S1. Methods

S1.1 $PM_{2.5}$ sampling

S1.1 Measurement instrument

Table S1 shows the descriptive statistics of $PM_{2.5}$ concentration from laser air quality monitor and the national monitoring instrument. The mean values of them were 72.99 and 71.89 μg m^{-3}, respectively. Their standard deviations were rather close (16.45 vs. 15.28). The laser air quality monitors have larger maximum value and smaller minimum value than national monitoring instrument.

S2. Results and Supplemental Analyses

S2.1 Model fitting of LUR

Table S2 shows the predictor variables for LUR models. All of the predictors listed in this table reveals statistical significance with very small p-values (<0.05). Clearly, models of two periods shared common predictor variables of the percentage of high-density residential area (H-Resi), other built-up area (O-Built), low-density forest (L-For), stacked substance area (Sta), natural bare surfaces (Bare), and crush stampede yard area (Yard). However, the optimal buffering sizes for some of these predictors were different. For example, the optimal buffering size for H-Resi of light-polluted period was 200m, while that of heavy-polluted period was 50m. Meteorological factors of wind speed, atmospheric pressure, relative humidity, and temperature were important predictors for LUR models.

Table S3 summarizes the model fitting results for LUR models. The fitting adjusted R^2 ranged from 0.38 (12:00) to 0.61 (8:00) for Period 1, and from 0.25 (17:00) to 0.36 (16:00) for Period 2. The AIC values for two periods were found to be lowest at 8:00 (1400.64) and 16:00 (907.38) respectively. And the highest and lowest RMSE were 11.79 and 10.39 for Period 1, and 17.53 and 11.51 for Period 2.

S2.2 Model performance for OK, LUR and UK

Average and standard deviation of RMSE and MRE between the observed and predicted concentrations of $PM_{2.5}$ in hold-out validation presented in Supporting Information (Table S4–S5) further demonstrated the better performance yet larger variation of these three methods with larger training data sets in $PM_{2.5}$ concentration estimation. The average RMSE and MRE of OK and UK were close and significantly smaller than LUR. Meanwhile, those of OK were generally smaller than UK in Period 1 while cases in Period 2 were the opposite.

For period 1, the RMSE (Mean±SD) of OK, LUR and UK ranged from (6.26±1.45) (10:00, n=184) to (12.54±0.83) (12:00, n=41), (11.25±1.59) (11:00, n=187) to (24.05±9.97) (8:00, n=36), and (8.65±1.07) (11:00, n=166) to (12.29±0.52) (8:00, n=36), respectively. Correspondingly, the MRE of OK, LUR and UK increased from (6.81±1.55)% (10:00, n=184) to (13.11±1.41)% (8:00, n=36), (13±2.26)% (12:00,

n=186) to (24.65 ±6.31)% (8:00, n=36), and (9.66 ±1.53)% (11:00, n=166) to (12.79 ±0.94)% (8:00, n=36), respectively.

For period 2, the lowest and highest values of RMSE were (9.82 ±3.27) (14:00, n=94) and (21.12 ±1.88) (18:00, n=32), (13.32 ±2.86) (16:00, n=103) and (44.59 ±10.55) (18:00, n=32), (8.70 ±2.20) (14:00, n=94) and (18.84 ±1.56) (18:00, n=32). The associated MRE were (4.36 ±1.25)% (14:00, n=94) and (8.80 ±1.19)% (18:00, n=32), (6.62 ±2.17)% (14:00, n=106) and (18.93 ±4.26)% (18:00, n=32), (4.11 ±1.03)% (14:00, n=94) and (8.42 ±0.90)% (8:00, n=36) (18:00, n=32).

Table S1. Descriptive statistics of $PM_{2.5}$ concentration ($\mu g/m^3$) for two kinds of instruments

	Mean	Standard deviation	Minimum	Maximum
SDL	72.99	16.45	37.33	120.67
NAT	71.89	15.28	42	103

SDL: laser air quality monitors (SDL307) used in this study.

NAT: national monitoring instrument.

Table S2. Predictor variables for LUR models

Time	N	Predictor variables
8:00	179	H-Resi(200) + L-Resi(500) + L-For(500) + O-Built(1000) + Bare(1000) + WS + PS + RH
9:00	207	H-Resi(200) + L-Resi(500) + L-For(500) + Sta(1000) + Bare(1000) + Yard(500) + PS + Temp
10:00	205	H-Resi(300) + L-Resi(200) + L-For(500) + Cons(300) + Bare(300) + Yard(500) + PS + Temp
11:00	208	H-Resi(200) + L-Resi(500) + O-Built(1000) + Cons(1000) + Yard(500) + WS + RH + Temp
12:00	207	L-Resi(500) + O-Built(1000) + Agri(200) + Indus(1000) + Yard(500) + Dust(1000) + PS + RH
14:00	118	H-Resi(50) + Green(1000) + L-For(300) + Sta(1000) + Bare(200) + Yard(1000) + PS + Temp
15:00	117	Green(1000) + Sta(1000) + Cons(1000) + Bare(100) + Dust(1000) + PS + RH
16:00	114	Green(1000) + Road(1000) + Sta(500) + Bare(1000) + WS + RH + Temp
17:00	116	H-Resi(100) + L-For(300) + Sta(1000) + Dust(1000) + WS
18:00	105	H-For(1000) + Cons(50) + Bare(1000) + WS + Temp

Significant with $p < 0.05$; N is the total amount of the records in compared pairs; H-Resi = high-density residential area; L-Resi = low-density residential area; Green = urban green land; O-Built = other built-up area; H-For = high-density forest; L-For = low-density forest, Agri = agricultural land; Sta = stacked substance; Cons = construction site; Bare = natural bare surfaces; Yard = crush stampede yard; Dust = Ground dust surface; Road = Road density; Indus = POI density including industrial and mining enterprises; (*x*) represent area proportion/ density within the *x* m buffering radius. WS = Average wind speed Atmospheric pressure; RH = Relative humidity; Temp = Temperature.

Table S3. Model fitting results for LUR models

	Adjusted R^2	AIC	RMSE
8:00	0.61	1400.64	11.44
9:00	0.50	1628.61	11.79
10:00	0.41	1610.10	11.69
11:00	0.42	1583.87	10.39
12:00	0.38	1607.56	11.20
14:00	0.31	931.46	11.51
15:00	0.32	951.70	12.97
16:00	0.36	907.38	11.86
17:00	0.25	985.91	15.96
18:00	0.30	913.41	17.53

Table S4. Mean and standard deviation (SD) of root-mean-square-errors (RMSE) between observed and estimated $PM_{2.5}$ concentration

		8:00		9:00		10:00		11:00		12:00		14:00		15:00		16:00		17:00		18:00	
%		Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
20	OK	12.29	1.11	11.30	0.94	10.42	0.81	9.88	0.65	12.54	0.83										
	LUR	24.05	9.97	19.11	4.75	20.33	5.35	18.82	10.31	19.40	4.77										
	UK	11.44	0.78	11.03	0.50	10.67	0.70	9.76	0.56	11.13	0.45										
30	OK	11.75	0.79	10.60	0.83	9.78	0.82	9.36	0.66	12.06	0.88	13.18	2.11	14.30	2.18	12.92	1.32	17.15	2.35	21.12	1.88
	LUR	17.03	3.39	15.22	4.39	15.57	2.30	14.45	5.32	15.37	2.90	29.92	17.26	26.61	6.71	27.18	9.74	35.27	22.10	44.59	10.55
	UK	12.29	0.52	10.75	0.57	10.26	0.51	9.68	0.51	11.07	0.48	11.42	1.31	13.09	1.14	13.02	1.04	16.16	1.87	18.84	1.56
40	OK	11.25	1.05	10.10	1.01	9.19	0.93	8.72	0.55	11.74	1.19	12.82	2.36	13.13	2.40	11.71	1.61	16.97	2.97	20.11	1.71
	LUR	14.88	1.82	13.33	1.14	14.97	2.64	12.65	1.07	14.01	1.50	22.09	9.48	20.95	3.97	19.74	5.31	23.69	8.53	31.99	6.83
	UK	12.11	0.70	10.48	0.61	10.16	0.66	9.22	0.51	11.06	0.53	11.26	1.42	12.32	1.58	12.00	1.17	15.70	2.24	18.30	1.69
50	OK	10.84	1.23	9.69	1.22	8.72	0.95	8.64	0.72	11.52	1.18	11.33	2.47	12.55	2.43	11.38	1.91	16.02	2.93	18.87	2.25
	LUR	14.47	2.58	12.94	1.23	13.73	2.16	12.20	1.01	13.23	1.01	17.46	2.98	19.10	3.12	15.90	3.06	21.76	4.90	27.56	5.63
	UK	10.66	0.68	10.48	0.78	9.95	0.76	9.32	0.60	11.03	0.66	10.54	1.66	11.87	1.58	11.70	1.47	15.22	2.30	16.74	2.13
60	OK	10.52	1.54	9.12	1.31	7.91	1.16	8.02	0.79	11.13	1.41	11.60	2.44	14.29	18.26	11.11	1.97	15.01	3.26	17.56	2.74
	LUR	13.70	1.70	12.55	1.22	12.91	1.14	11.61	1.08	12.87	1.10	19.95	13.42	17.95	2.81	15.48	2.56	19.50	3.56	23.98	3.98
	UK	10.46	0.86	10.14	0.83	9.59	0.82	8.96	0.65	10.85	0.76	10.32	1.80	11.28	1.71	11.44	1.62	14.20	2.86	16.19	2.70
70	OK	10.52	1.74	8.36	1.28	7.47	1.19	7.69	0.95	10.53	1.64	10.93	3.00	11.26	3.09	10.55	2.12	14.78	4.41	17.42	3.55
	LUR	13.16	1.34	11.98	1.22	13.61	1.22	11.66	1.18	12.33	1.19	16.04	3.15	16.27	2.49	14.22	2.04	18.57	4.03	22.89	2.89
	UK	10.48	0.99	9.82	0.94	9.61	1.17	9.01	0.77	10.54	1.03	9.88	2.10	10.30	2.06	10.98	1.73	13.82	4.19	15.53	3.33
80	OK	9.53	2.03	8.25	1.63	7.03	1.43	7.26	1.23	10.66	2.29	9.82	3.27	11.31	3.53	10.35	2.36	13.37	5.47	16.12	4.26
	LUR	12.68	1.48	11.94	1.44	12.77	1.51	11.29	1.28	12.33	1.47	15.93	3.55	16.41	2.61	13.74	1.75	18.53	5.35	22.79	3.99
	UK	10.12	1.23	9.69	1.38	9.33	1.40	8.65	1.07	10.58	1.36	8.81	1.88	11.04	1.88	11.20	1.93	12.46	4.76	15.11	3.50
90	OK	9.65	3.87	7.73	2.12	6.26	1.45	7.30	1.69	10.16	2.51	10.16	3.93	12.49	4.09	10.38	3.76	14.21	6.04	15.59	7.57
	LUR	13.01	2.09	11.81	2.00	12.35	1.92	11.25	1.59	12.12	1.90	15.60	5.67	15.93	3.82	13.32	2.86	17.41	6.61	22.09	6.49
	UK	10.26	1.87	9.49	2.02	8.90	1.77	8.70	1.41	10.54	2.06	8.70	2.20	10.00	2.73	11.35	2.13	12.29	6.13	14.24	5.81

Table S5. Mean and standard deviation (SD) of mean relative error (MRE, %) between observed and estimated $PM_{2.5}$ concentration

		8:00		9:00		10:00		11:00		12:00		14:00		15:00		16:00		17:00		18:00	
%		Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
20	OK	13.11	1.41	12.37	1.20	11.50	1.10	10.99	0.86	12.09	1.09										
	LUR	24.65	6.31	20.37	3.26	21.30	3.20	18.81	3.45	19.37	3.49										
	UK	12.79	0.94	12.48	0.81	12.38	0.92	10.80	0.70	11.54	0.63										
30	OK	12.50	1.00	11.48	1.01	10.58	0.93	10.32	0.86	11.56	0.98	5.56	0.71	5.98	0.71	5.65	0.50	6.71	0.93	8.80	1.19
	LUR	18.98	2.86	16.67	1.82	17.35	1.79	15.76	1.74	15.67	1.49	11.82	4.58	11.80	2.88	12.02	3.39	13.16	4.40	18.93	4.26
	UK	12.24	0.81	12.05	0.86	11.80	0.80	10.68	0.74	11.54	0.73	5.13	0.49	6.02	0.52	6.35	0.64	6.76	0.88	8.42	0.90
40	OK	11.80	1.17	10.74	1.11	9.91	1.12	9.41	0.71	11.09	0.96	5.39	0.82	5.48	0.74	5.16	0.64	6.64	1.17	8.33	0.99
	LUR	16.85	1.84	15.23	1.08	16.74	1.59	14.37	0.91	14.67	1.55	8.84	2.07	9.34	1.82	9.03	1.41	10.16	2.38	13.85	2.63
	UK	11.96	0.90	11.71	0.89	11.75	1.14	10.22	0.68	11.56	0.69	5.06	0.52	5.68	0.64	5.81	0.72	6.56	0.88	8.06	0.89
50	OK	11.16	1.08	10.25	1.16	9.33	1.15	9.23	0.84	10.60	0.92	4.77	0.87	5.29	0.91	4.95	0.71	6.12	0.94	7.59	0.93
	LUR	16.23	1.99	15.01	1.35	15.90	1.46	13.98	1.19	13.94	1.08	7.35	1.09	8.49	1.27	7.78	1.11	9.11	1.46	11.83	2.28
	UK	11.93	0.79	11.83	0.97	11.46	1.24	10.35	0.93	11.48	0.89	4.78	0.62	5.43	0.60	5.61	0.79	6.15	0.89	7.08	1.07
60	OK	10.95	1.43	9.76	1.32	8.47	1.14	8.55	0.96	10.46	1.14	4.90	0.87	6.48	9.81	4.90	0.81	5.97	0.96	7.20	1.19
	LUR	15.57	1.79	14.62	1.43	15.19	1.64	13.45	1.43	13.68	1.28	7.61	2.46	8.27	1.21	7.55	0.99	8.57	1.46	10.75	1.79
	UK	11.73	1.11	11.45	0.99	10.86	1.26	10.07	0.96	11.31	1.01	4.60	0.62	5.18	0.66	5.46	0.91	5.95	0.84	6.96	1.24
70	OK	10.52	1.51	8.89	1.26	8.14	1.38	8.16	1.06	9.82	1.14	4.66	1.07	4.78	1.10	4.65	0.98	5.89	1.36	7.16	1.17
	LUR	14.96	1.57	14.09	1.63	15.53	1.89	13.45	1.60	13.11	1.35	6.81	1.10	7.44	1.04	7.15	0.94	7.92	1.24	10.27	1.24
	UK	11.55	1.09	11.12	1.24	10.97	1.71	10.01	1.12	10.93	1.02	4.49	0.78	4.81	0.84	5.26	0.96	5.65	1.15	6.51	1.25
80	OK	10.02	1.83	8.67	1.64	7.55	1.48	7.77	1.27	9.79	1.24	4.36	1.25	4.80	1.48	4.50	0.91	5.52	1.83	6.64	1.61
	LUR	14.81	2.00	14.23	1.79	15.21	2.31	13.22	2.10	13.08	1.69	6.64	1.51	7.56	1.14	6.87	0.88	8.15	1.81	10.17	1.97
	UK	11.56	1.50	10.94	1.52	10.52	1.96	9.66	1.53	10.93	1.34	4.13	0.75	5.08	0.83	5.31	1.00	5.35	1.39	6.35	1.54
90	OK	9.91	3.02	8.20	2.09	6.81	1.55	7.54	1.68	9.79	1.90	4.57	1.59	5.34	1.73	4.80	1.59	6.22	2.05	6.24	2.34
	LUR	15.00	2.89	14.18	2.89	14.49	2.98	13.10	2.67	13.00	2.26	6.62	2.17	7.14	1.47	6.97	1.59	7.95	2.48	10.19	2.56
	UK	11.66	2.19	10.80	2.31	9.79	2.25	9.73	1.88	10.95	1.97	4.11	1.03	4.62	1.17	5.61	1.23	5.52	1.83	5.77	1.83