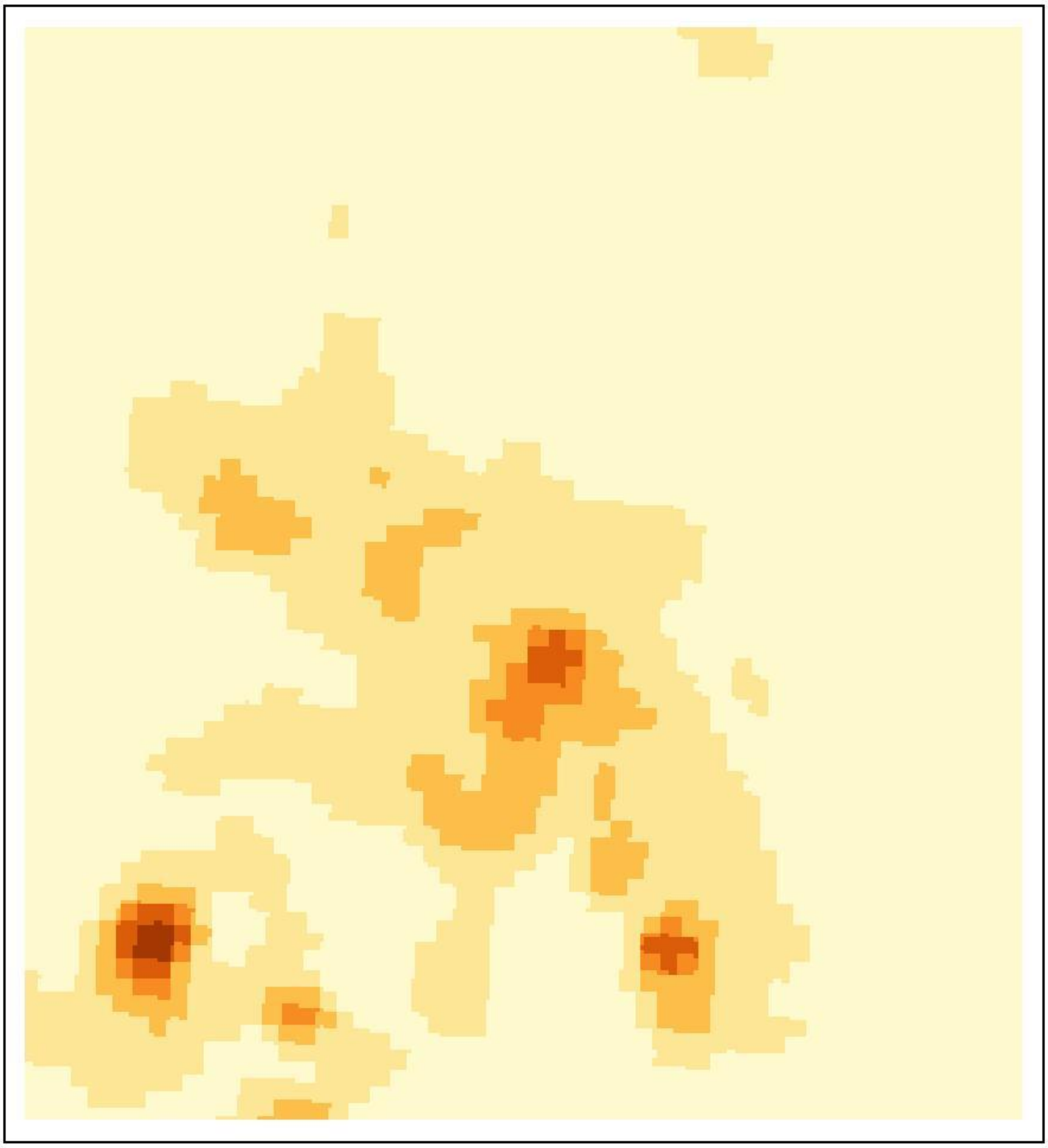


Figure S34 – local predictor nightlight 4950

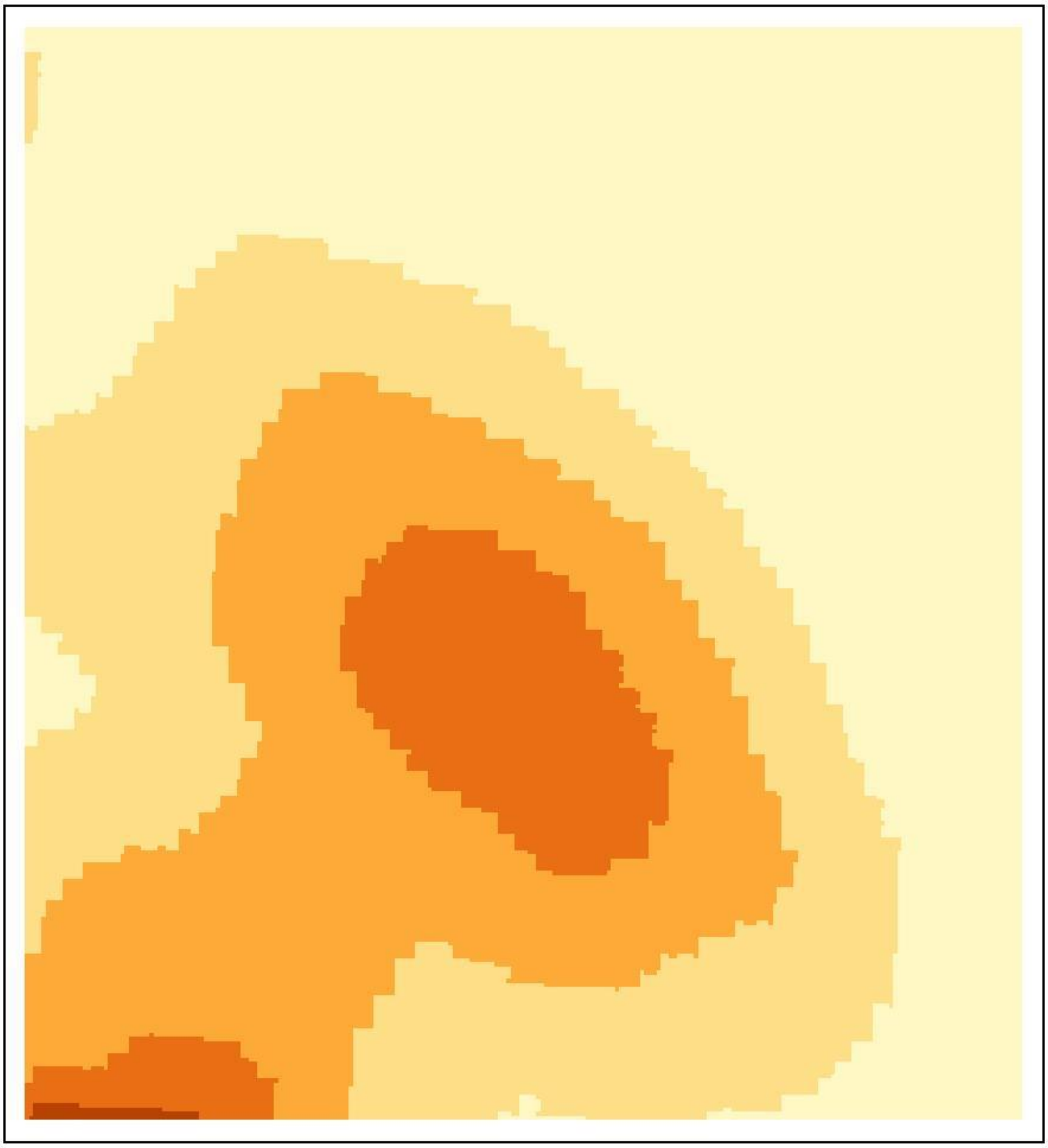


Figure S35 – local predictor population 3000

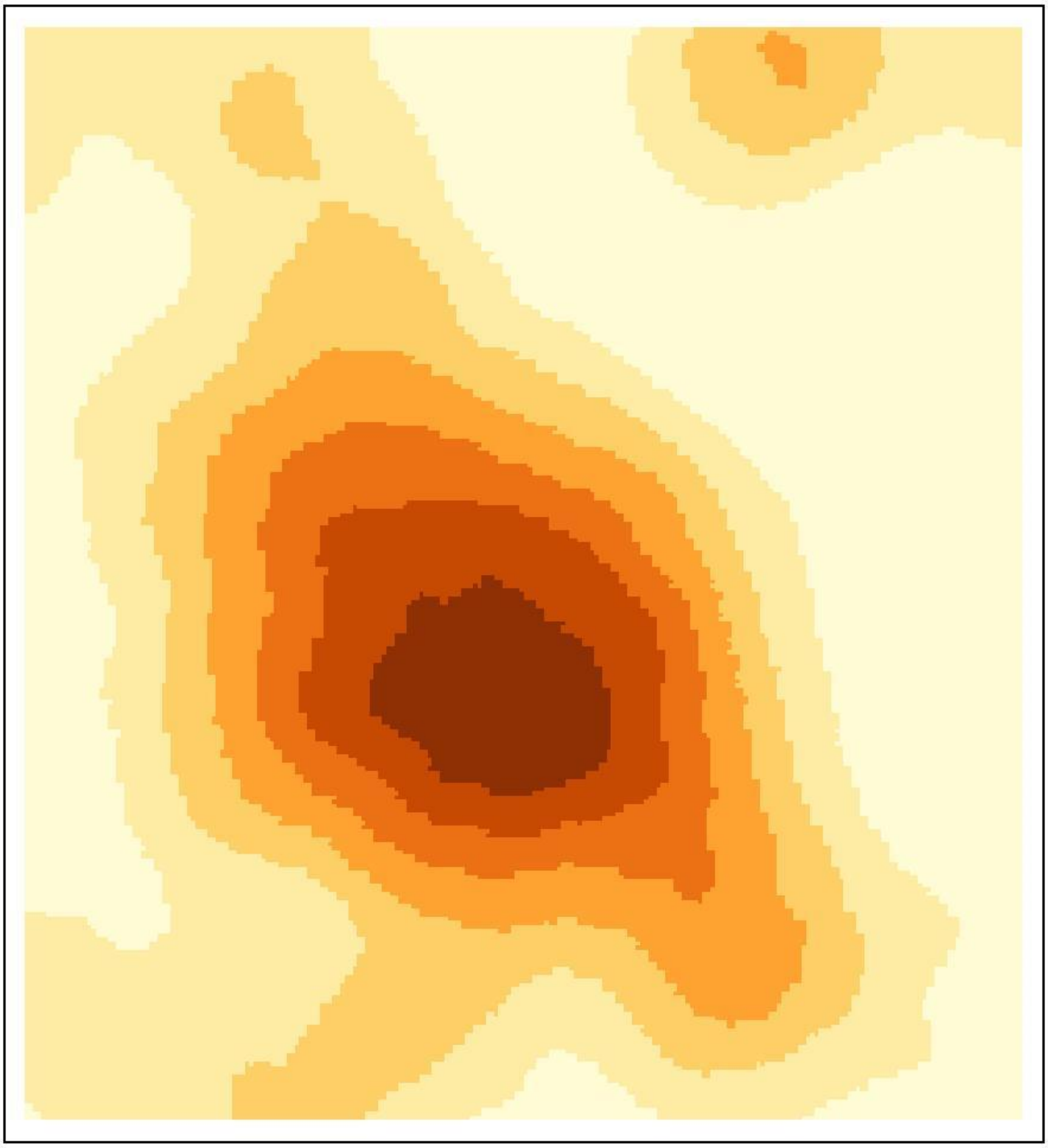


Figure S36 – local predictor road class 1 5000

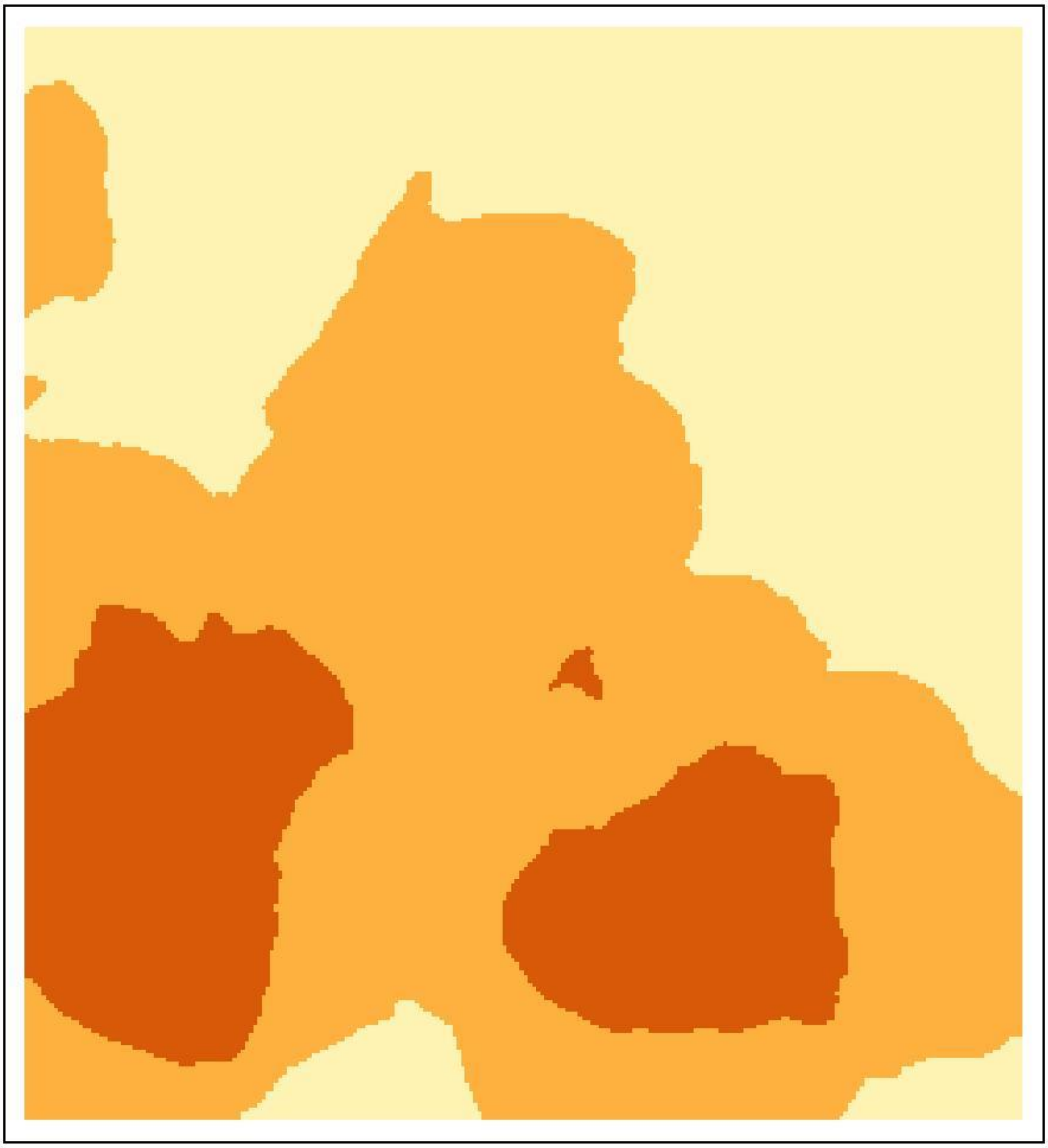


Figure S37 – local predictor road class 2 1000

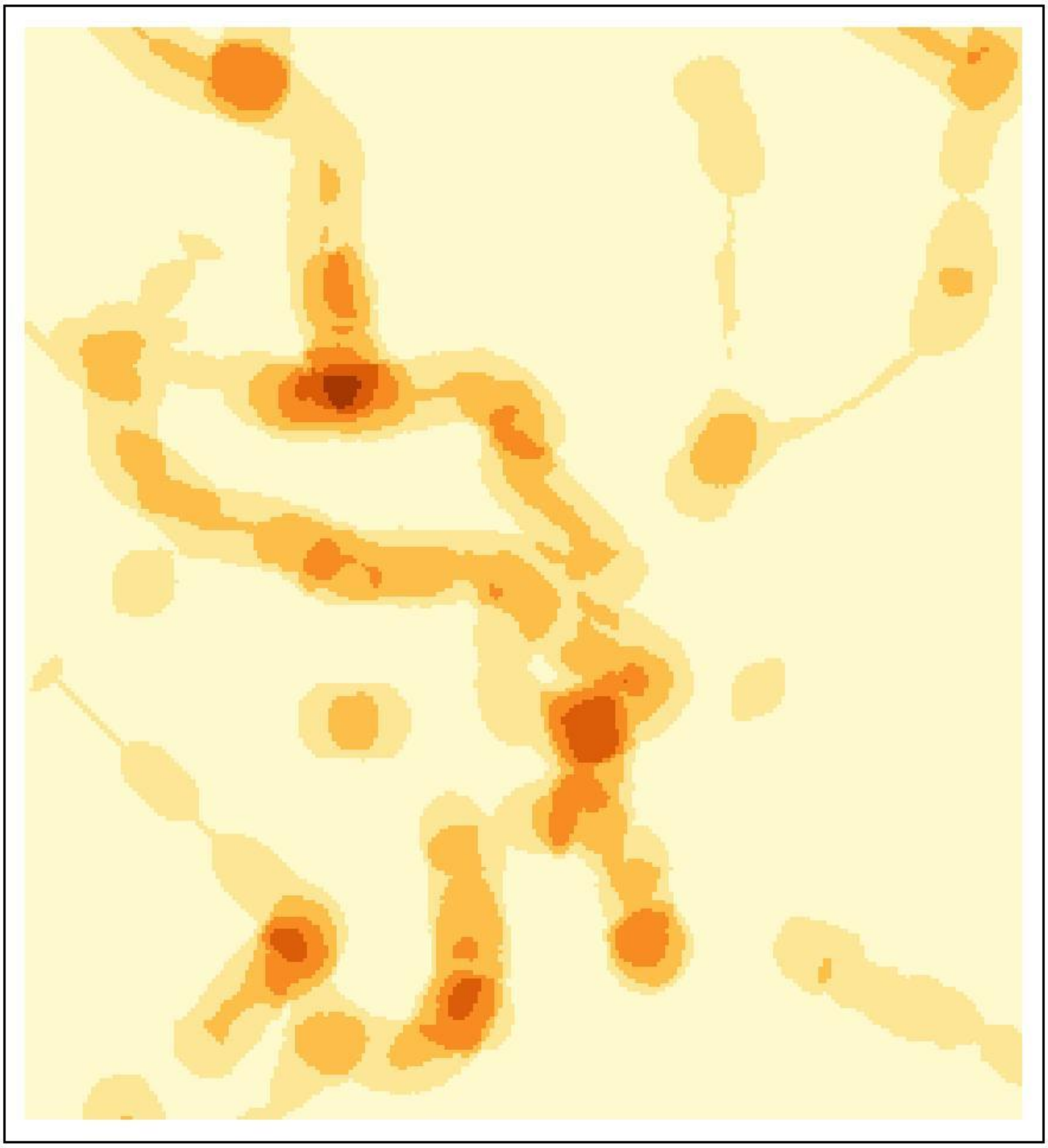


Figure S38 – local predictor road class 2 5000

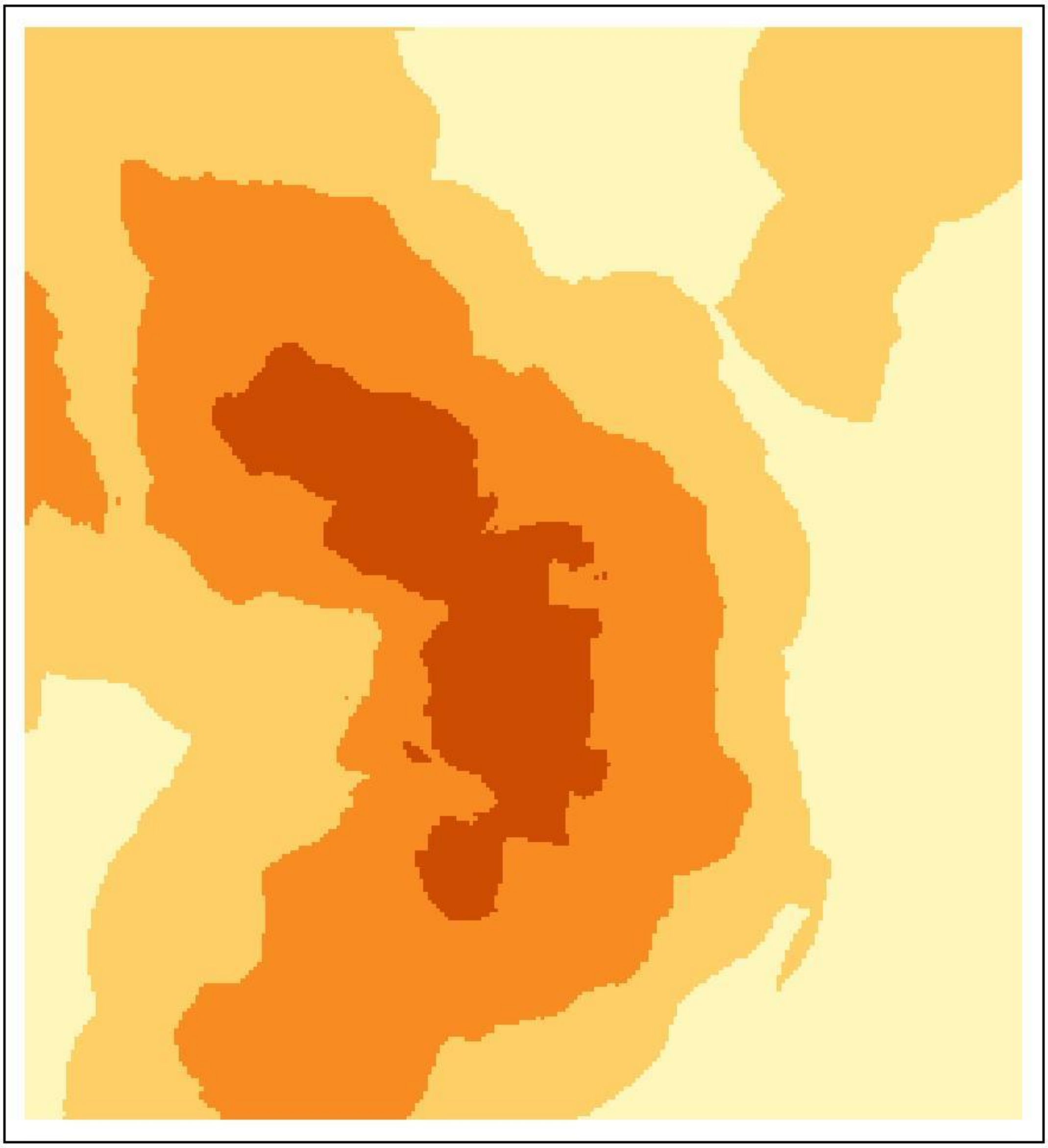


Figure S39 – local predictor road class 3 100

Figure S40 – local predictor road class 3 300

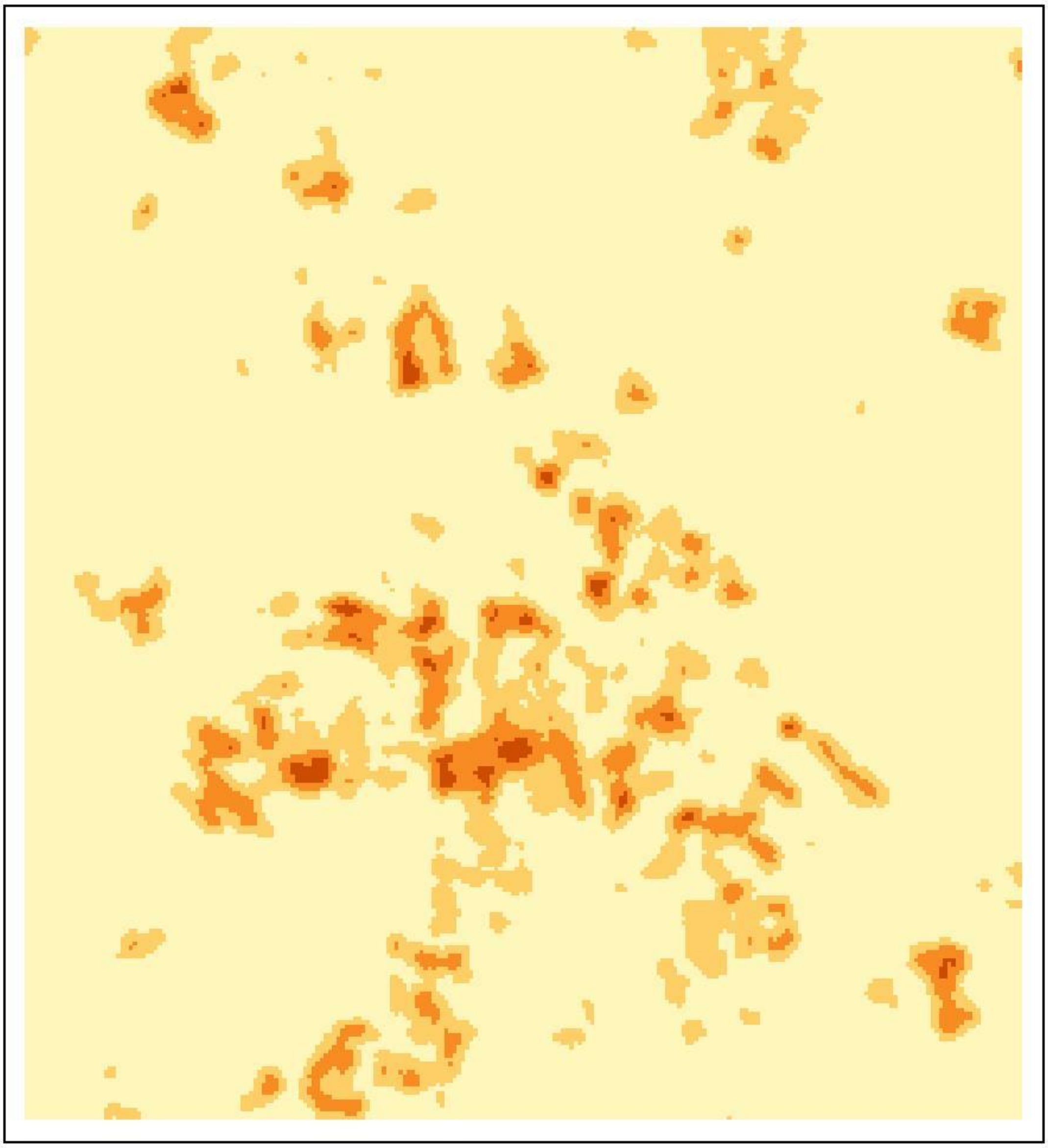


Figure S41 – local predictor trafbuf50

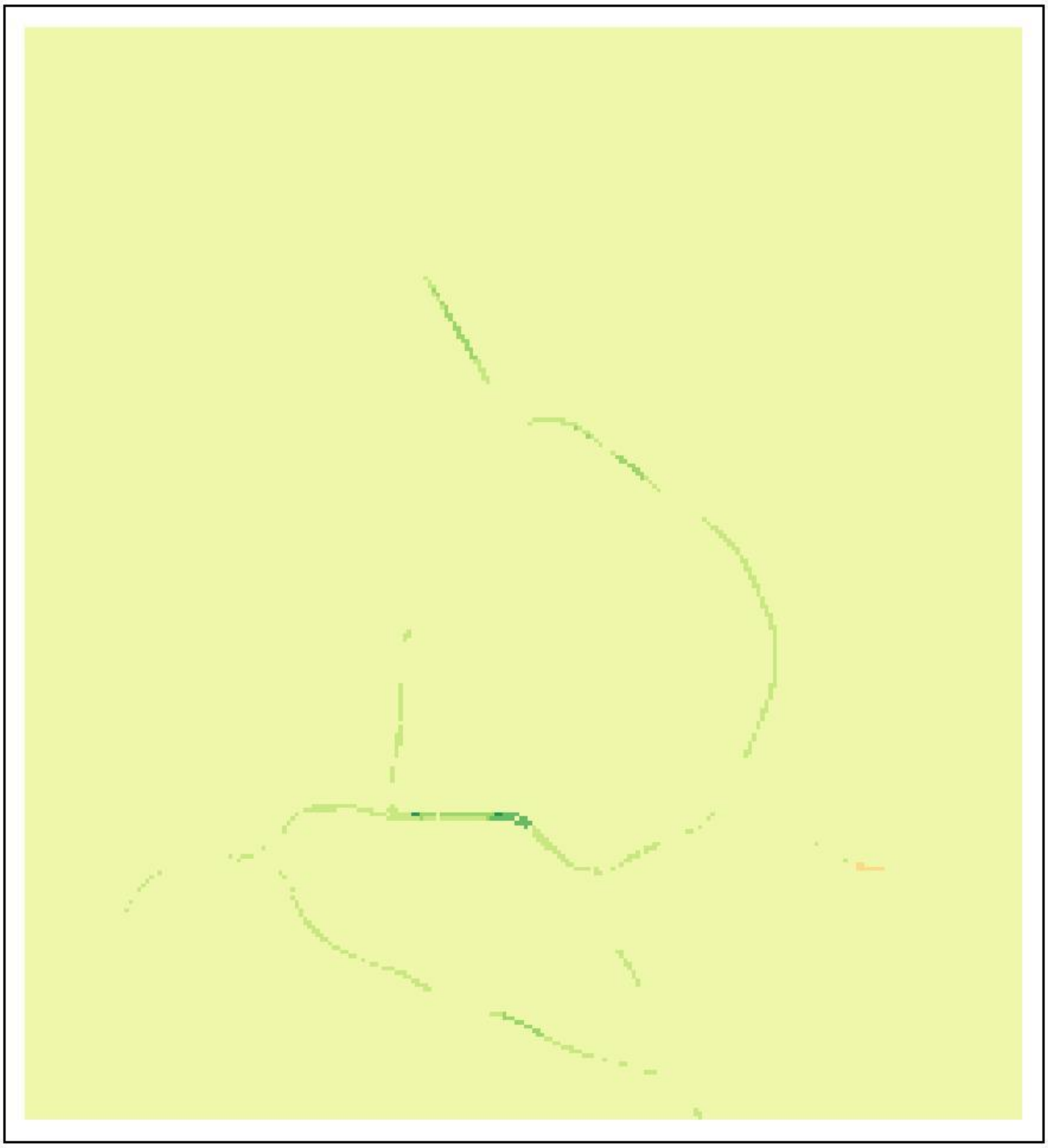


Figure S42 – Spatial distribution per spatial group – local - GRID100mx100m

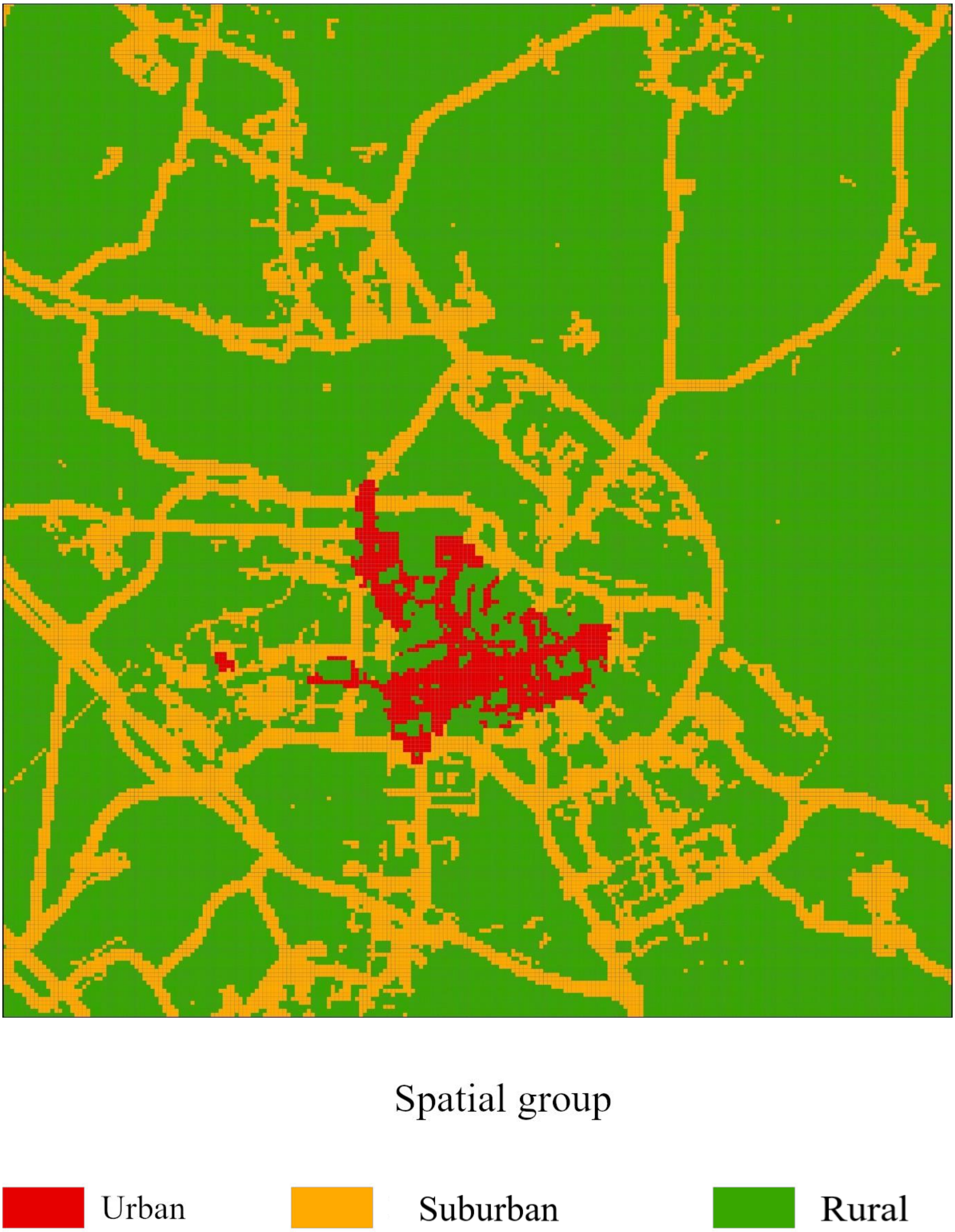


Figure S43 – Local data distribution per spatial group - nightlight_450

**In the main text, "suburban" refers to low population areas, while "rural" refers to far from road areas.*

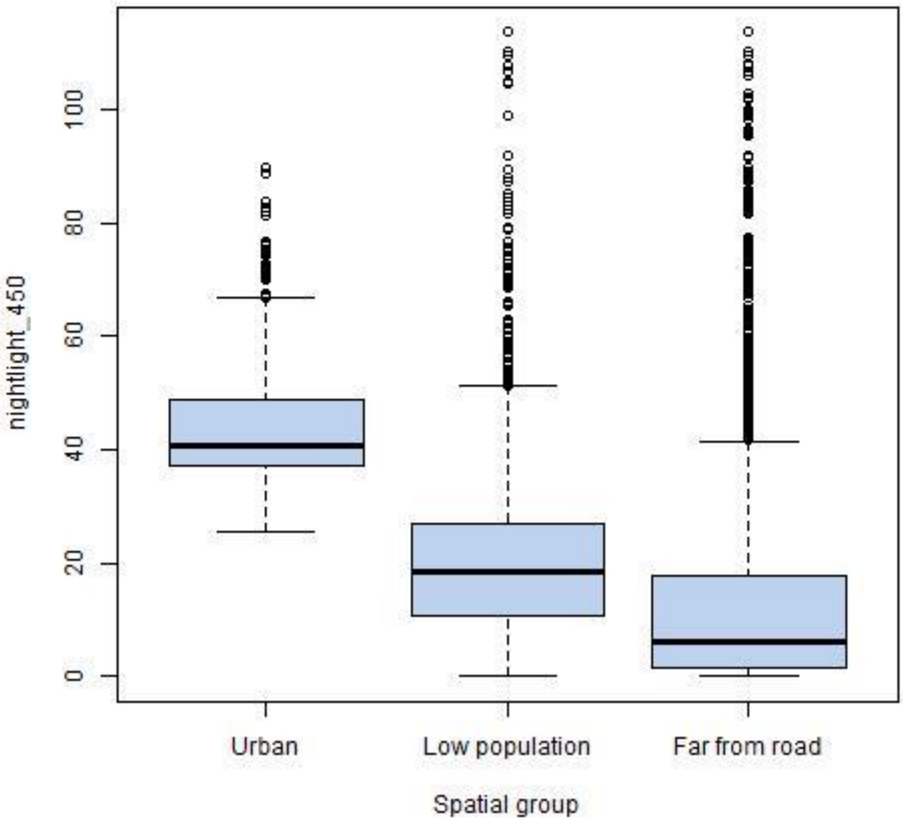


Figure S44 - Local data distribution per spatial group - nightlight_4950

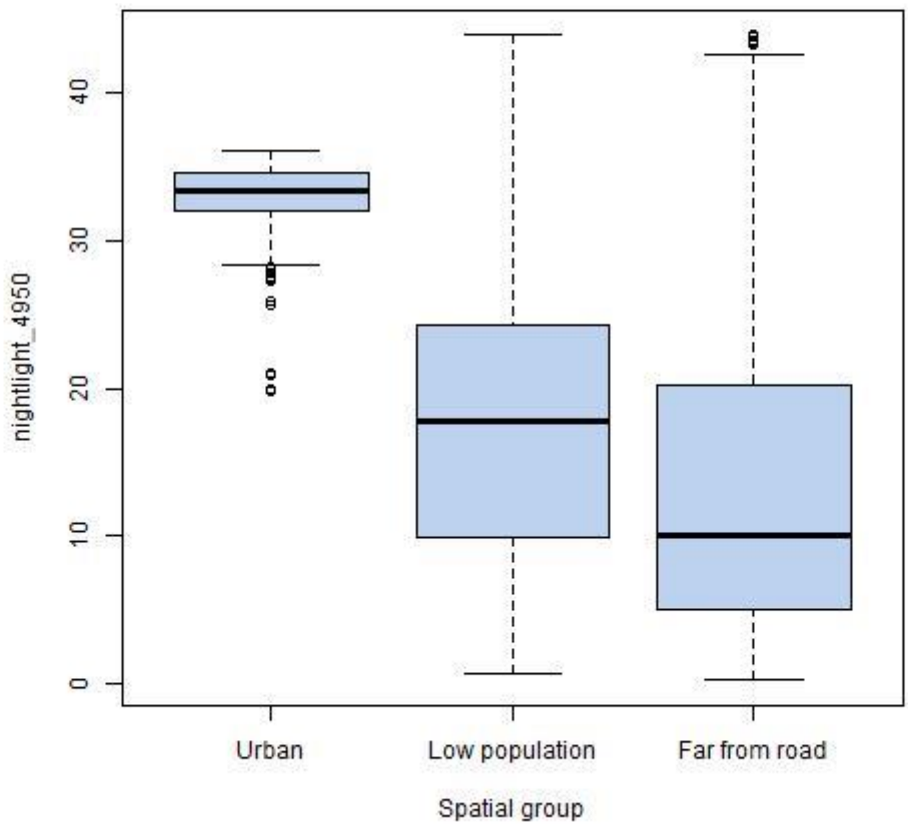


Figure S45 Local data distribution per spatial group – population 3000

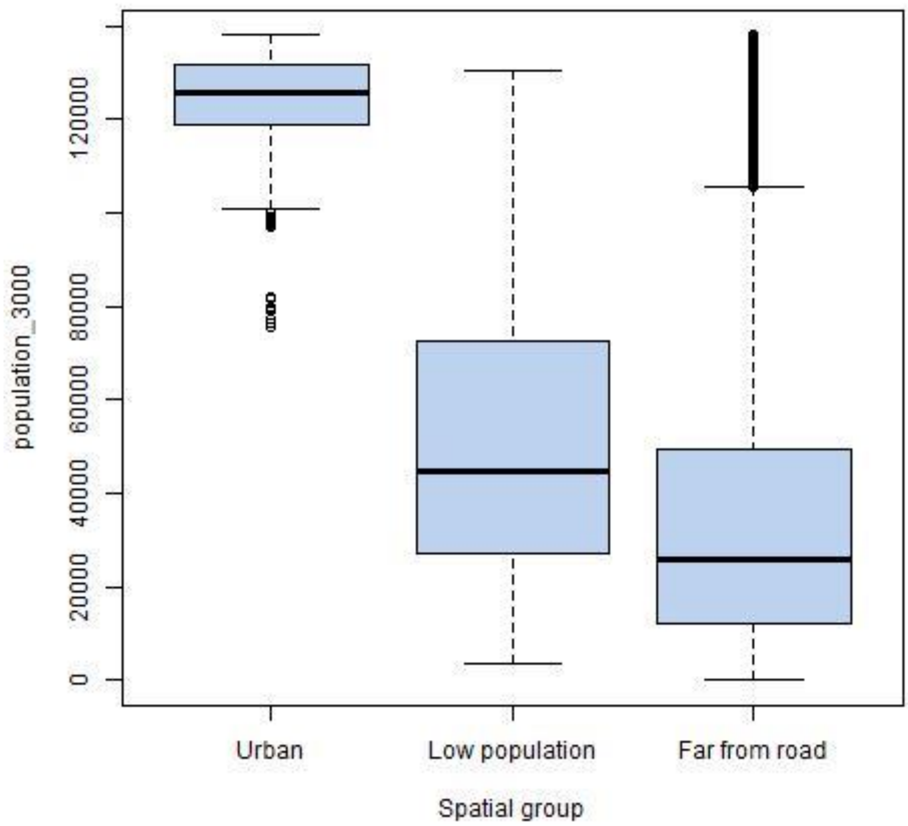


Figure S46 - Local data distribution per spatial group - road class 1 5000

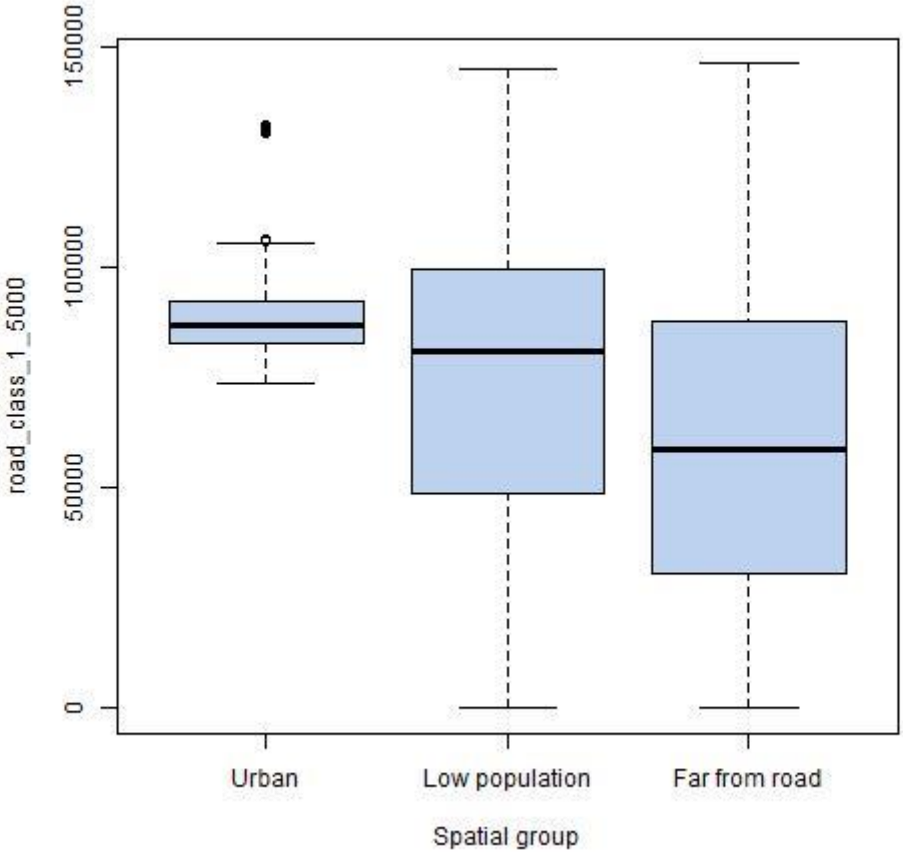


Figure S47 - Local data distribution per spatial group - road class 2 1000

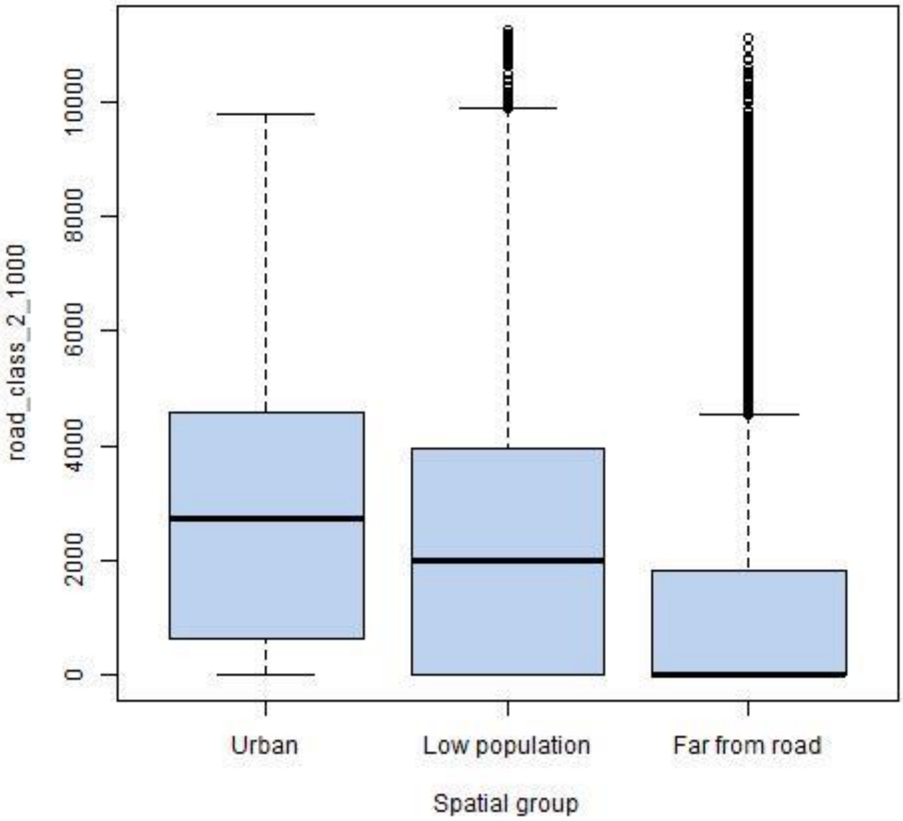


Figure S48 - Local data distribution per spatial group - road class 2 5000

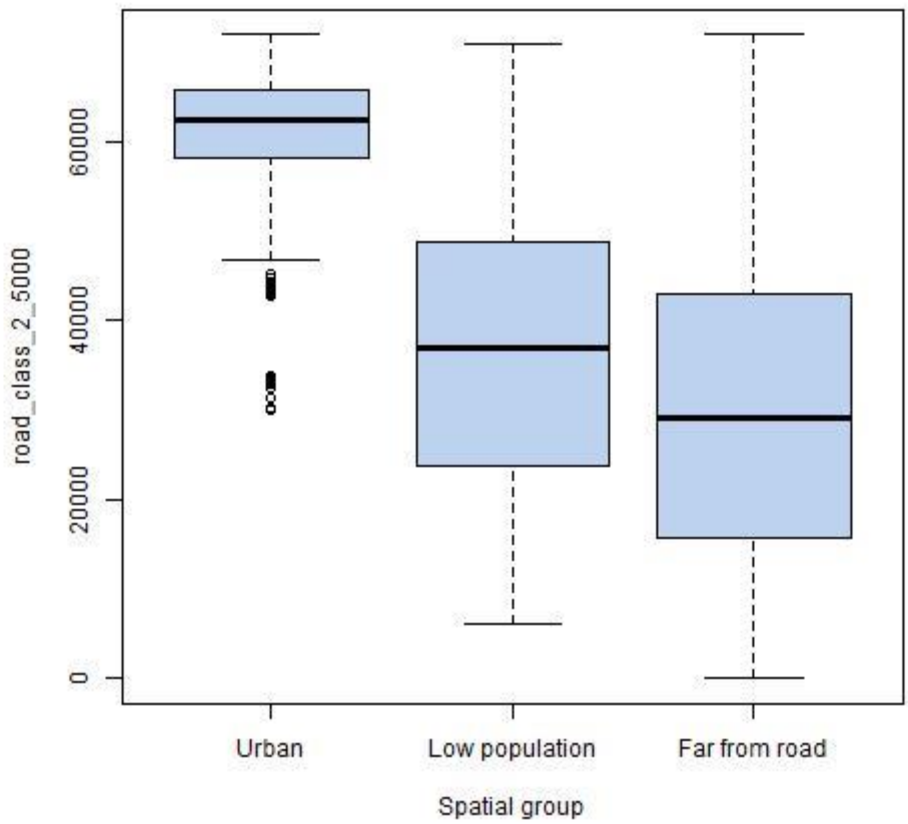


Figure S49 - Local data distribution per spatial group - road class 3 100

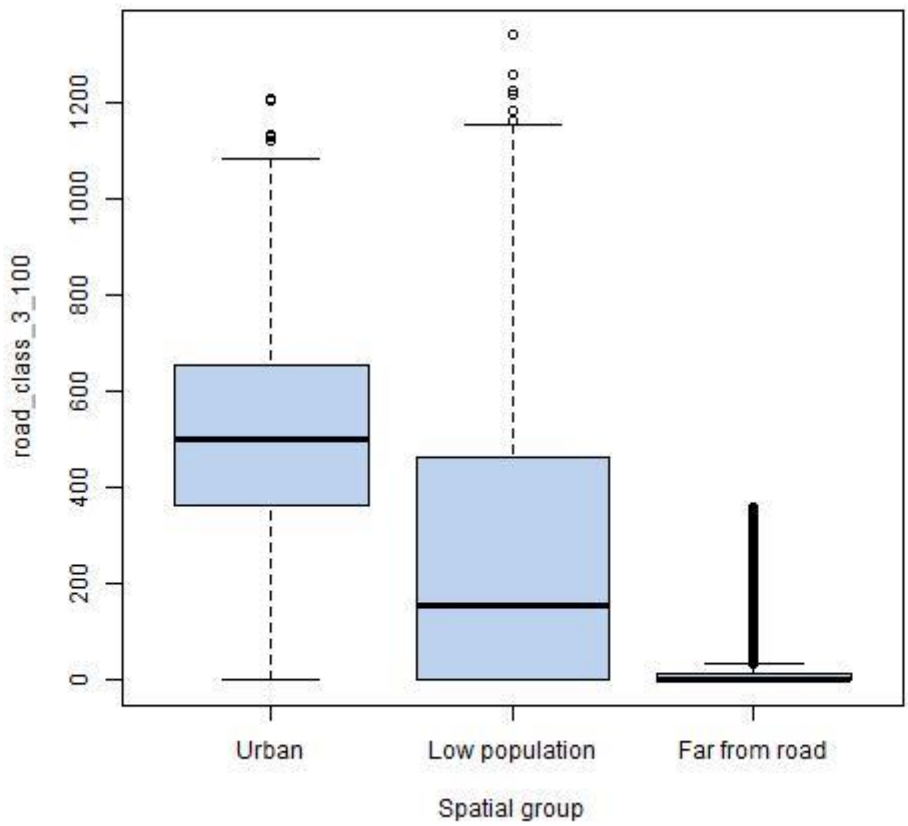


Figure S50 - Local data distribution per spatial group - road class 3 300

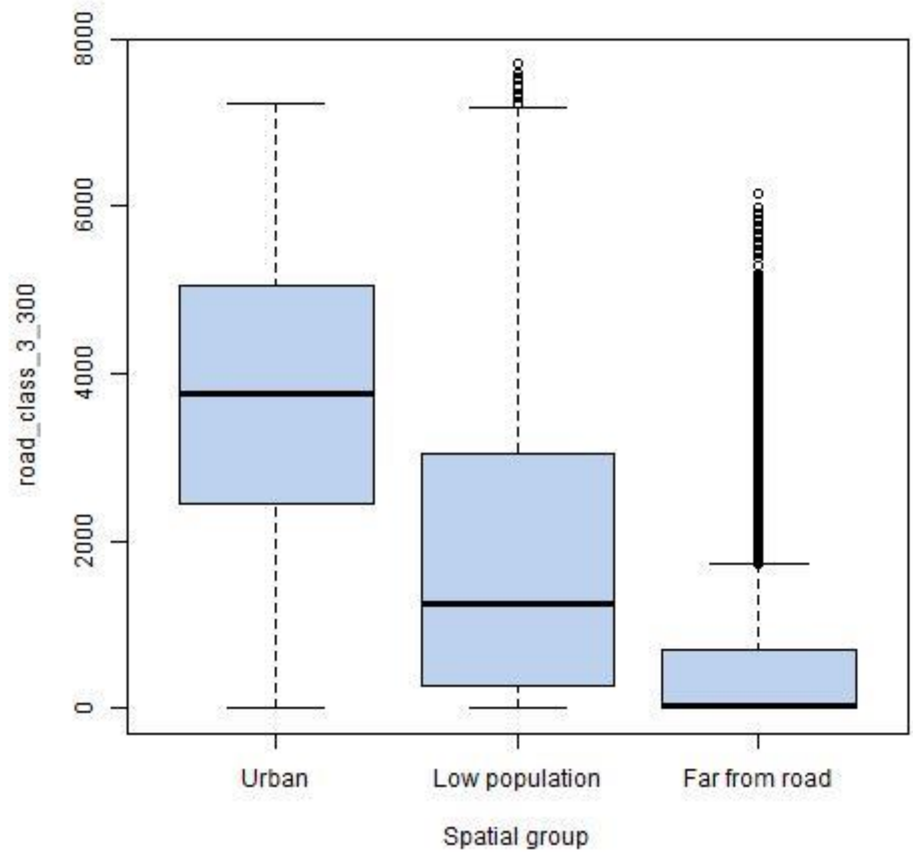


Figure S51 - Local data distribution per spatial group - trafbuf50

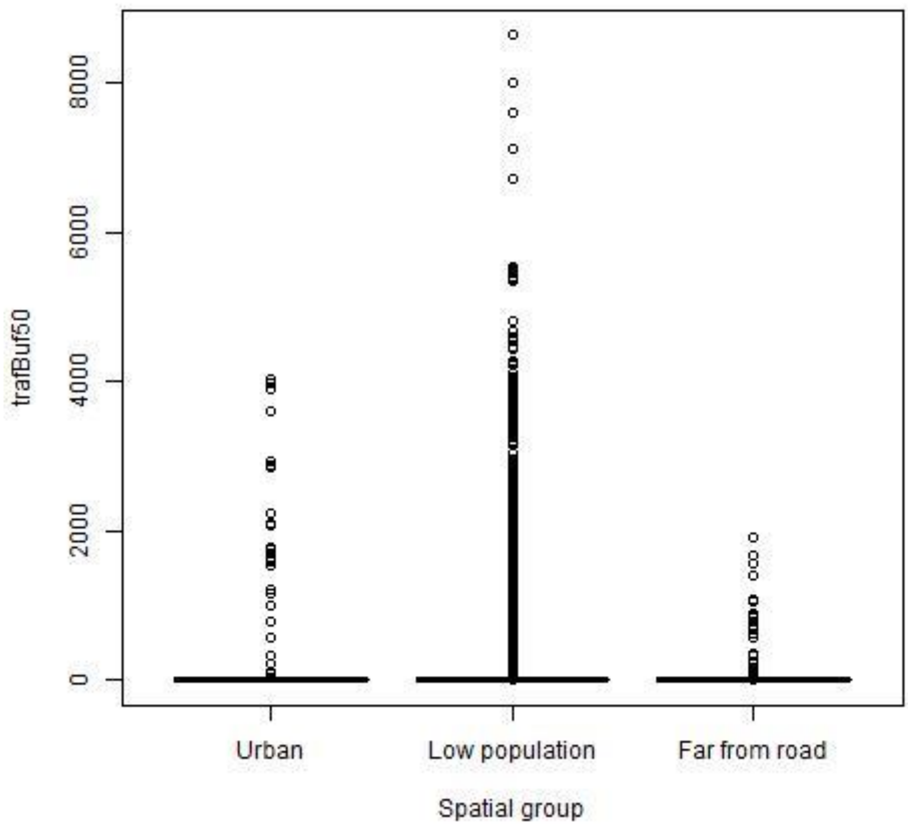


Figure S52 Predicted NO2 values by local models no outliers

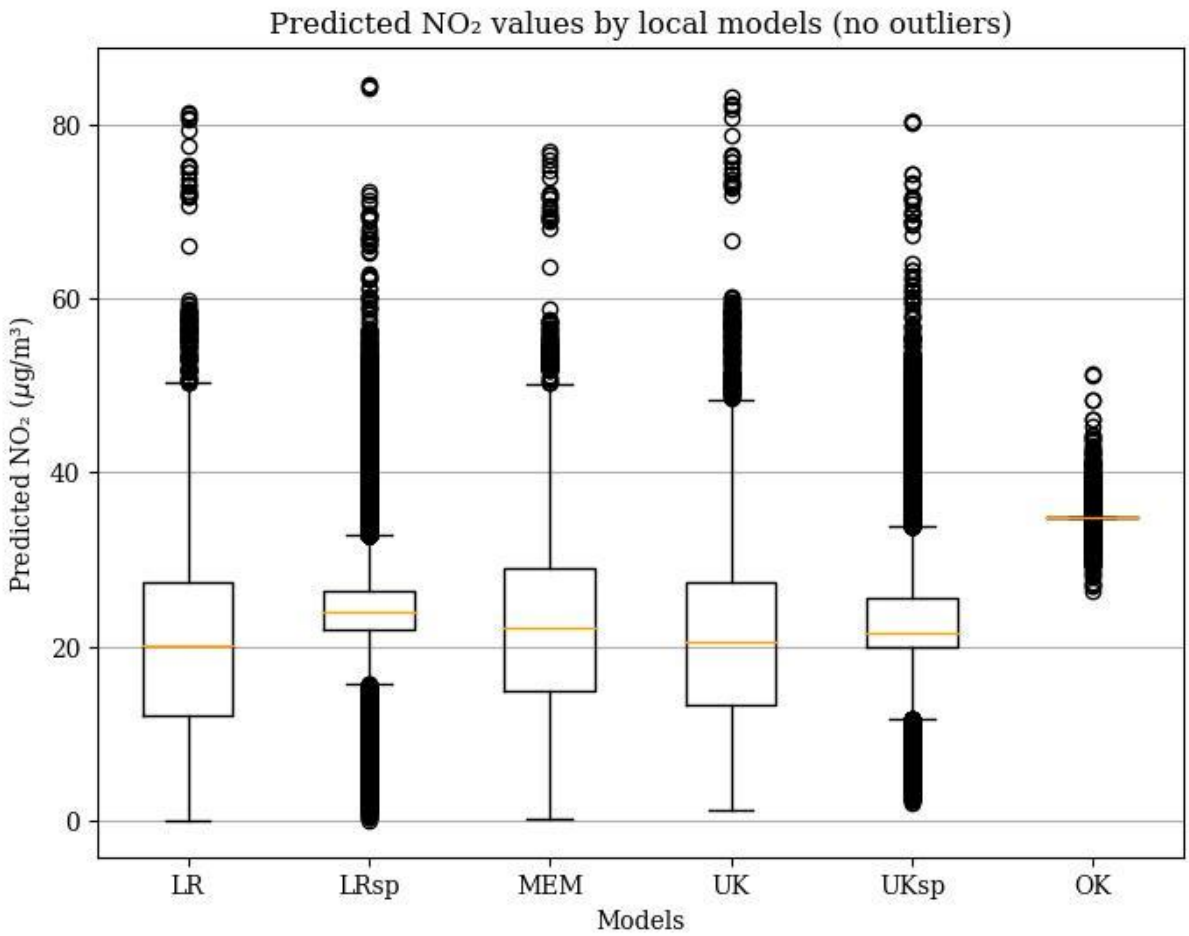


Figure S53 NO2 map Kerckhoffs

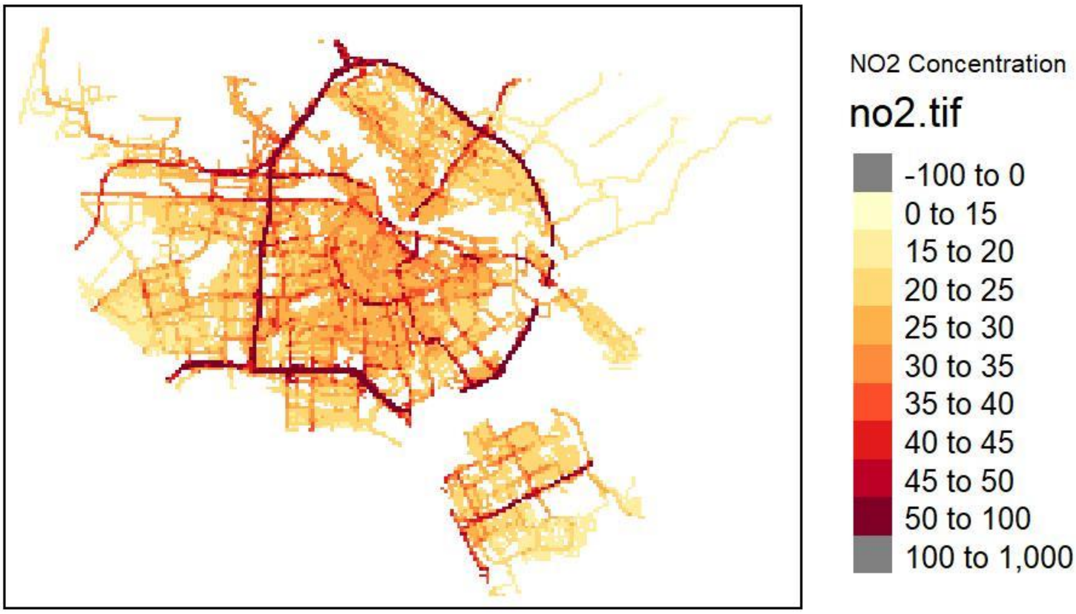


Figure S54. Comparing model predictions whereby the numbers equal the Pearson correlation coefficient. RF: Random Forest, XGB: XGBoost, LR: linear regression, LRsp: Linear Regression accounting for spatial groups, MEM: Mixed-Effects Model, UK: Universal Kriging, UKsp: Universal Kriging accounting for spatial groups, OK: Ordinary Kriging, no2: mobile NO$_{2}$ map.

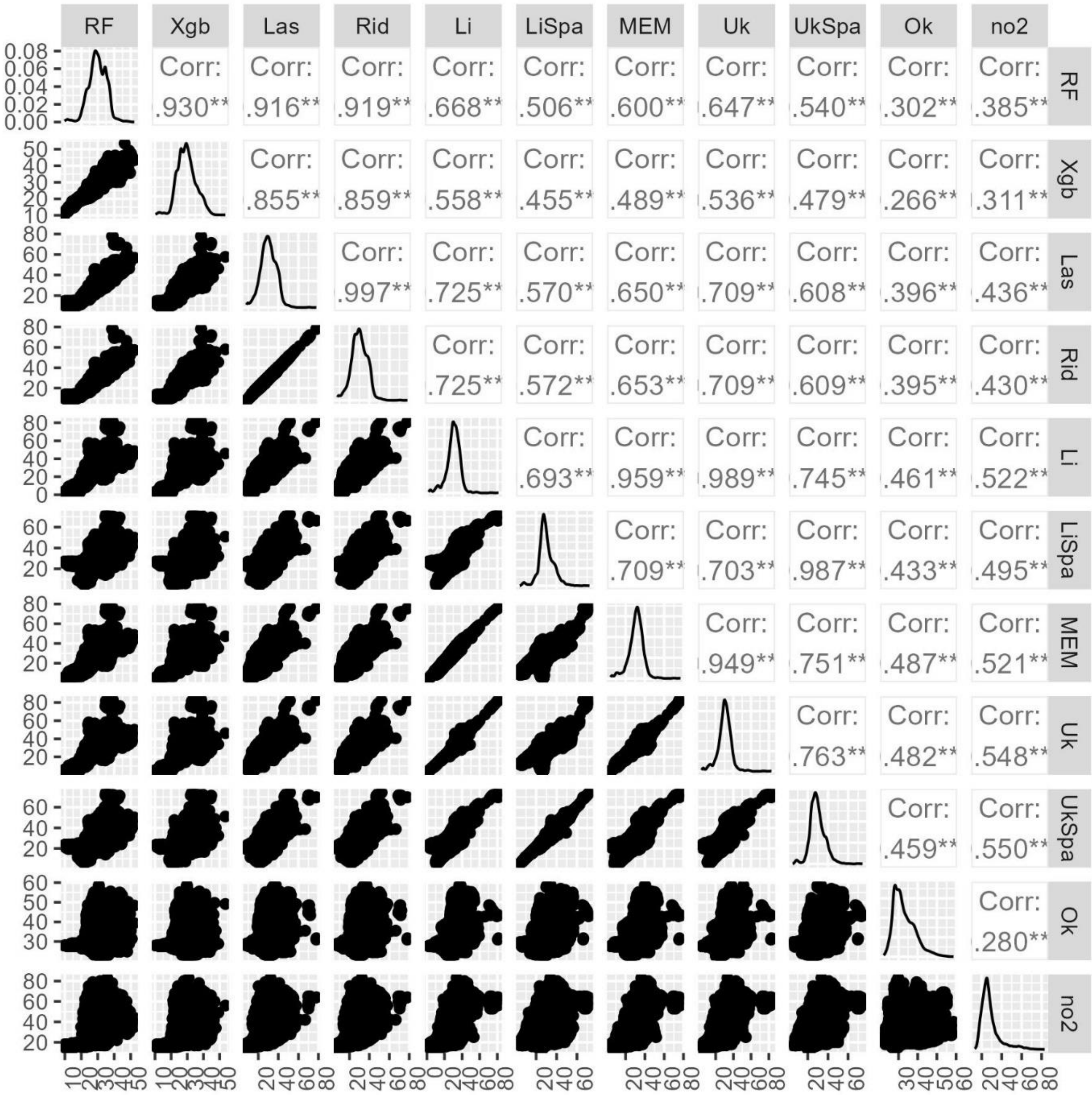


Figure S55 Spatial residual random forest (Predicted NO2 values by random forest - NO2 map Kerckhoffs)

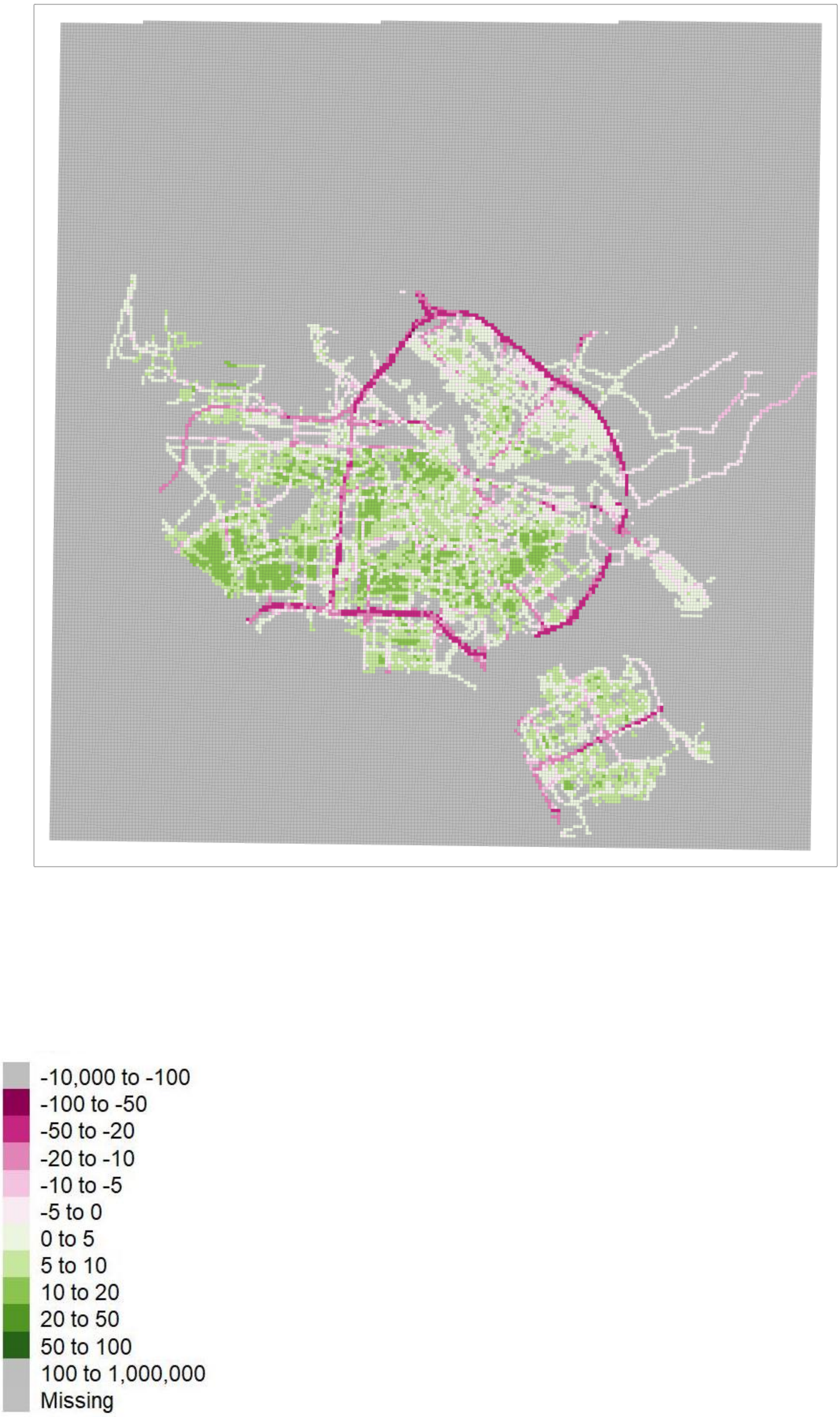


Figure S56 - Spatial residual LightGBM (Predicted NO2 values by LightGBM model - NO2 map Kerckhoffs)

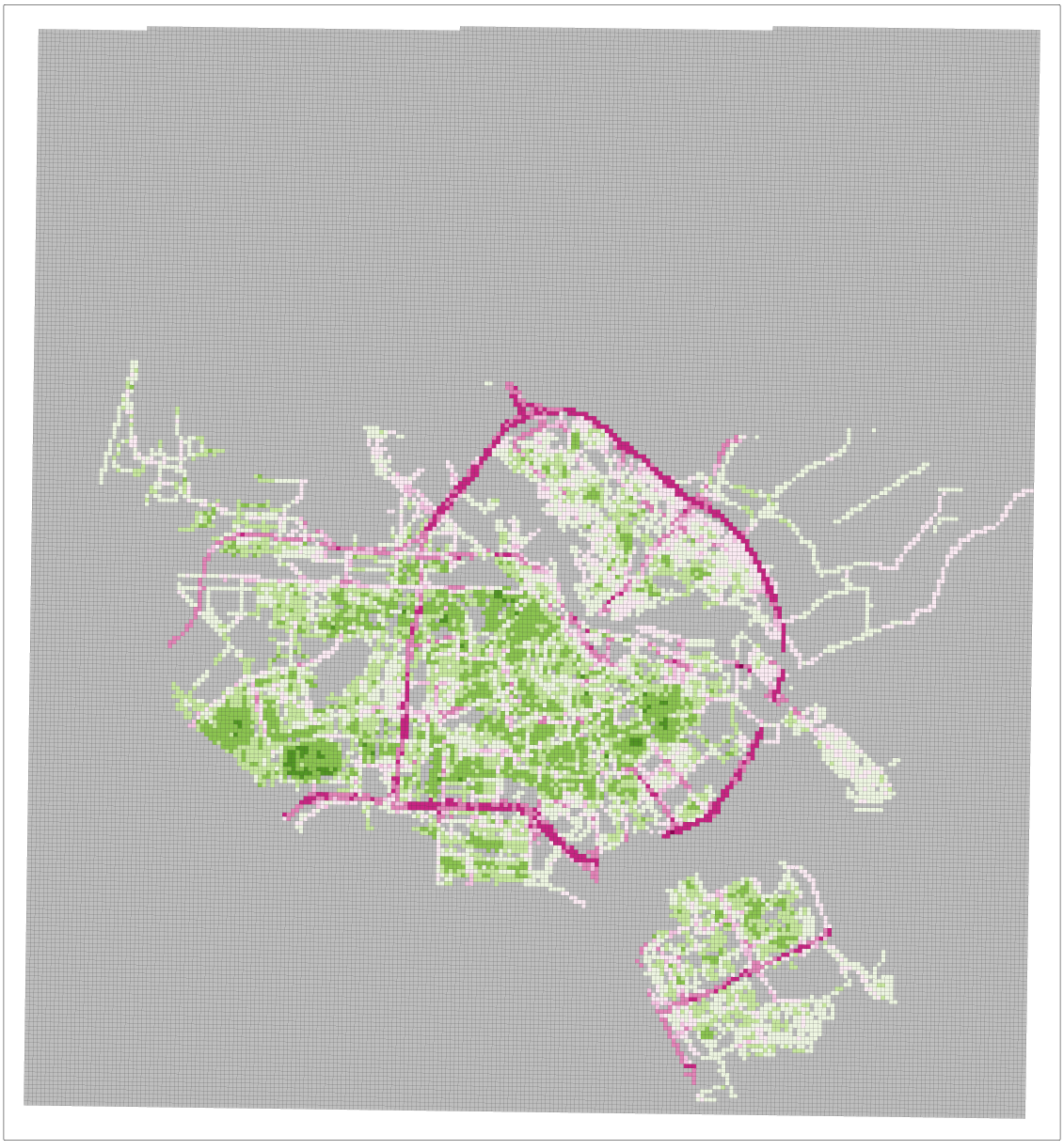


Figure S57 - Spatial residual Universal kriging – XGBoost kriging (Predicted NO2 values by XGBoost model - NO2 map Kerckhoffs)

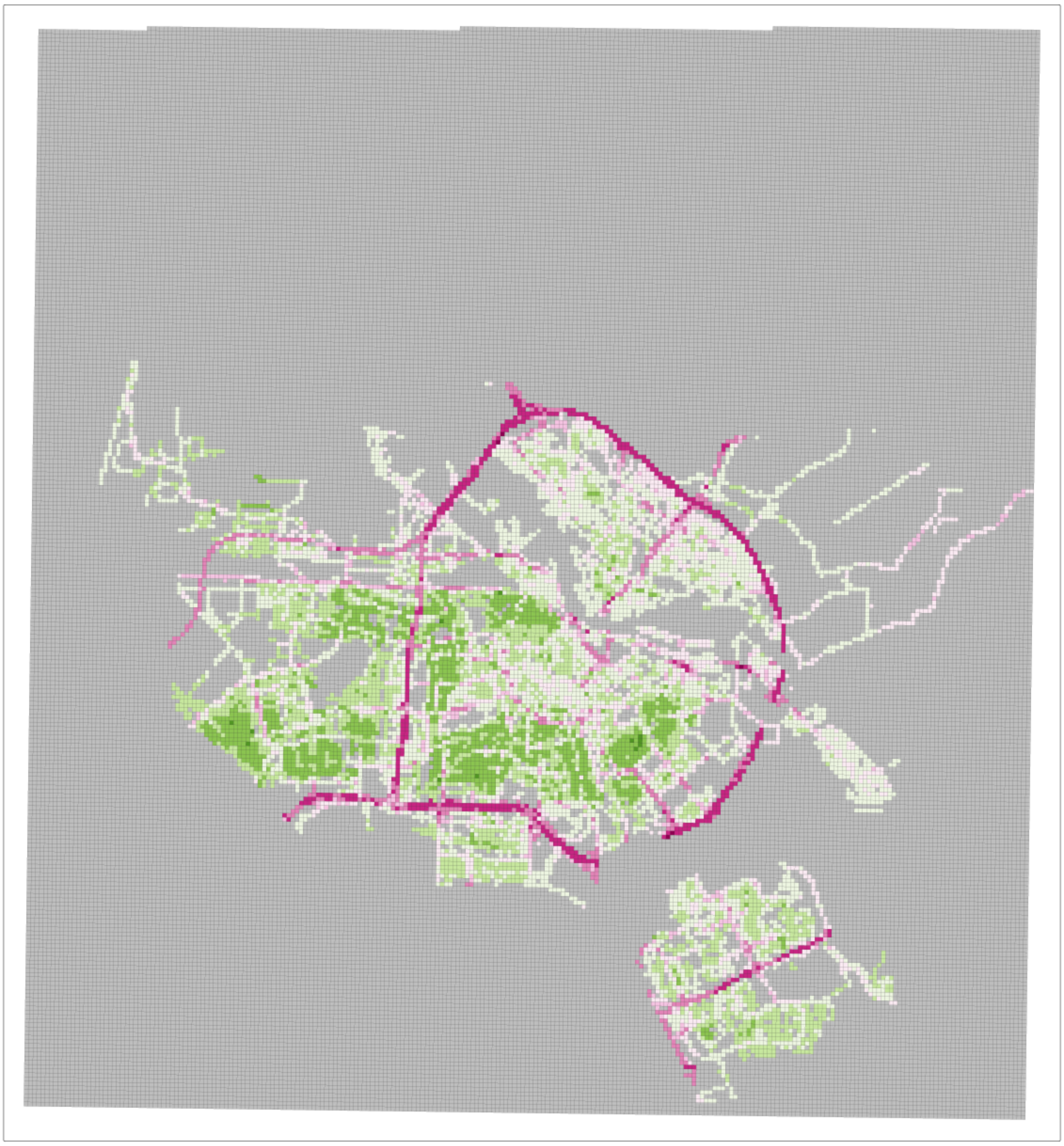


Figure S58 - Spatial residual LASSO (Predicted NO2 values by Lasso model - NO2 map Kerckhoffs)

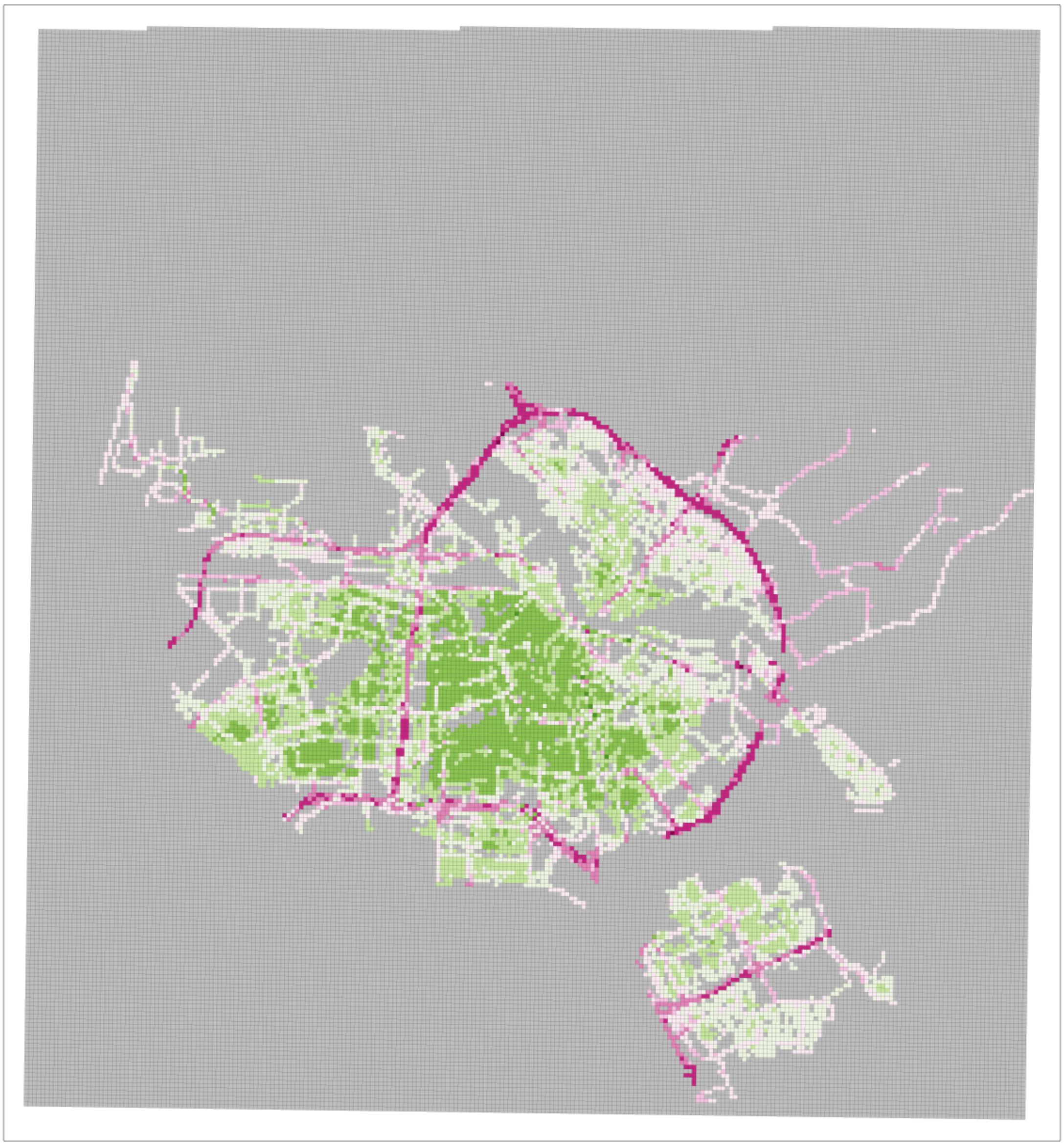


Figure S59 - Spatial residual Ridge (Predicted NO2 values by ridge model - NO2 map Kerckhoffs)

Figure S60 - Spatial residual Linear (Predicted NO2 values by Linear model - NO2 map Kerckhoffs)

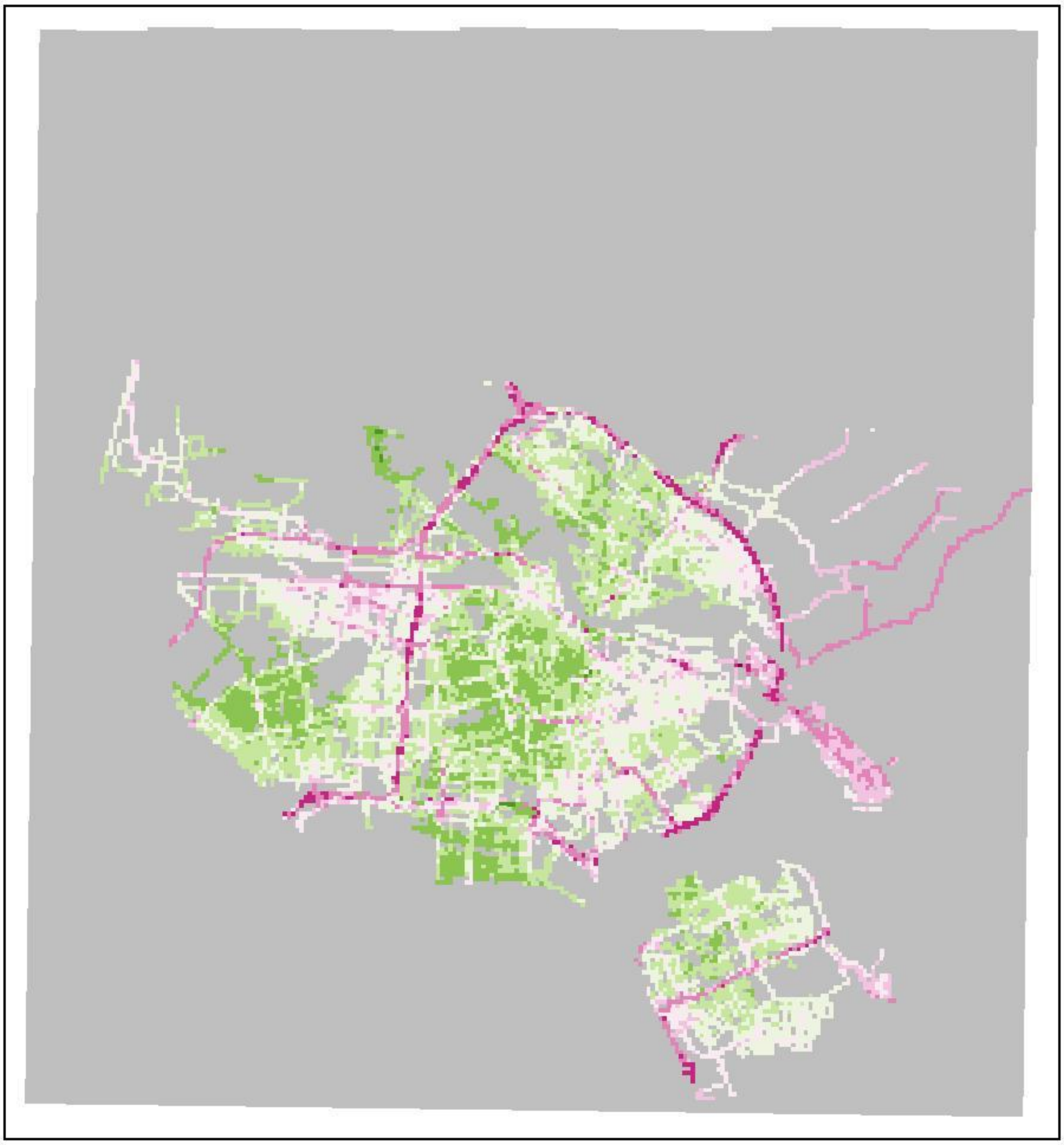


Figure S61 - Spatial residual Linear separated per spatial group (Predicted NO2 values by Linear model separated per spatial group - NO2 map Kerckhoffs)

Figure S62 - Spatial residual Mixed effects model (Predicted NO2 values by mixed-effects model model - NO2 map Kerckhoffs)

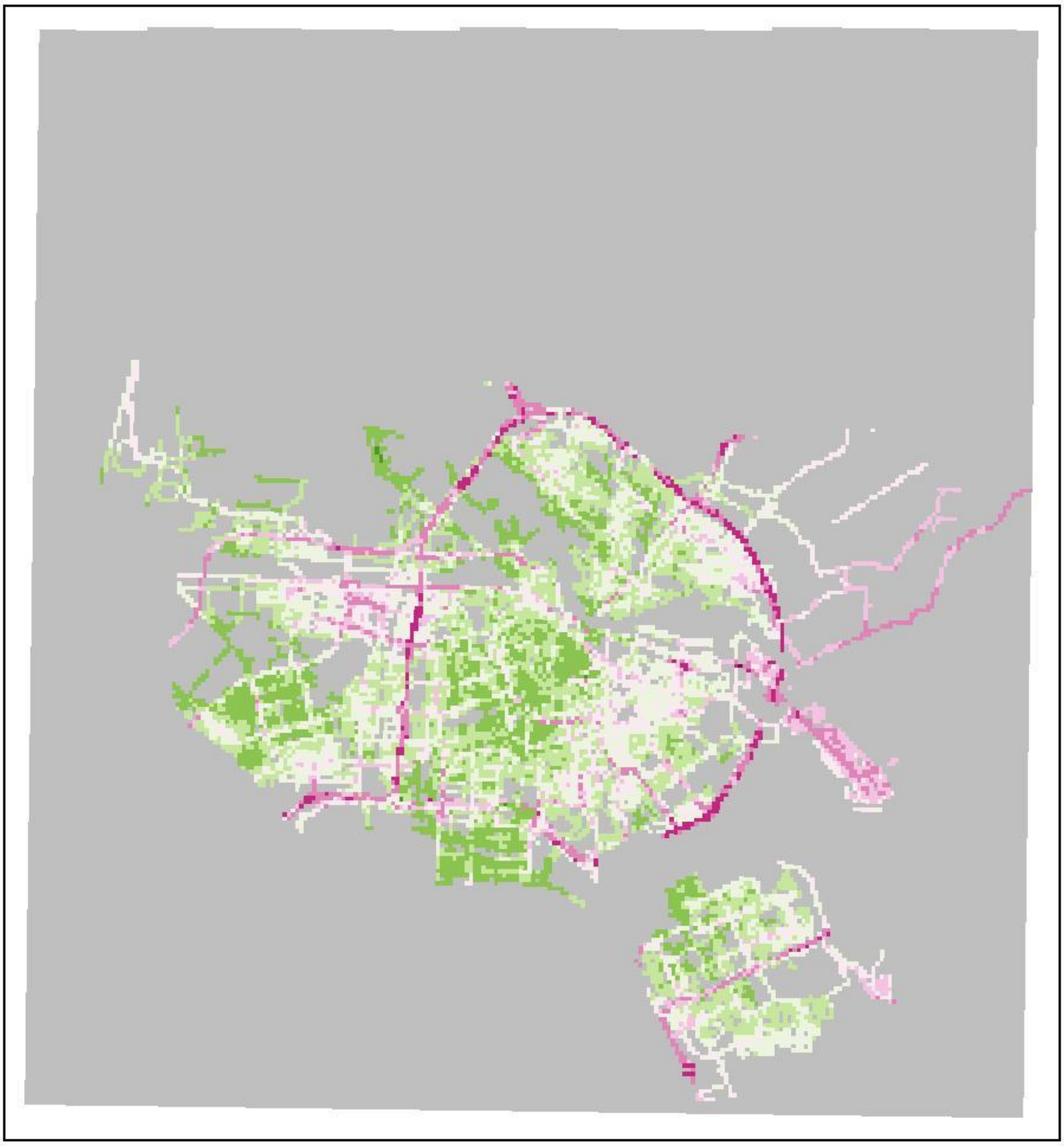


Figure S63 - Spatial residual ordinary kriging (Predicted NO2 values by ordinary kriging - NO2 map Kerckhoffs)

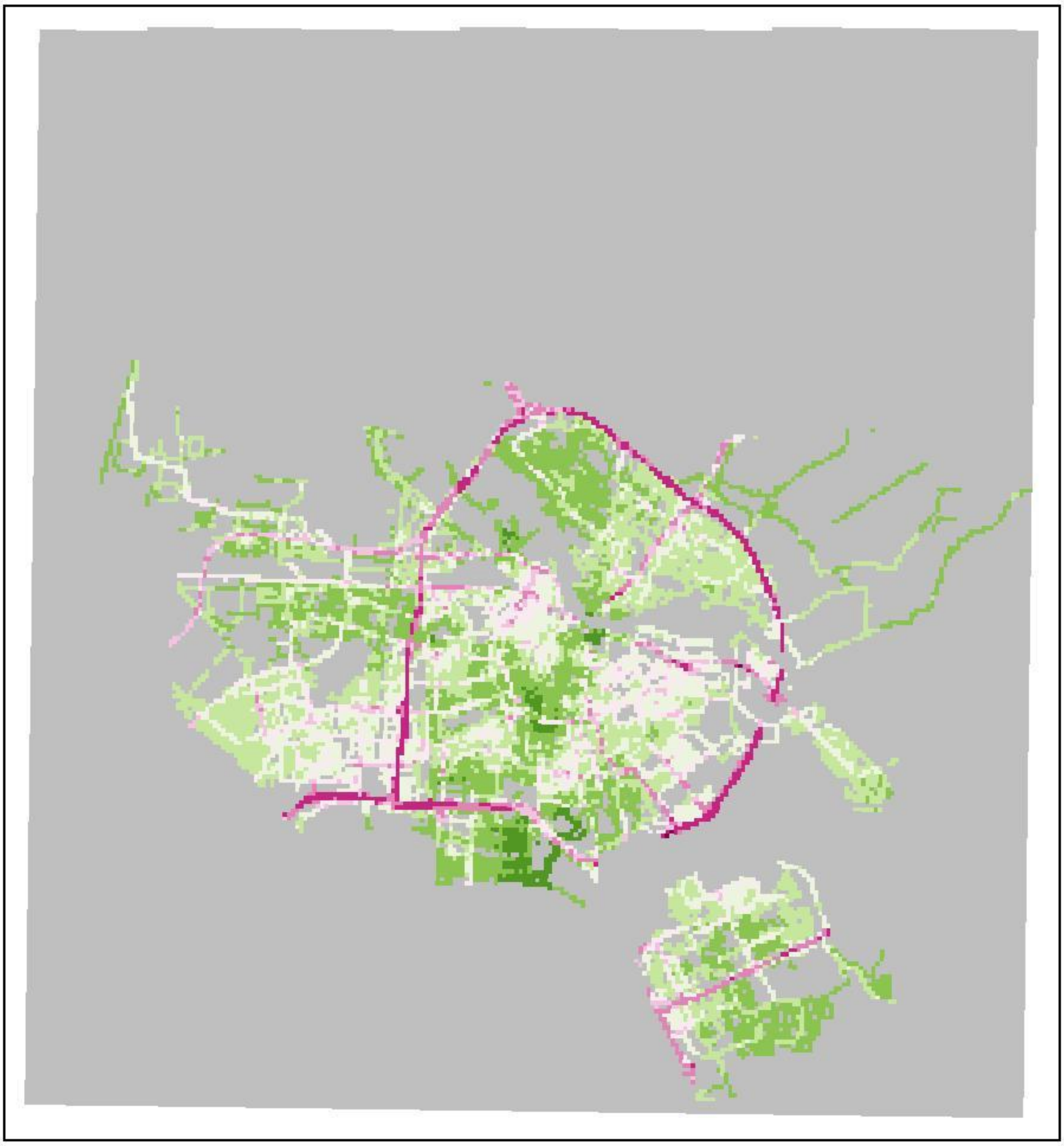


Figure S64 - Spatial residual universal kriging (Predicted NO2 values by universal kriging model - NO2 map Kerckhoffs)

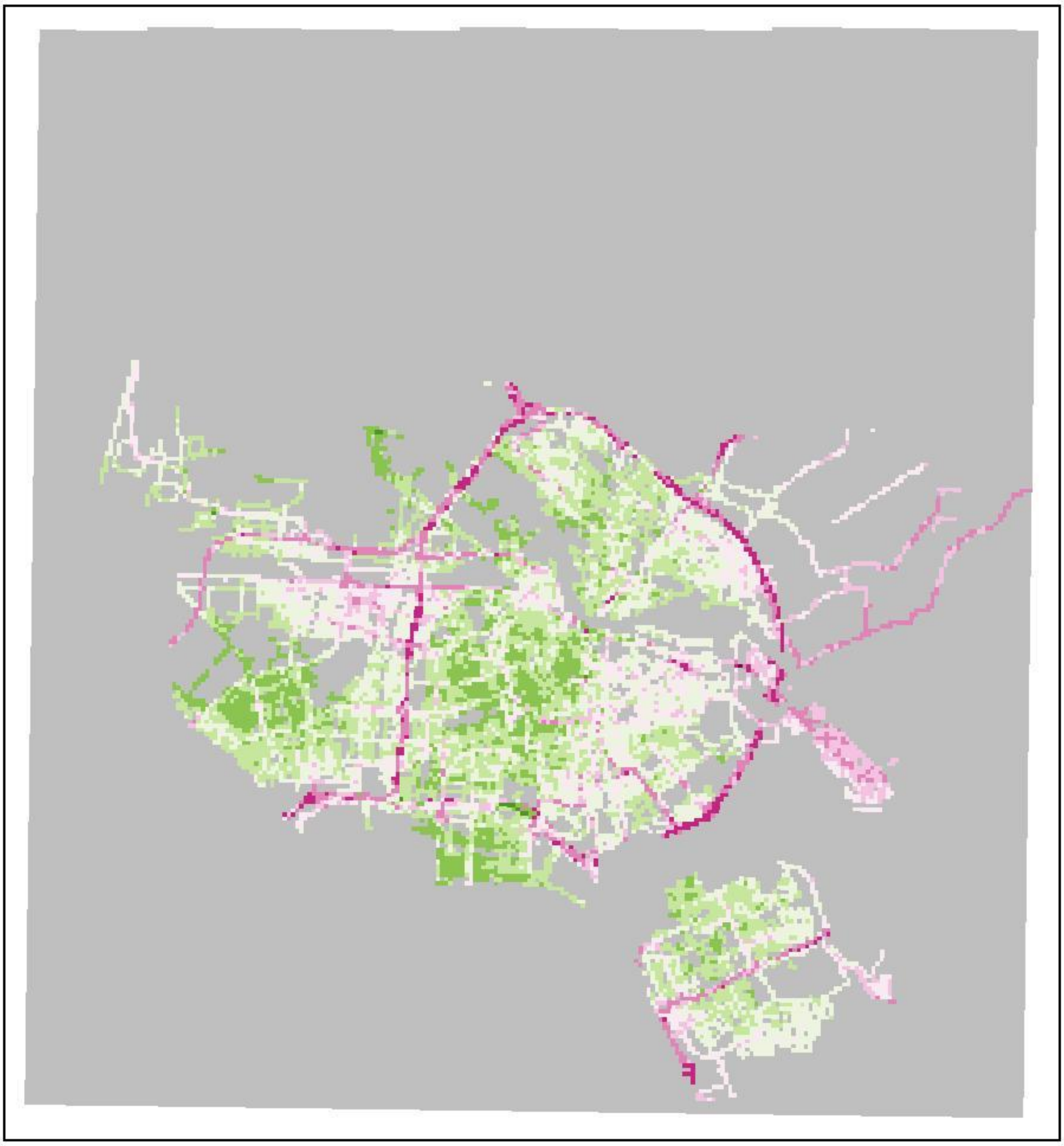


Figure S65 - Spatial residual Universal kriging – separated per spatial group (Predicted NO2 values by universal kriging separated per spatial group model - NO2 map Kerckhoffs)

Figure S66 – Differences in model prediction vs ground measurement stations in Amsterdam random forest (global)

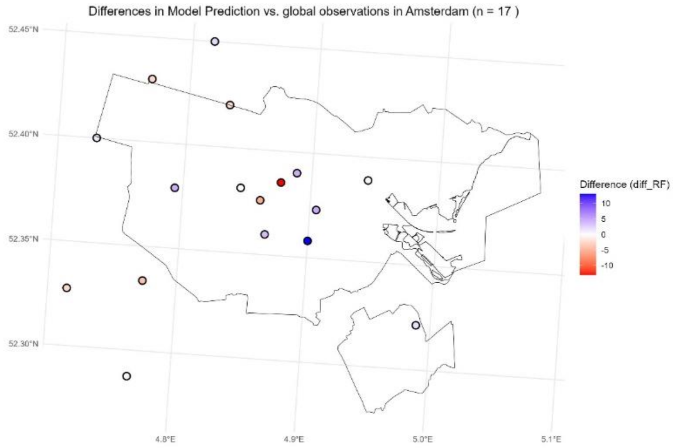


Figure S67 – Differences in model prediction vs ground measurement stations in Bayreuth random forest (global)

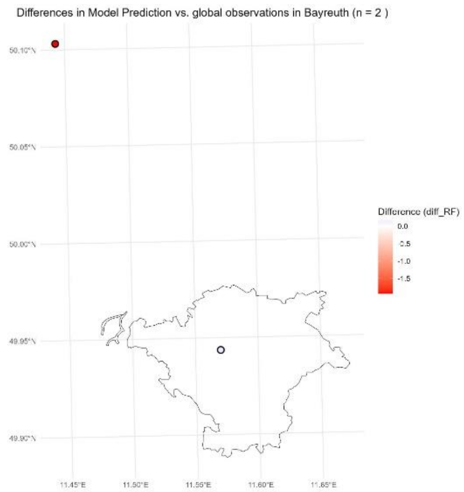


Figure S68 – Differences in model prediction vs ground measurement stations in Hamburg random forest (global)

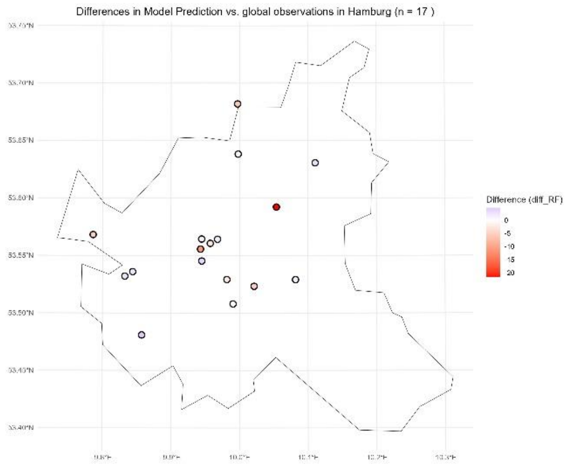


Figure S69 – Differences in model prediction vs ground measurement stations in Utrecht random forest (global)

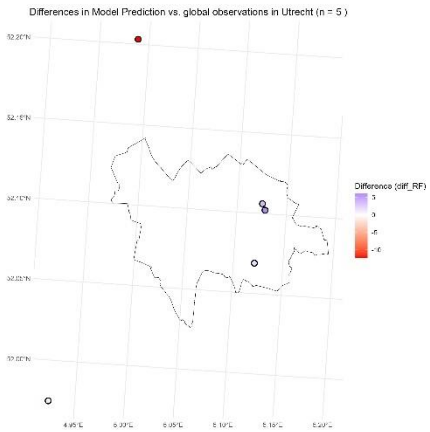


Figure S70 – Differences in model prediction vs ground measurement stations in Amsterdam XGBoost (global)

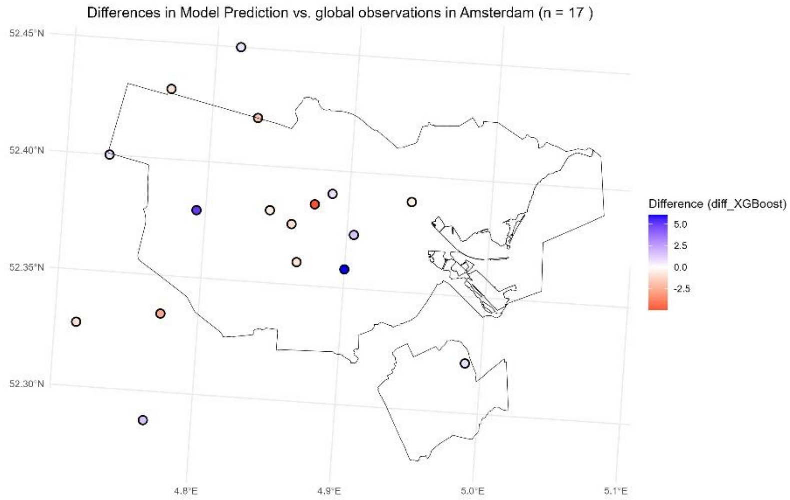


Figure S71 – Differences in model prediction vs ground measurement stations in Bayreuth XGBoost (global)

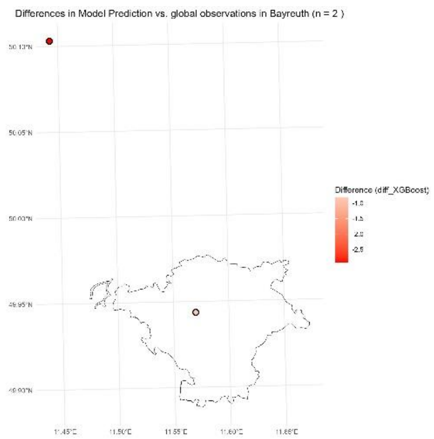


Figure S72 – Differences in model prediction vs ground measurement stations in Hamburg XGBoost (global)

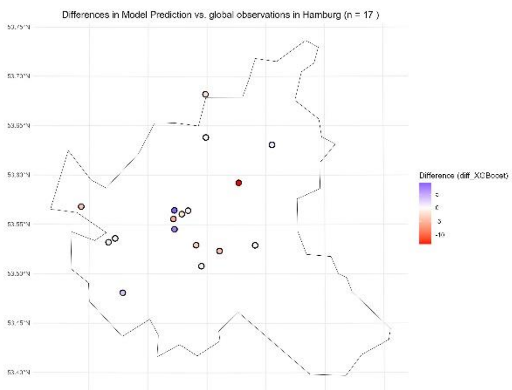


Figure S73 – Differences in model prediction vs ground measurement stations in Utrecht XGBoost (global)

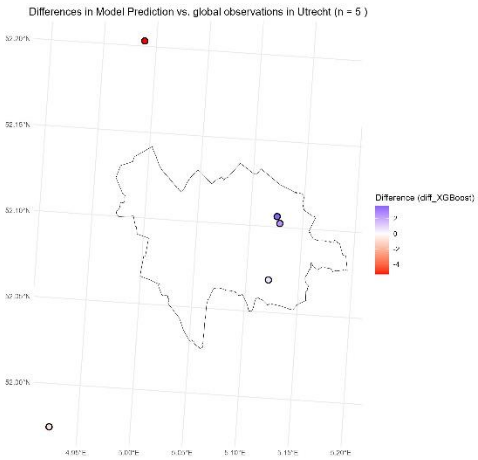


Figure S74 – Differences in model prediction vs ground measurement stations in Amsterdam LightGBM (global)

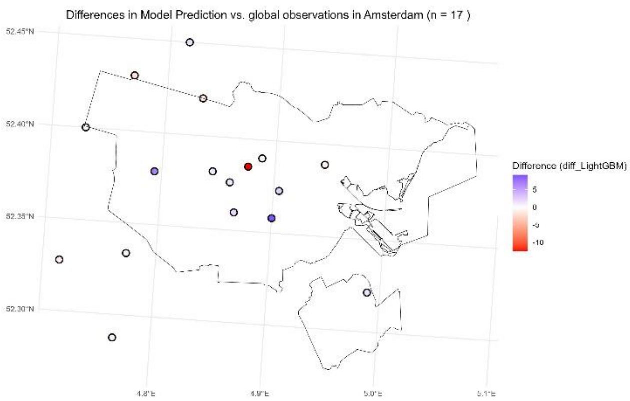


Figure S75 – Differences in model prediction vs ground measurement stations in Bayreuth LightGBM (global)

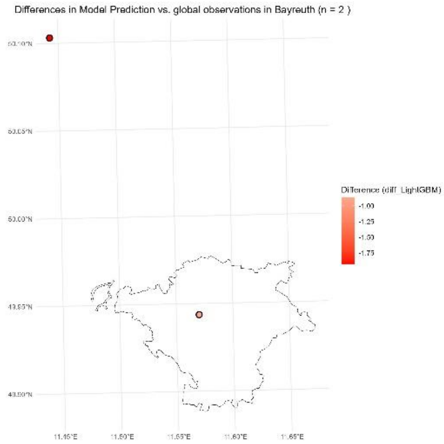


Figure S76 – Differences in model prediction vs ground measurement stations in Hamburg LightGBM (global)

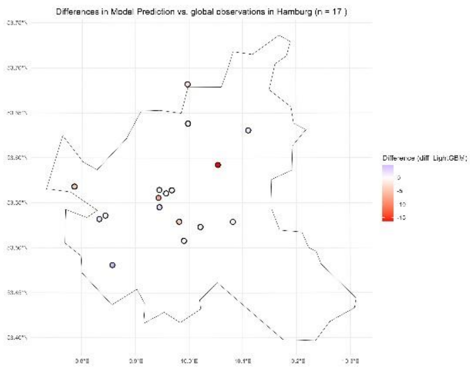


Figure S77 – Differences in model prediction vs ground measurement stations in Utrecht LightGBM (global)

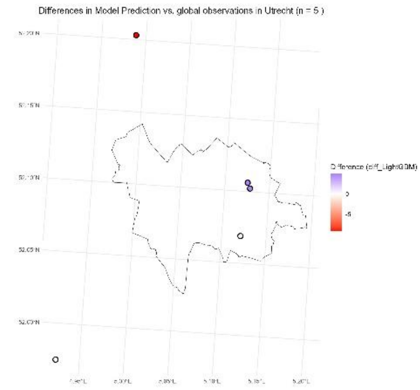


Figure S78 – Differences in model prediction vs ground measurement stations in Amsterdam LASSO (global)

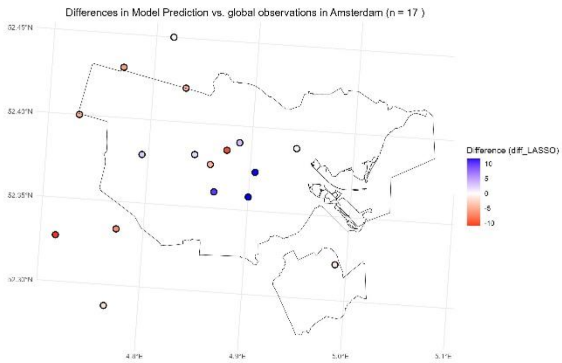


Figure S79 – Differences in model prediction vs ground measurement stations in Bayreuth LASSO (global)

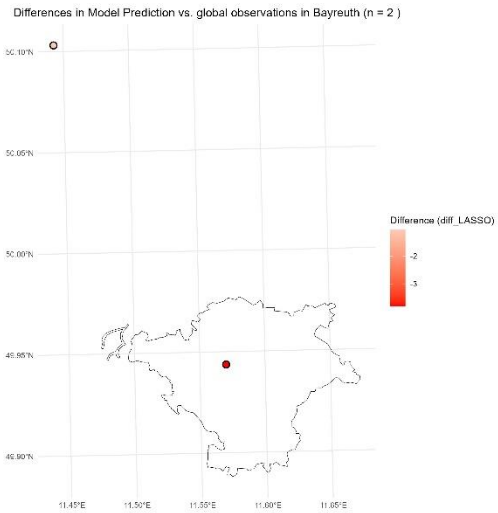


Figure S80 – Differences in model prediction vs ground measurement stations in Hamburg LASSO (global)

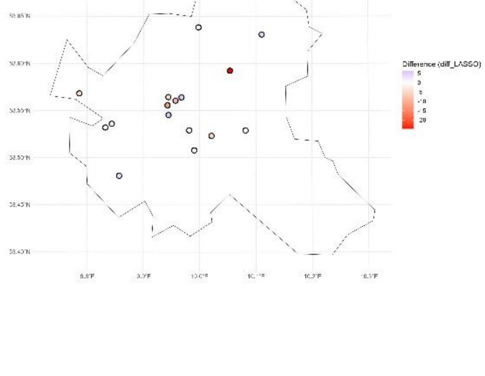


Figure S81 – Differences in model prediction vs ground measurement stations in Utrecht LASSO (global)

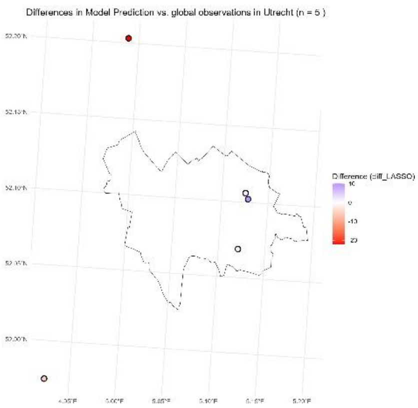


Figure S82 – Differences in model prediction vs ground measurement stations in Amsterdam Ridge (global)

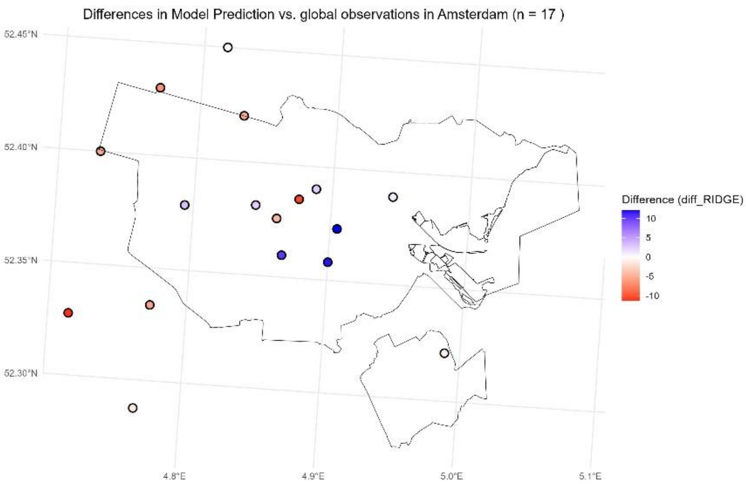


Figure S83 – Differences in model prediction vs ground measurement stations in Bayreuth Ridge (global)

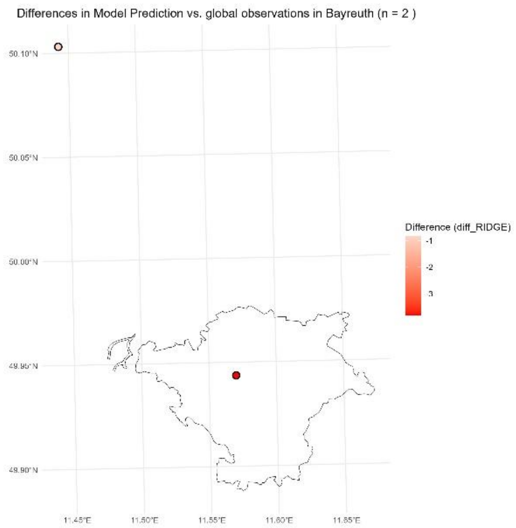


Figure S84 – Differences in model prediction vs ground measurement stations in Hamburg Ridge (global)

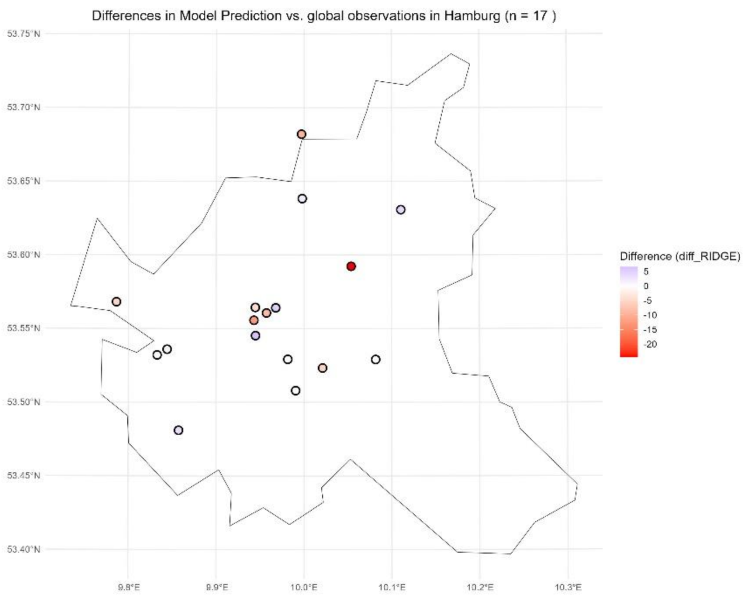


Figure S85 – Differences in model prediction vs ground measurement stations in Utrecht Ridge (global)

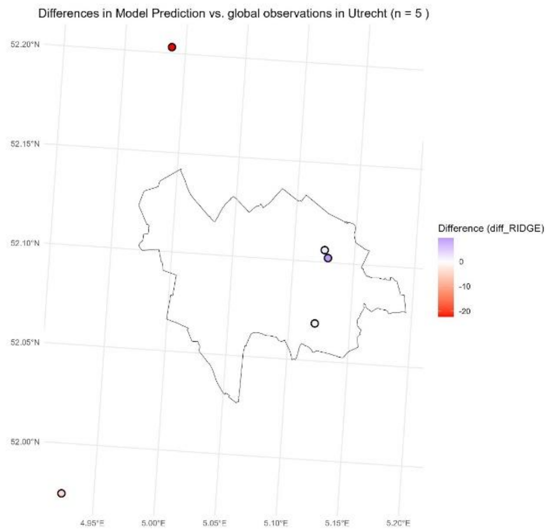


Figure S86 – Differences in model prediction vs ground measurement stations in Amsterdam linear (local)

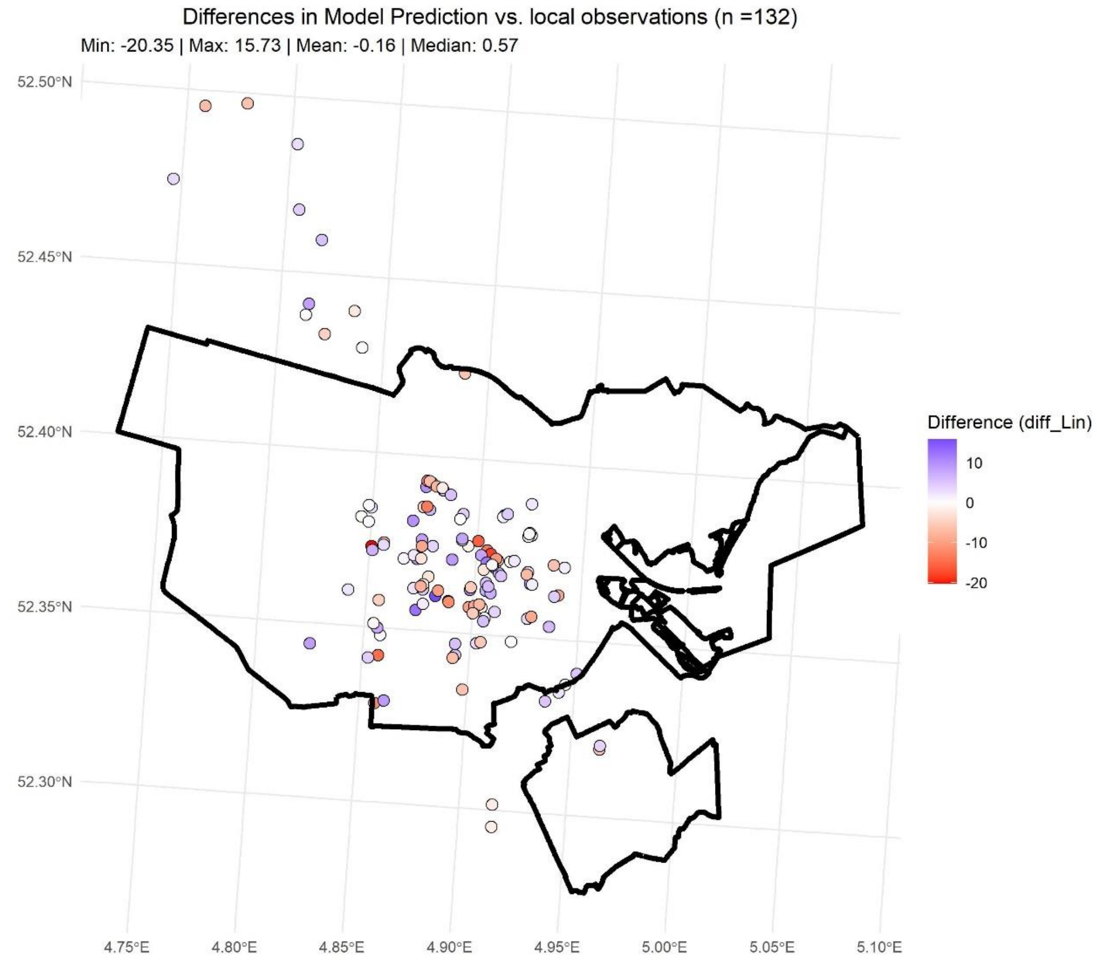


Figure S87 – Differences in model prediction vs ground measurement stations in Amsterdam linear separating for spatial groups (local)

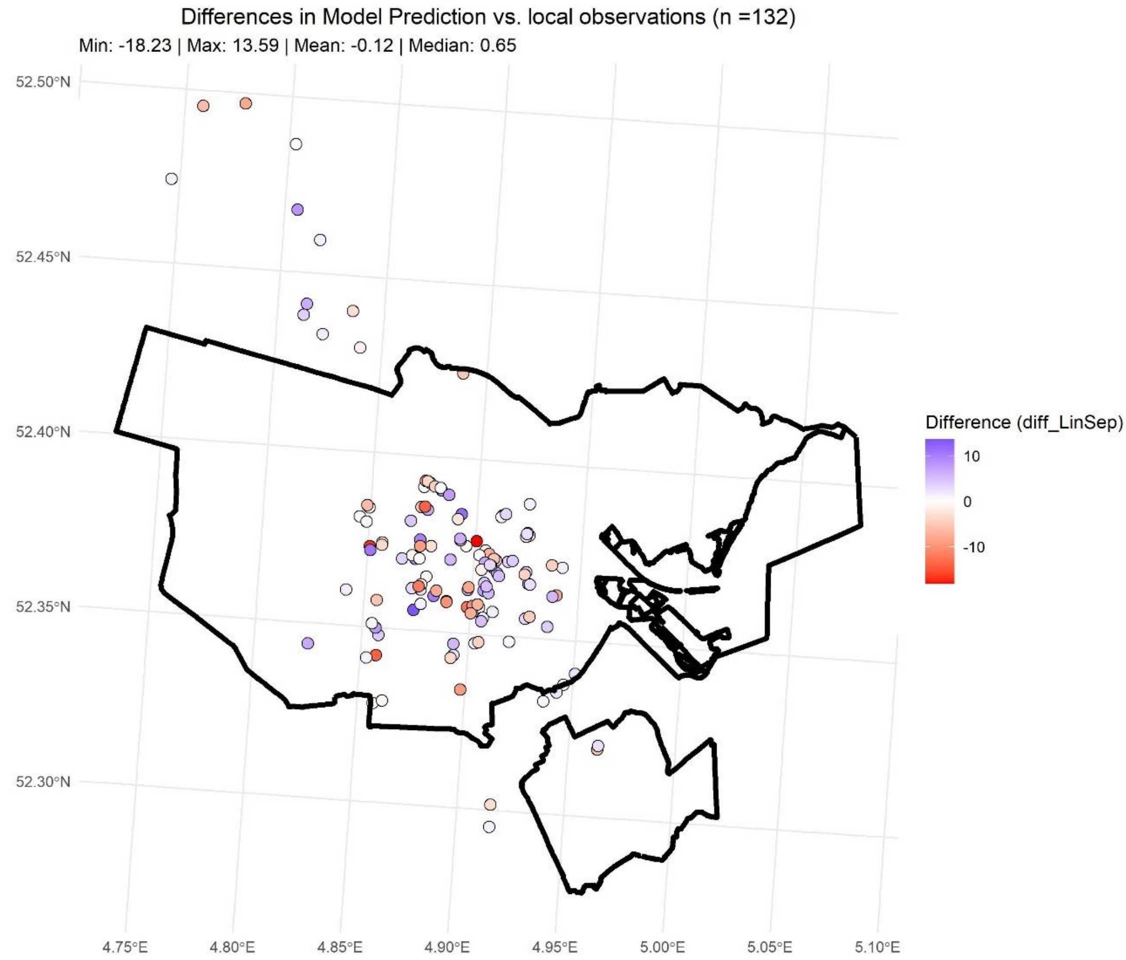


Figure S88 – Differences in model prediction vs ground measurement stations in Amsterdam mixed-effects model (local)

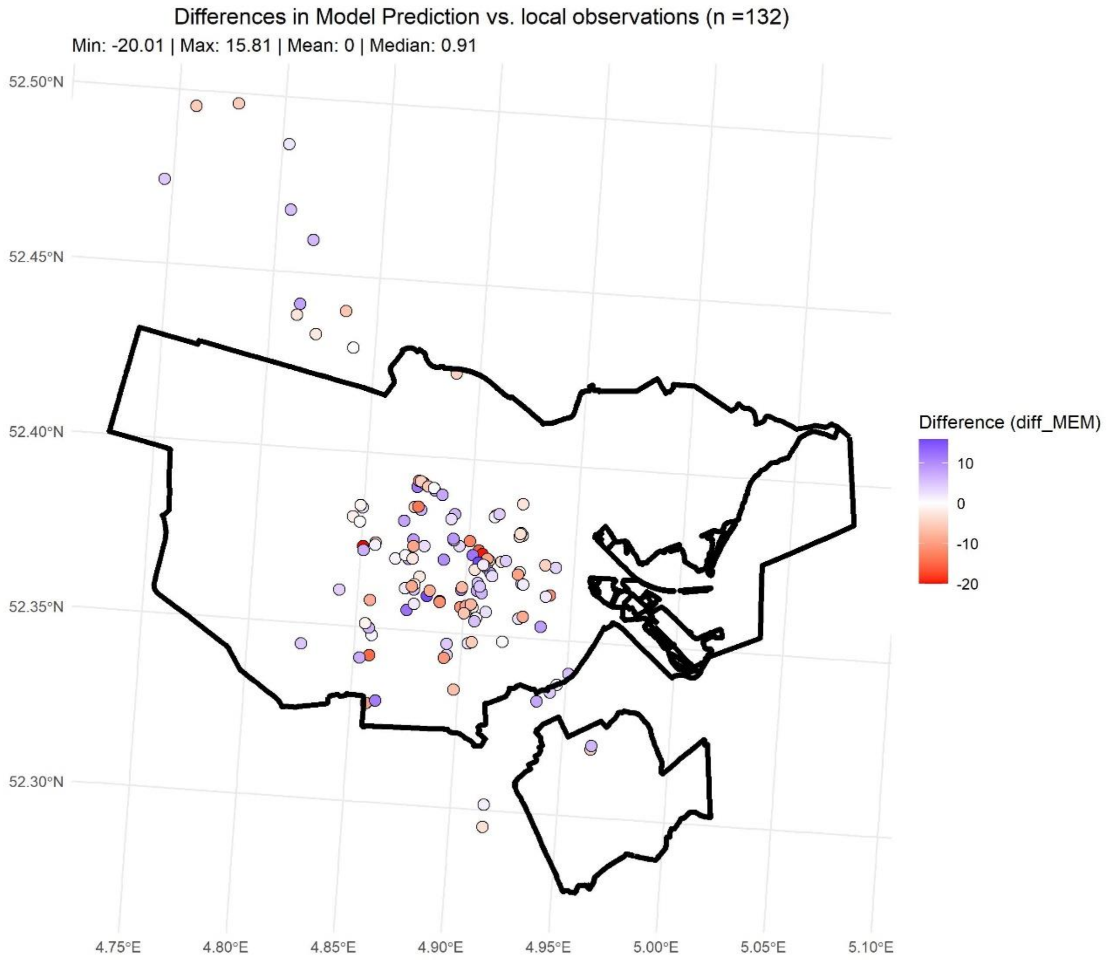


Figure S89 – Differences in model prediction vs ground measurement stations in Amsterdam universal kriging (local)

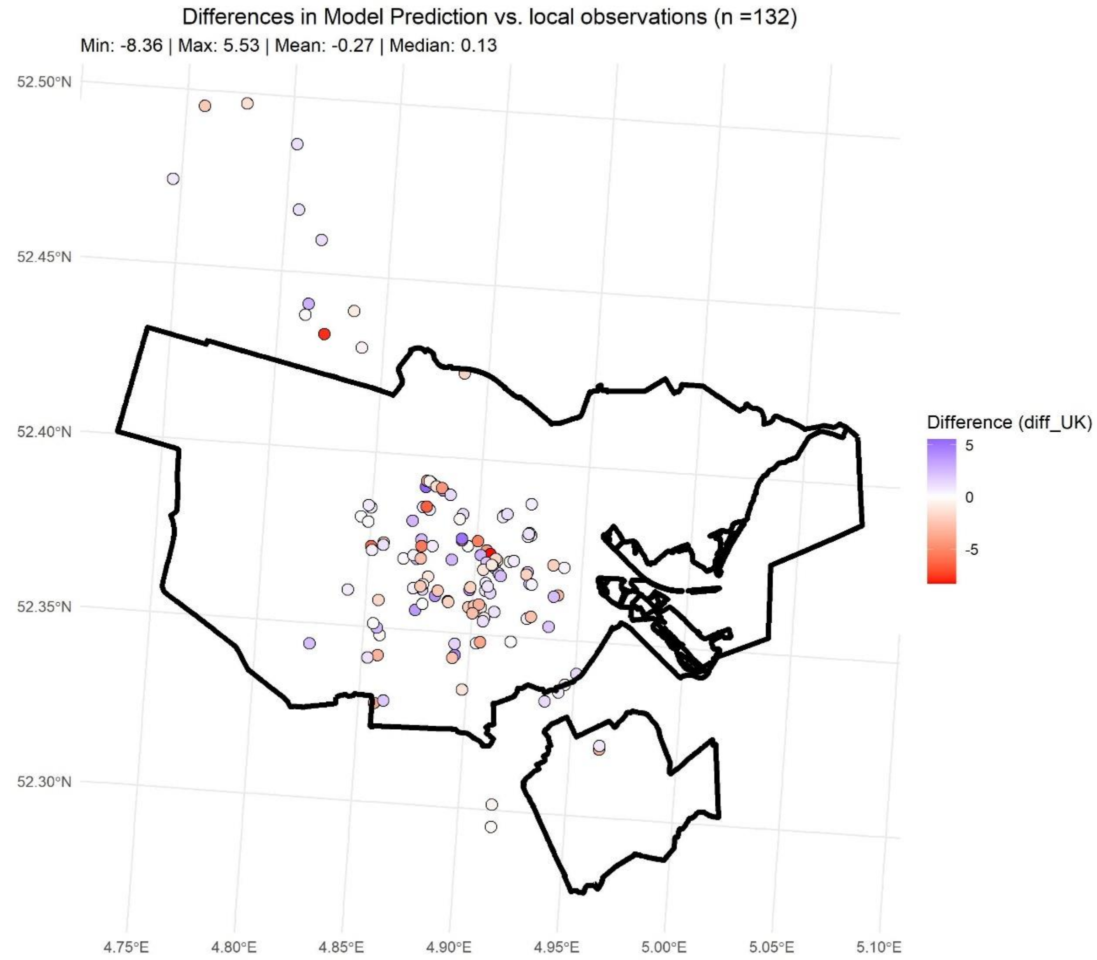


Figure S90 – Differences in model prediction vs ground measurement stations in Amsterdam universal kriging separating for spatial groups (local)

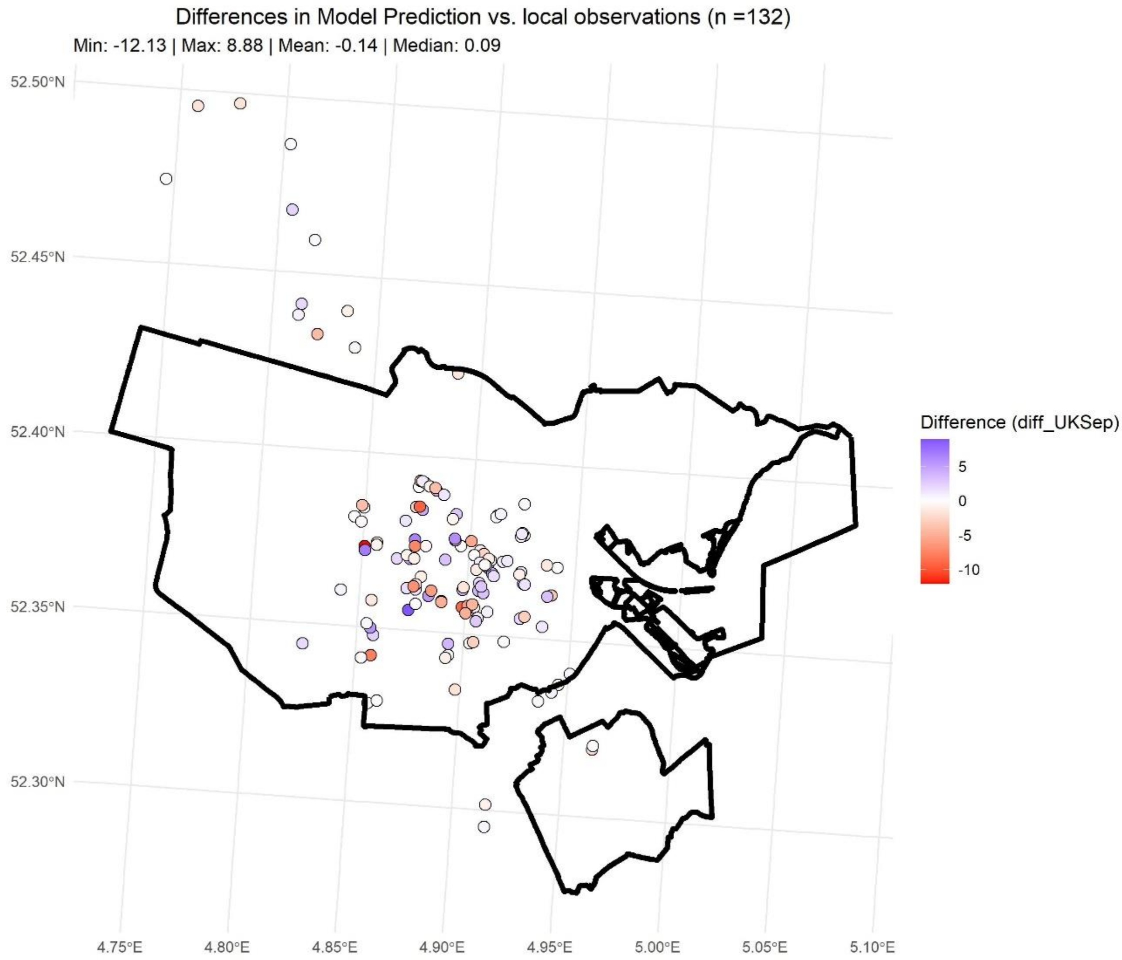


Figure S91 – Differences in model prediction vs ground measurement stations in Amsterdam ordinary kriging (local)

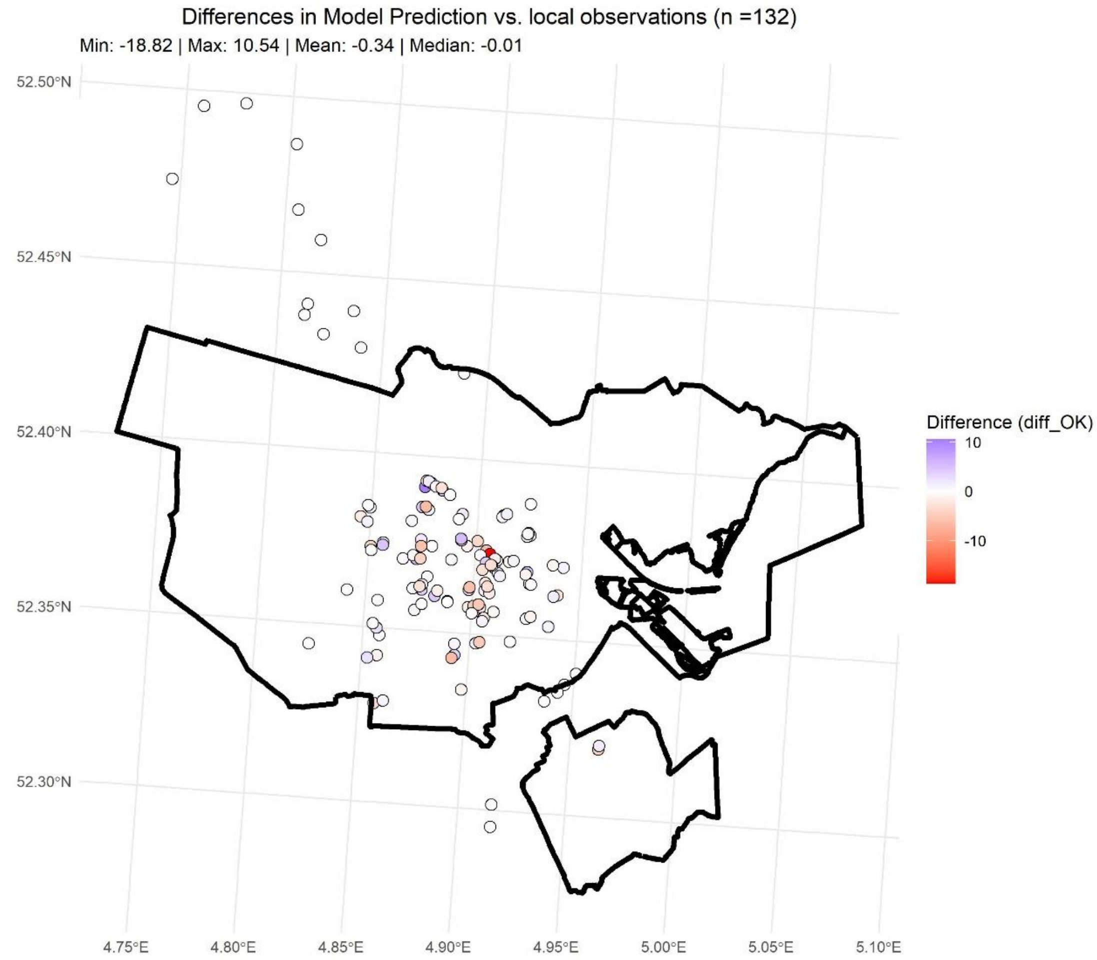


Table S1 initial dataset Lu et al. (2020)

Table 1: Description of variables used in the study.

Variable	Data	Unit	Location	Time	Source
road_class_1	Total length of highway in various buffer sizes	m (25m-, 50m-, 100m-, 300m-, 1000m-, 3000m-, 5000m-buffers)	The Netherlands, Germany	2017	OpenStreetMap
road_class_2	Total length of primary roads in various buffer sizes	m (25m-, 50m-, 100m-, 300m-, 1000m-, 3000m-, 5000m-buffers)	The Netherlands, Germany	2017	OpenStreetMap
road_class_3	Total length of local roads in various buffer sizes	m (25m-, 50m-, 100m-, 300m-, 1000m-, 3000m-, 5000m-buffers)	The Netherlands, Germany	2017	OpenStreetMap
industry	Area of industry in various buffer sizes	m^2 (25m-, 50m-, 100m-, 300m-, 1000m-, 3000m-, 5000m-buffers)	The Netherlands, Germany	2017	OpenStreetMap
wind	Monthly wind speed at 10 m altitude, for each month of the year	km/hr	The Netherlands, Germany	2017	ERA-Interim
temperature	Monthly temperature at 2 m altitude, for each month of the year	Celsius	The Netherlands, Germany	2017	ERA-Interim
OMI_mean_filt	Annual mean vertical column density (level 3 product)	mol/cm^2	The Netherlands, Germany	2017	Earthdata
TROPOMI	Vertical column density (level 3 product), mean of monthly average from 2018/02–2019/01	mol/cm^2	The Netherlands, Germany	2018-2019	Tropomi
RSP	Remote sensing product from SCIAMACHY, GOME-2, global GEOS-Chem, 2011	g/m^3	The Netherlands, Germany	2011	SCIAMACHY
population	Global Human Settlement Layer population grid	count (absolute) (1000m-, 3000m-, 5000m-buffers)	The Netherlands, Germany	2015	-

Table S2: Model performance metrics for different spatial characteristics for local dataset. (nfold=20 repeated random sampling validation),

Models		Urban			Suburban			Rural		
		R^2	RMSE	MAE	R^2	RMSE	MAE	R^2	RMSE	MAE
Random Forest	Mean	-0.07	7.603	6.606	0.373	6.436	4.821	-0.208	7.511	6.515
	SD	0.38	1.091	0.996	0.278	2.144	1.639	0.802	1.094	1.071
LightGBM	Mean	-0.134	7.802	6.627	0.337	6.448	5.25	-0.251	7.743	6.518
	SD	0.425	1.043	0.941	0.336	1.788	1.38	0.726	1.462	1.279
XGBoost	Mean	-0.398	8.495	6.824	0.347	6.341	4.542	-1.148	9.931	8.037
	SD	0.669	1.327	1.12	0.424	2.283	1.473	1.574	1.621	1.187
Ridge	Mean	-0.003	7.518	6.437	0.421	6.171	4.428	-0.276	7.62	6.685
	SD	0.257	1.327	1.214	0.229	1.859	1.276	0.942	1.078	0.996
Lasso	Mean	-0.03	7.564	6.466	0.407	6.121	4.392	-0.334	7.717	6.637
	SD	0.297	1.216	1.153	0.309	1.791	1.304	1.046	1.197	1.108

Table S3: Performance Metrics for Linear, LASSO and Ridge Regression Models (LOOCV)

Local model	Metric	Total	Urban	Suburban	Rural
Linear	RMSE	7.4121	7.8901	6.8003	7.3902
	R2	0.3073	0.1397	0.5087	0.1469
	MAE	5.9548	6.3598	5.3008	6.2019
Lasso	RMSE	7.4142	7.8774	6.8123	7.4080
	R2	0.3061	0.1400	0.5083	0.1460
	MAE	5.9603	6.3581	5.2977	6.2338
Ridge	RMSE	7.4420	7.8929	6.8174	7.4924
	R2	0.2968	0.1253	0.5197	0.1549
	MAE	6.0096	6.4493	5.2278	6.3877

Table S4 Variance Inflation Factor (VIF) for Features (global dataset)

Feature	**VIF**
nightlight_450	3.775581
nightlight_3150	4.002998
population_1000	15.366872
population_3000	18.274317
road_class_2_25	1.368278
road_class_3_3000	6.047955
road_class_3_300	3.052999
trop_mean_filt_2019	1.552954
BldDen100	2.993308
NDVI	1.912893
trafBuf25	10.976549
trafBuf50	10.841305

Table S5: Variance Inflation Factor (VIF) for Features (local dataset)

Feature	VIF
nightlight_450	2.156364
nightlight_4950	25.911217
population_3000	20.571758
road_class_1_5000	2.926517
road_class_2_1000	1.538339
road_class_2_5000	4.499247
road_class_3_100	2.116426
road_class_3_300	2.265685
trafBuf50	1.168518

Code S1

```
df['Urban']= np.where(df['population_1000'].gt(df['population_1000'].quantile(0.75)) &
(df['road_class_1_100'].gt(0) | df['road_class_2_100'].gt(0) |
df['road_class_3_100'].gt(df['road_class_3_100'].quantile(0.75))), 1, 0)

df['Suburban'] = np.where(df['population_1000'].lt(df['population_1000'].quantile(0.75)) &
(df['road_class_1_100'].gt(0) | df['road_class_2_100'].gt(0)  |
df['road_class_3_100'].gt(df['road_class_3_100'].quantile(0.75))), 1, 0)

df['Rural'] = np.where(df['Urban'] | df['LowPop'] = = 1, 0, 1)
```