Supporting Information

Spatial and Spatiotemporal Variability of Regional Background Ultrafine Particle Concentrations in The Netherlands

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# Contributed equally

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This file includes 22 pages containing:

Supplement A: Data Correction

Differences between devices were assessed by co-locating all 5 devices for 1-2 days before and after every two-week measurement. All devices were placed in a laboratory setting with all inlets connected to the same airstream. Concentrations were fluctuated between 0 and 100.000 particles/cm^3 throughout the colocation measurement. We corrected each measurement campaign based on calculated median ratios between the reference device and other devices of each co-located comparison. All individual ratios are shown in table A.1 with average medians ranging from 0,99 to 1,19.

Table A.1: Ratios for all individual co-location measurements.

Date	DM1:DM5	DM2:DM5	DM3:DM5	DM4:DM5
06/09/2016	1,20	1,23	1,20	0,80
23/09/2016	1,16	1,16	1,07	0,85
26/09/2016	/	1,19	1,15	0,85
13/10/2016	1,18	1,25	1,15	0,93
14/10/2016	1,10	1,23	1,12	0,96
07/11/2016	1,03	1,20	0,99	0,98
25/11/2016	0,94	1,13	1,01	0,88
29/11/2016	1,10	1,22	1,00	1,02
16/12/2016	1,22	1,20	0,99	1,01
21/12/2016	1,24	1,16	1,02	1,02
20/01/2017	1,14	1,22	0,94	1,01
23/01/2017	1,11	1,22	0,96	1,06
09/02/2017	1,12	1,23	0,98	0,98
10/02/2017	0,98	1,23	0,92	0,98
02/03/2017	1,13	1,17	1,04	1,05
23/03/2017	1,16	1,30	1,03	1,11
27/03/2017	1,18	1,27	1,08	1,12
28/03/2017	1,17	1,22	1,07	1,02
13/04/2017	1,19	1,26	1,13	1,05
10/07/2017	0,88	0,88	0,98	0,87
01/08/2017	/	1,11	1,16	1,01
04/09/2017	1,17	1,20	1,47	1,20
03/10/2017	1,08	0,93	1,07	0,98
23/10/2017	1,25	1,18	1,09	1,01
14/11/2017	1,38	1,32	1,09	1,07
Total	**1,14**	**1,19**	**1,07**	**0,99**

Temporal Correction

We used a reference site in the middle of the country (Bunnik) to correct for temporal variation. This reference site had the same restrictions as the sampling locations regarding potential local sources. Reference equipment was mounted into a bike-trailer and was placed at ground level in the back garden of a relatively secluded house.

The difference method we used is as follows:

1. Calculate the overall average UFP concentration on the reference site for the full study period.
2. Calculate for the reference site the difference between the overall average UFP concentration and each of the 15 averages of the measurement periods.
3. Add this difference to the time-corresponding individual site average concentrations on all sampling locations.

Results

Table A.2: Individual 2-week average concentrations per site

Site	Round 1	Round 2	Round 3	Total	Region
Bedum	5569	3964	/	4767	North
Budel	5914	5257	/	5586	South
Burgerbrug	4800	4966	3637	4468	North
De Zilk	7908	5449	/	6678	West
Donkerbroek	/	4600	3028	3814	North
Grijpskerke	6215	5171	3854	5080	South
Huybergen	7658	7180	5355	6731	South
Lemelerveld	6065	4749	/	5407	East
Mantgum	/	4330	/	4330	North
Maurik	8202	5565	4352	6040	East
Monnickendam	6593	5680	5064	5779	West
Odiliapeel	7723	6751	5252	6575	South
Oosterhesselen	4717	4117	/	4417	North
Rockanje	6477	5480	5121	5693	West
s Gravenmoer	7185	5530	5707	6141	West
Schimmert	/	5228	5390	5309	South
Stolwijk	6561	5388	/	5975	West
Swifterband	/	3920	3899	3909	East
Voorthuizen	7584	7599	6056	7080	East
Zelhem	6153	5388	4300	5280	East

Concentrations in particles/cm^3

Supplement B: Spatial Analysis

Geostatistics

Geostatistics can be applied to create maps of a spatially sampled property, a so-called regionalized variable (Cressie, 1991). The most widely used geostatistical technique is kriging, which provides estimates with minimum error variance of the regionalized variable at unsampled locations (Davis, 2002). In order to apply kriging, first the spatial correlation between observations of the regionalized variable needs to be quantified. The concept used in geostatistics is that observations of a regionalized variable at locations that are close to each other are more similar than observations that are further apart. This spatial correlation can be quantified with the semi-variogram, which is a model of the semi-variance between spatial observations at a certain distance apart (the lag distance). In a 2-dimensional plane the semi-variance between pairs of observations (z_i , z_{i+h}) which are separated by a distance falling in a distance class h is calculated as (Davis, 2002):

$$\gamma_h = \frac{1}{2}\frac{\sum_{i=1}^{Nh}\left(z_i - z_{i+h}\right)^2}{Nh} \qquad \text{S1}$$

where γ_h is the semi-variance at lag distance class h, and Nh is the total number of observation pairs in lag distance class h. By fitting an equation through the calculated semi-variogram a model of the semi-variance is obtained. This model is characterized by three parameters: the nugget, the range and the sill. The nugget is the semi-variance at distance zero. In theory this semi-variance is zero, but due to measurement errors or spatial variation at small lag distances that are not included in the observations there can be a non-zero semi-variance. The range of the semi-variogram is the spatial distance over which spatial correlation exists. Beyond the range any two point are no longer related to each other. The sill is the semi-variance that is reached at the range. It is approximately equal to the variance of all observations.

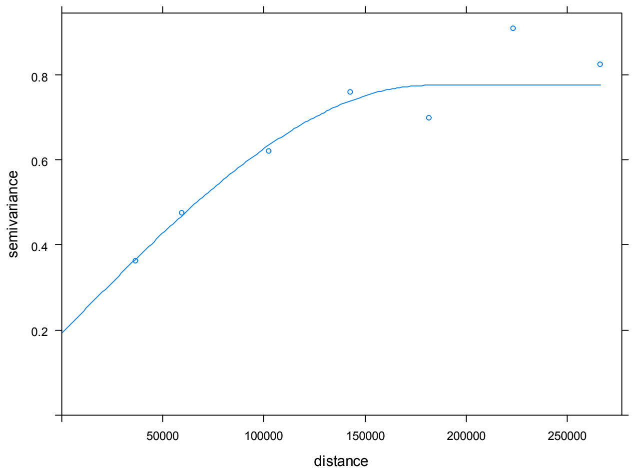
Webster and Oliver (1992) recommended that semi-variograms should be computed using at least 50 observations, but preferably 100 or more observations of the regionalized variable should be used. In practice, often less observations of a regionalized variable are available due to varying problems, such as lack of instruments, time and financial resources. To overcome this problem of an insufficient number of observations, Sterk and Stein (1997) developed a space-time procedure to compute stable semi-variograms. In their procedure multiple events with observations of the regionalized variable taken at different moments in time are used to calculate a standardized semi-variogram. This standardized semi-variogram is subsequently converted to an event-based semi-variogram, by re-calculating the nugget and sill, which are proportional to the variance of the observations for the event. The range of the semi-variograms is assumed equal for all events, assuming that the spatial correlation structure is unchanged. The Sterk and Stein (1997) space-time procedure was used in this study to calculate separate semi-variograms for each of the three UFP measurement times.

In the first step the average concentrations per location and measurement period were standardized by withdrawing the mean of all 49 observations from each individual observation, and by dividing with the standard deviation of all 49 observations. The semi-variogram was calculated using equation S1, but to avoid that calculations between the observations at the three measurement periods were used, the coordinates were shifted 1000 km per time interval. The maximum distance of pairs of observations was set at 500 km, which guaranteed that only pairs of observations within a certain time period were used. A spherical model was fitted through the computed standardized semi-variogram values (figure S1). The spherical model is defined as (Davis, 2002):

$$\gamma_h = \gamma_0 + (\sigma_0^2 - \gamma_0)\left(\frac{3h}{2a} - \frac{h^3}{2a^3}\right) \quad for\ h < a$$
$$\gamma_h = \sigma_0^2 \quad for\ h \geq a \qquad \text{S2}$$

where γ_0 is the nugget, σ_0^2 is the sill and a is the range of the semi-variogram.

Figure B.1: Fitted semi-variogram model through calculated semi-variance values of standardized UFP measurements in the Netherlands.



The fitted spherical model has a range of 180.9 km, which indicates the presence of spatial correlation up until 181 km. The nugget and sill values are respectively 0.193 and 0.582. The standardized semi-variogram was converted into three semi-variograms, one for each measurement time. This was done by multiplying the nugget and sill of the standardized semi-variogram with the variance of the UFP values per measurement time, while the range was kept constant. The parameters of the three spherical semi-variogram models are provided in Table B.1.

Table B.1: Fitted spherical semi-variogram models for three UFP measurement periods in the Netherlands.

Observation time	Spherical semi-variogram		
	Nugget (particles $cm^{-3})^2$	Sill (particles $cm^{-3})^2$	Range (m)
Round 1	217606	657807	180893
Round 2	184765	558532	180893
Round 3	151692	458553	180893

The three semi-variogram models and the original UFP concentrations of every measurement period were used to make three separate kriging maps, applying ordinary kriging (Davis, 2002):

$$\hat{Z}(x_o) = m + \sum_{i=1}^{k} \lambda_i [Z(x_i) - m] \qquad \text{S3}$$

where $\hat{Z}(x_o)$ is the kriging estimate at position x_o, m is the mean of the observations, λ_i are the kriging weights, and $Z(x_i)$ the UFP observations. The kriging weights are calculated using the semi-variogram model, and determine the weight of each observation for the kriging estimate. A raster was used with a cell size of 500 x 500m. The three produced kriging maps (Figure B.2) were then averaged in ArcGIS pro, to create one final mean kriging map of the Netherlands. Kriging error maps (i.e. the variance of the kriging estimates) for each measurement period were made to estimate the uncertainty of the kriging prediction over the area (Figure B.2). In sensitivity analysis we performed a leave-one-out-cross-validation to further analyse potential error in the map. While deletion of a site would affect the standardisation procedure and development of the variogram, we assumed these to be unchanged. So, we created 20 kriging maps, each based on 19 sites and the same variogram. Predictions made for the remaining site was then compared to the same site in the kriging map with all 20 sites included. Results are shown in figure B.3. All geostatistical analyses were done using the gstat package in R.

References:

Cressie, N.A.C. 1991. Statistics for Spatial Data. John Wiley & Sons, New York

Davis, J.C. 2002. Statistics and Data Analysis in Geology, 3rd Edition. John Wiley & Sons, New York.

Sterk, G. and A. Stein. 1997. Mapping wind-blown mass transport by modeling variability in space and time. Soil Science Society of America Journal 61: 232-239.

Webster R. and M.A. Oliver 1992. Sample adequately to estimate variograms of soil properties. Journal of Soil Science 43: 177-192.

Figure B.2. Top: Maps of UFP regional background concentrations (particles/cm³) in the Netherlands produced by kriging. Dates of measurement periods are June 8 – Nov 23 2016 (Round 1), Nov 30 2016 – Mar 22 2017 (Round 2) and Mar 29 - Nov 8 2017 (Round 3). Bottom: Maps of the kriging variance for the corresponding periods, providing a measure of the uncertainty of the kriging estimates in the top row figures. Values are the root of the standard error.

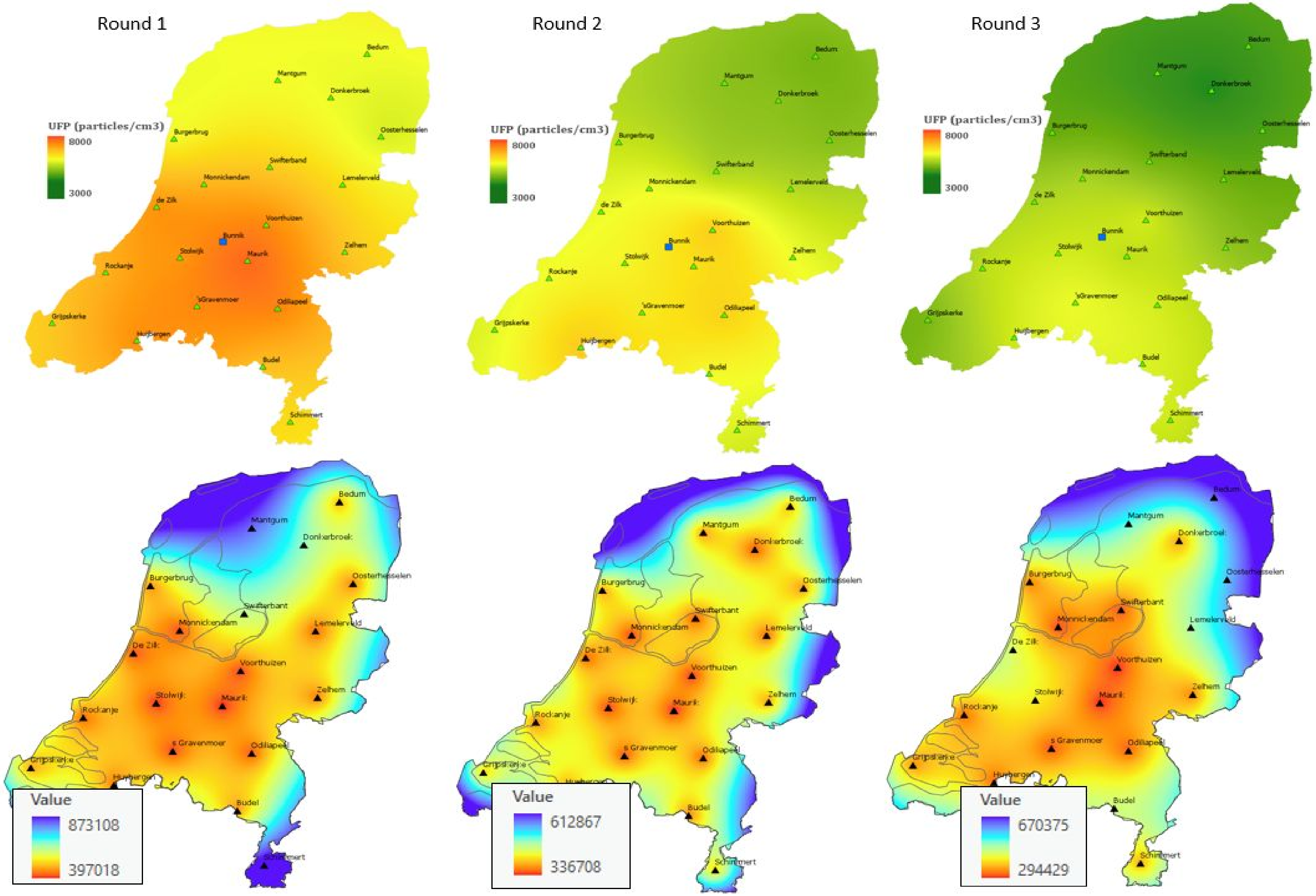


Figure B.3: Scatterplot of UFP predictions generated by leave-one-out-cross-validation and kriged values with the full data set.

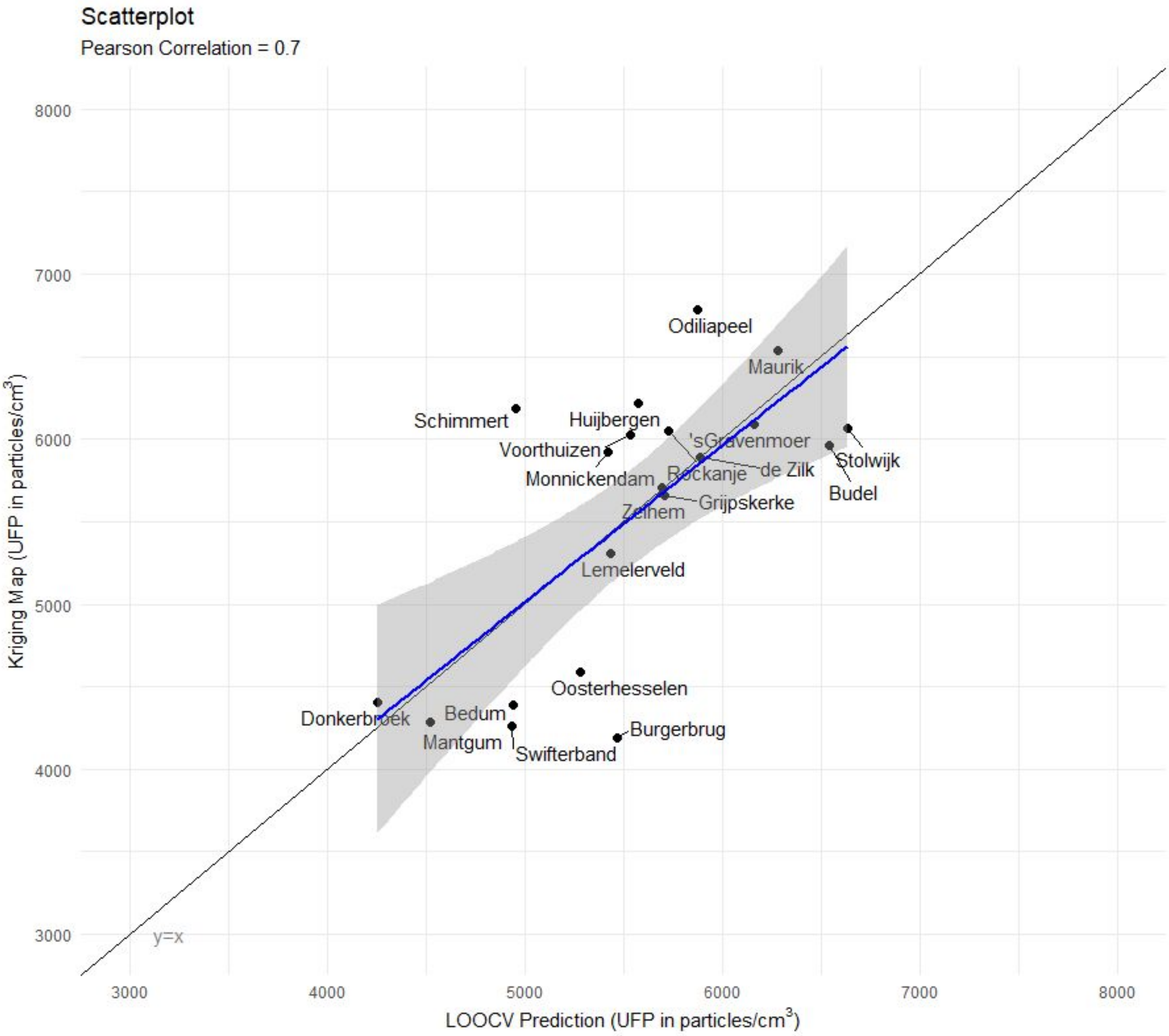


Figure B.4: Scatterplot of UFP predictions generated by leave-one-out-cross-validation and measured concentrations.

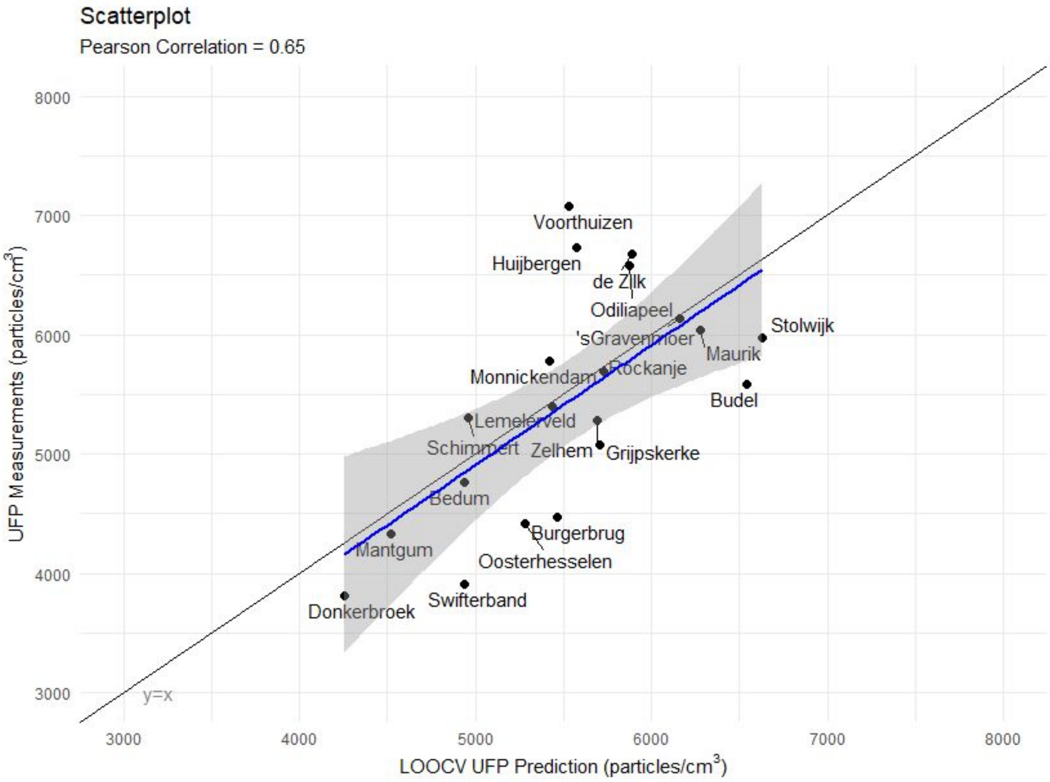
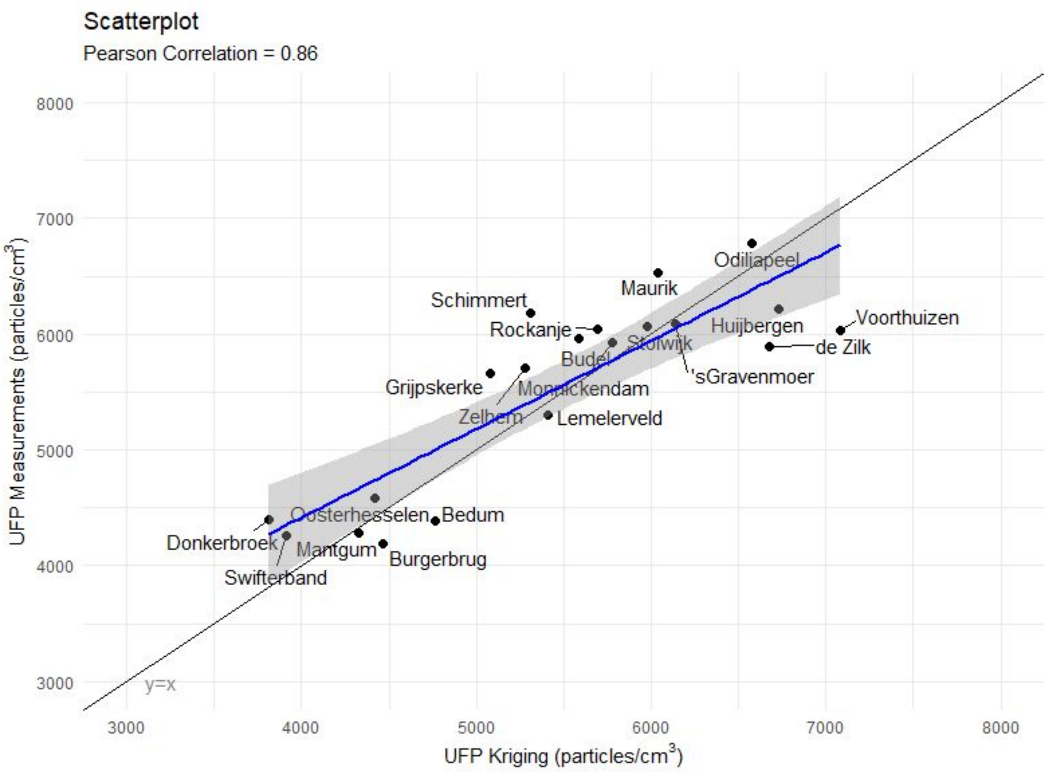


Figure B.5: Scatterplot of UFP concentrations generated by kriging with the measured concentrations.



Supplement C: Spatiotemporal analysis

Table C.1: Distance, Pearson and Spearman correlations, maximum cross-correlations and proportion of variance explained (PVE), between the sampling locations and the reference site (Bunnik).

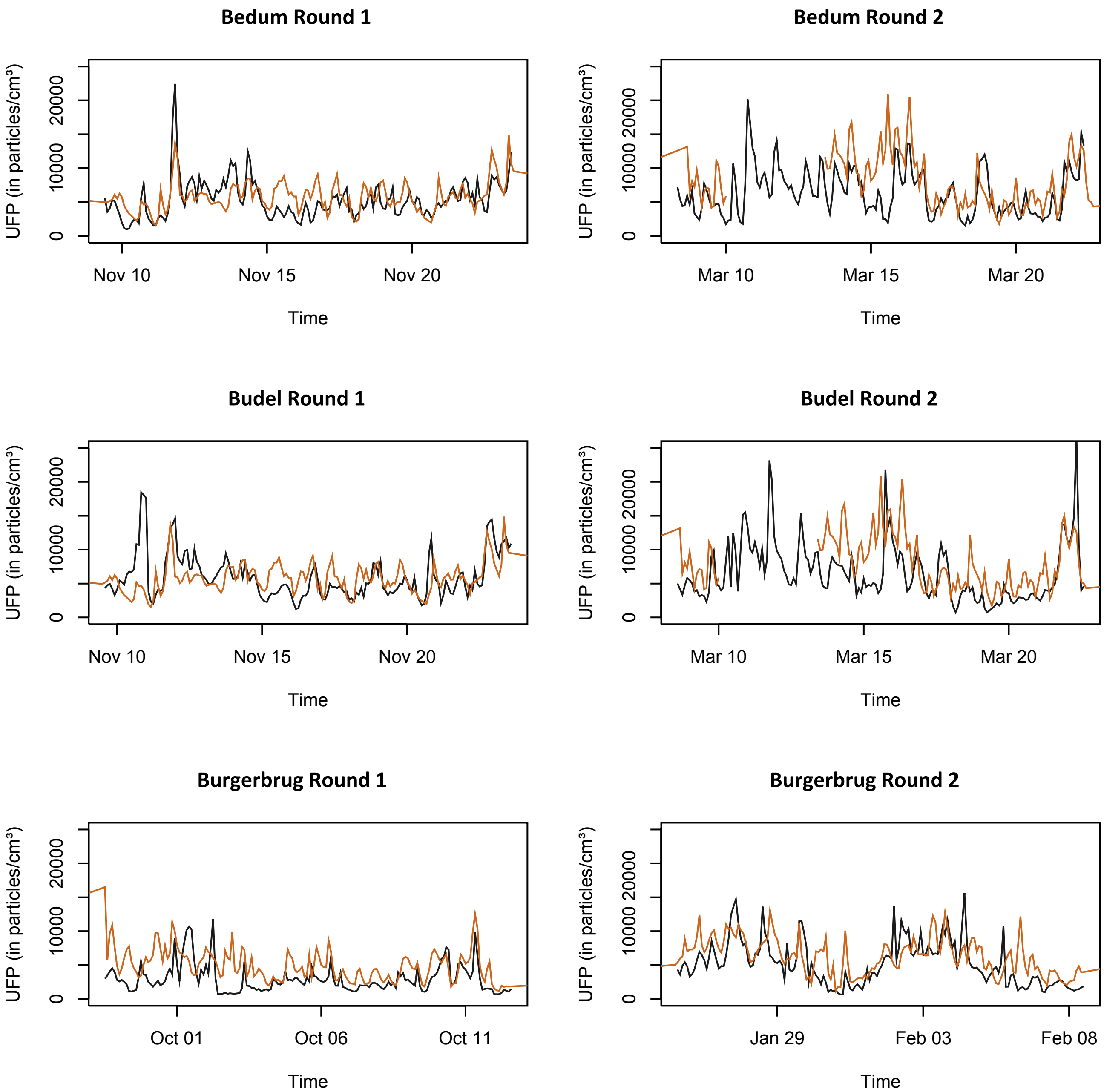
Site	Distance	Pearson	Spearman	2hr Cross-Correlation	Time lag (Hours)	24hr Cross-Correlation	PVE 2hr	PVE 24hr
Bedum	165	0,59	0,50	0,59	0	0,39	0,33	0,15
	165	0,58	0,57	0,58	0	0,52	-0,13	0,02
Budel	100	0,42	0,38	0,44	1	0,26	0,11	0,05
	100	0,54	0,64	0,55	1	0,56	0,08	0,27
Burgerbrug	77	0,43	0,53	0,45	1	0,40	-0,25	0,11
	77	0,57	0,62	0,58	0,5	0,66	0,27	0,41
	77	0,36	0,38	0,46	2¥	0,58	-0,05	0,15
De Zilk	46	-0,05	-0,05	-0,24	3¥	-0,24	-0,41	-0,28
	46	0,44	0,44	0,45	-0,5	0,33	-0,06	0,00
Donkerbroek	125	0,63	0,68	0,67	1,5	0,67	-0,06	0,37
	125	0,61	0,62	0,61	-0,5	0,65	-0,03	0,39
Grijpskerke	125	0,28	0,27	0,33	-1,5	0,38	-0,16	-0,04
	125	0,48	0,51	0,49	0,5	0,57	-0,31	0,22
	125	0,23	0,34	0,25	-1,5	0,23	-0,47	-0,02
Huijbergen	92	0,60	0,66	0,60	0	0,56	0,36	0,27
	92	0,53	0,55	0,54	0,5	0,48	0,20	0,17
	92	0,45	0,50	0,45	0,5	0,53	-0,22	0,12
Lemelerveld	90	0,73	0,70	0,73	0	0,54	0,47	0,29
	90	0,67	0,63	0,67	0	0,63	0,31	0,34
Mantgum	120	0,63	0,65	0,63	0	0,65	0,22	0,36
Maurik	25	0,35	0,87	0,38	-1,5	0,17	0,12	0,03
	25	0,90	0,90	0,90	0	0,75	0,78	0,55
	25	0,70	0,71	0,70	0	0,50	0,36	0,24
Monnickendam	40	0,57	0,35	0,58	-1	0,38	0,32	0,13
	40	0,53	0,66	0,53	0	0,51	0,20	0,23
	40	0,49	0,51	0,49	0	0,49	0,07	0,19
Odiliapeel	65	0,73	0,74	0,74	-0,5	0,61	0,49	0,37
	65	0,66	0,65	0,66	0	0,60	0,38	0,36
	65	0,53	0,53	0,55	1	0,45	0,12	0,19
Oosterhesselen	130	0,44	0,47	0,48	-1	0,43	-0,53	-0,21
	130	0,44	0,42	0,47	1,5	0,33	-0,30	-0,03
Rockanje	78	0,32	0,42	0,36	-2,5	0,36	-0,23	0,12
	78	0,45	0,59	0,46	0,5	0,55	0,15	0,24
	78	0,34	0,35	0,34	0	0,25	-0,57	-0,20
s'Gravenmoer	52	0,70	0,71	0,70	0	0,52	0,47	0,23
	52	0,63	0,65	0,63	0	0,59	0,09	0,33
	52	0,63	0,65	0,63	0	0,30	0,23	0,03
Schimmert	142	0,44	0,48	0,45	-1	0,43	-0,14	0,05
	142	0,42	0,41	0,42	0,5	0,41	-0,32	0,04
Stolwijk	28	0,78	0,83	0,78	0	0,56	0,59	0,31
	28	0,74	0,74	0,74	-0,5	0,59	0,29	0,24
Swifterband	62	0,71	0,67	0,72	0,5	0,69	0,05	0,40
	62	0,29	0,39	0,30	1	0,41	-0,60	-0,10
Voorthuizen	32	0,66	0,76	0,67	0,5	0,62	0,42	0,29
	32	0,60	0,66	0,60	0	0,58	0,36	0,28
	32	0,59	0,57	0,59	0	0,59	0,31	0,30
Zelhem	84	0,44	0,50	0,50	2	0,26	0,12	0,04
	84	0,72	0,71	0,72	0	0,67	0,21	0,41
	84	0,62	0,63	0,65	1	0,61	0,15	0,36

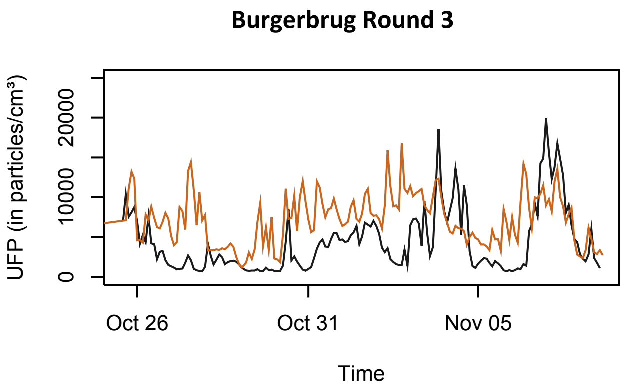
¥Lag was limited to 3 hours maximum

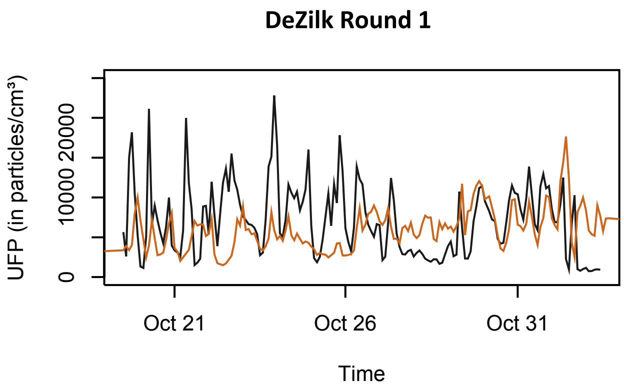
Table C.2: Influence of distance between sites on spatiotemporal correlation, with log-transformed UFP concentrations.

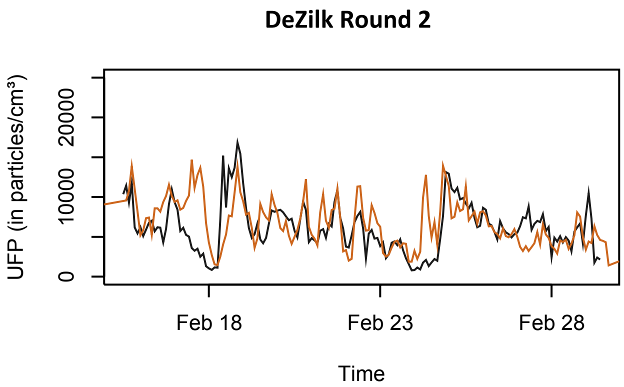
Distance between sites	Median Pearson correlation 2h	Median cross correlation 2h	Median cross correlation 24h	Median PVE 2h	Median PVE 24h
< 50 km	0,69	0,69	0,60	0,47	0,35
50 - 80km	0,61	0,62	0,53	0,24	0,24
> 80km	0,55	0,56	0,52	0,18	0,23

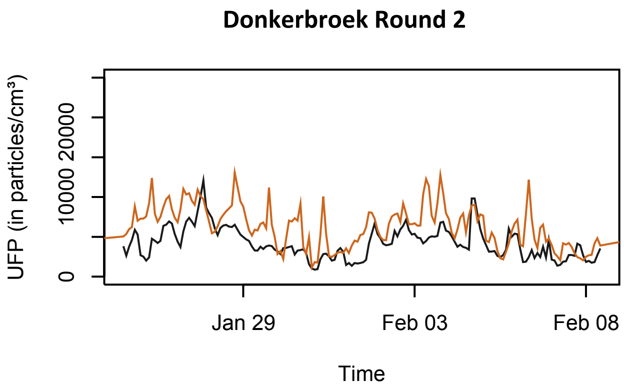
Figure C.1: Temporal variation of 2-hourly averaged UFP concentrations (particles/cm³) at the reference site (black) and the sampling locations (brown).

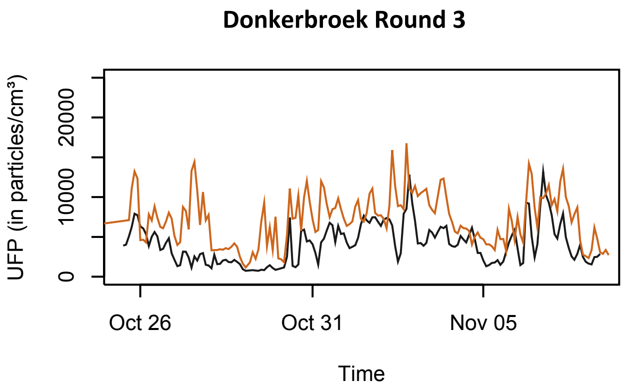



Burgerbrug Round 3
UFP (in particles/cm³)
0
10000
20000
Oct 26
Oct 31
Nov 05
Time


DeZilk Round 1
UFP (in particles/cm³)
0
10000 20000
Oct 21
Oct 26
Oct 31
Time


DeZilk Round 2
UFP (in particles/cm³)
0
10000
20000
Feb 18
Feb 23
Feb 28
Time


Donkerbroek Round 2
UFP (in particles/cm³)
0
10000 20000
Jan 29
Feb 03
Feb 08
Time


Donkerbroek Round 3
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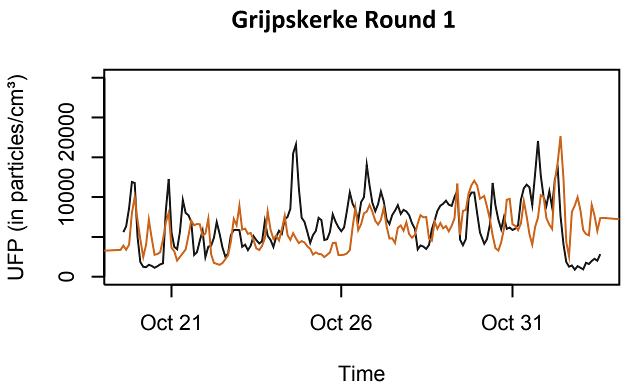
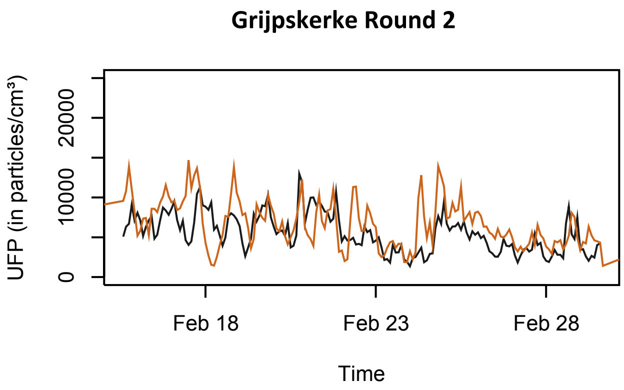
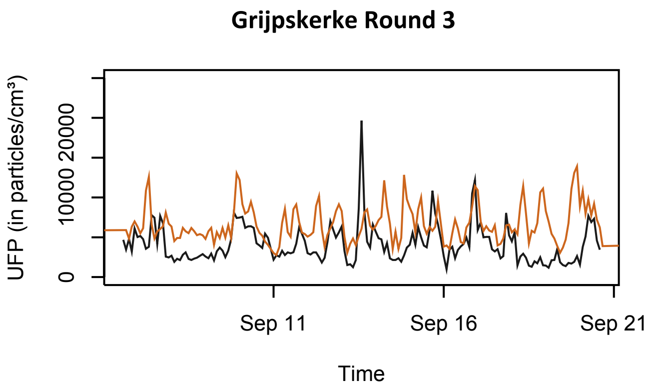
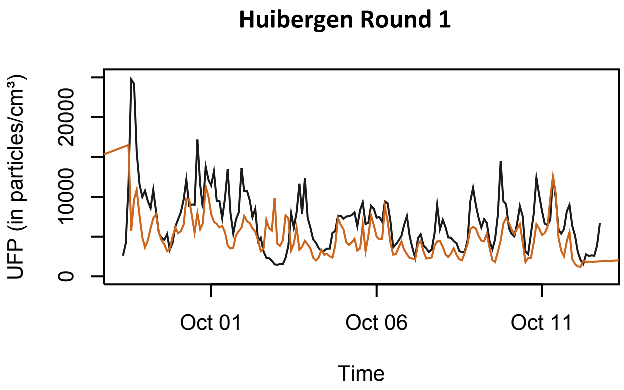
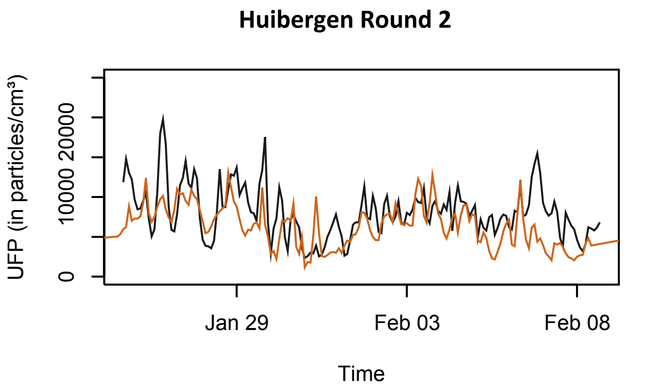
0
10000
20000
Oct 26
Oct 31
Nov 05
Time

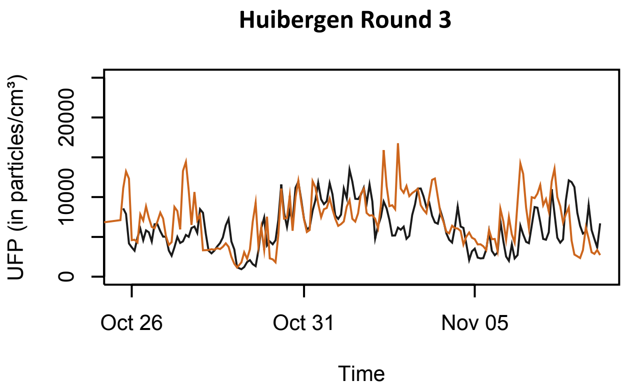

Grijpskerke Round 1
UFP (in particles/cm³)
0
10000 20000
Oct 21
Oct 26
Oct 31
Time


Grijpskerke Round 2
UFP (in particles/cm³)
0
10000
20000
Feb 18
Feb 23
Feb 28
Time


Grijpskerke Round 3
UFP (in particles/cm³)
0
10000 20000
Sep 11
Sep 16
Sep 21
Time


Huibergen Round 1
UFP (in particles/cm³)
0
10000
20000
Oct 01
Oct 06
Oct 11
Time


Huibergen Round 2
UFP (in particles/cm³)
0
10000 20000
Jan 29
Feb 03
Feb 08
Time


Huibergen Round 3
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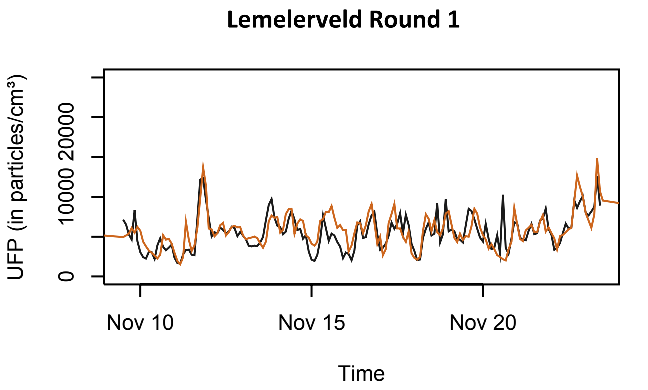
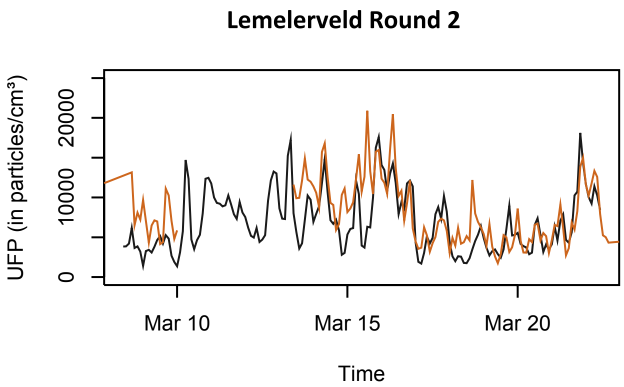
0
10000
20000
Oct 26
Oct 31
Nov 05
Time


Lemelerveld Round 1
UFP (in particles/cm³)
0
10000 20000
Nov 10
Nov 15
Nov 20
Time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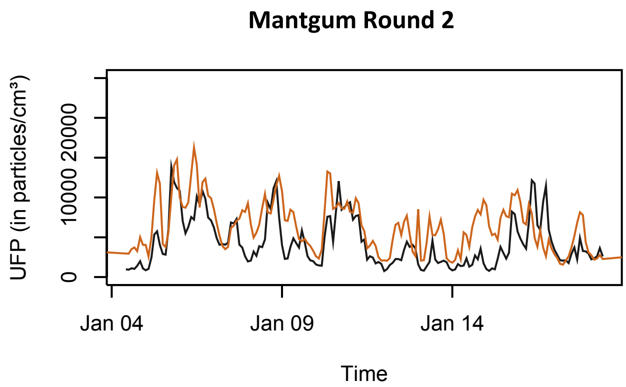
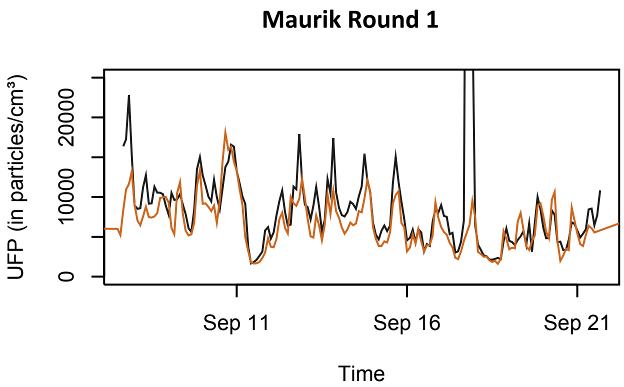
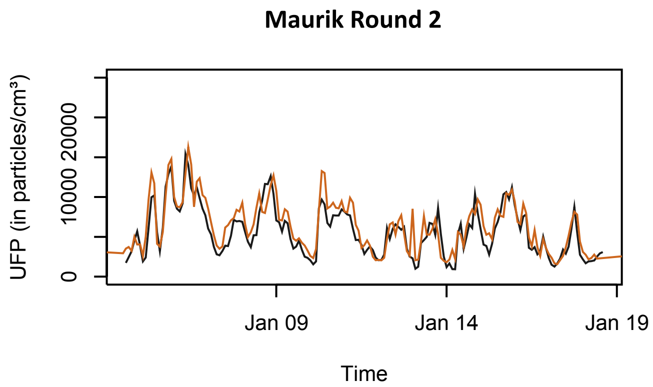
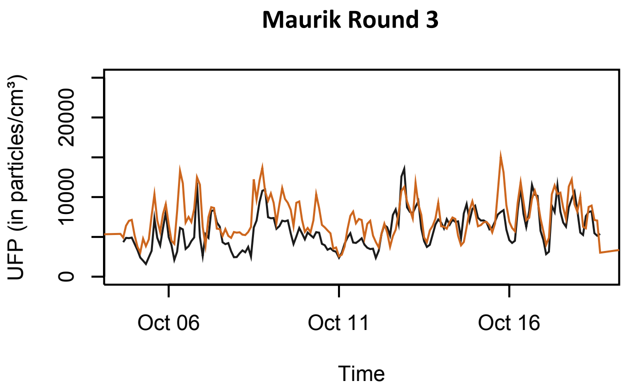
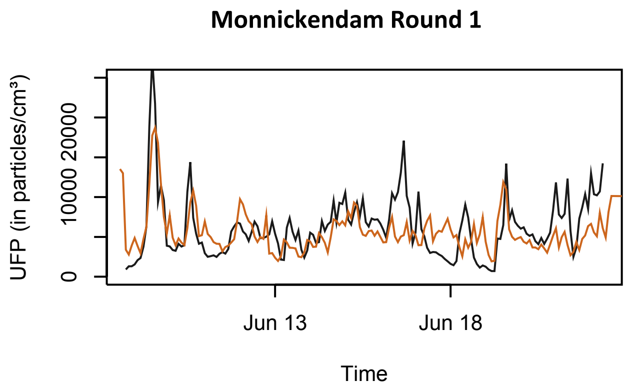
UFP (in particles/cm³)
0
10000
20000
Mar 10
Mar 15
Mar 20
Time

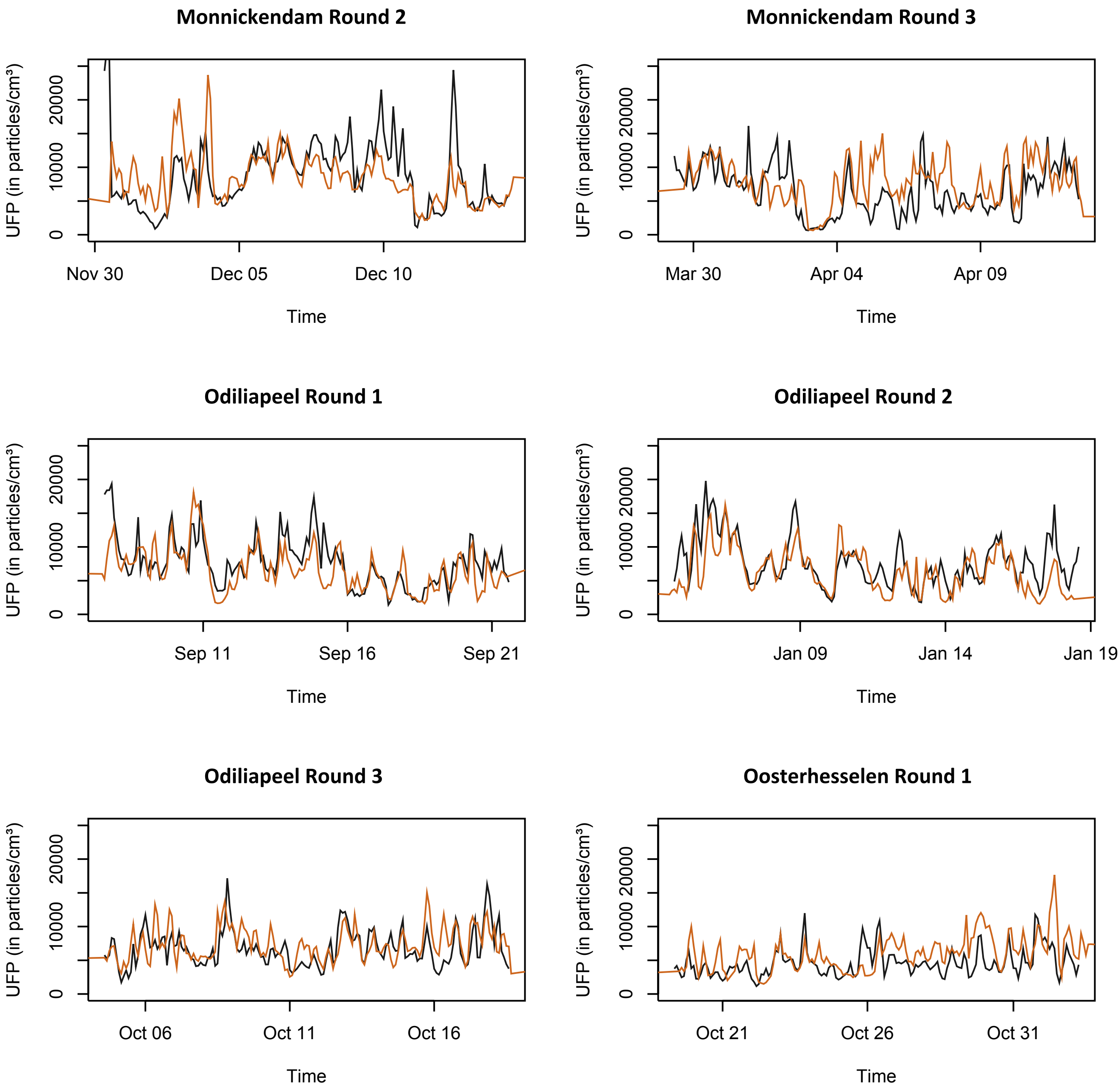

Mantgum Round 2
UFP (in particles/cm³)
0
10000 20000
Jan 04
Jan 09
Jan 14
Time

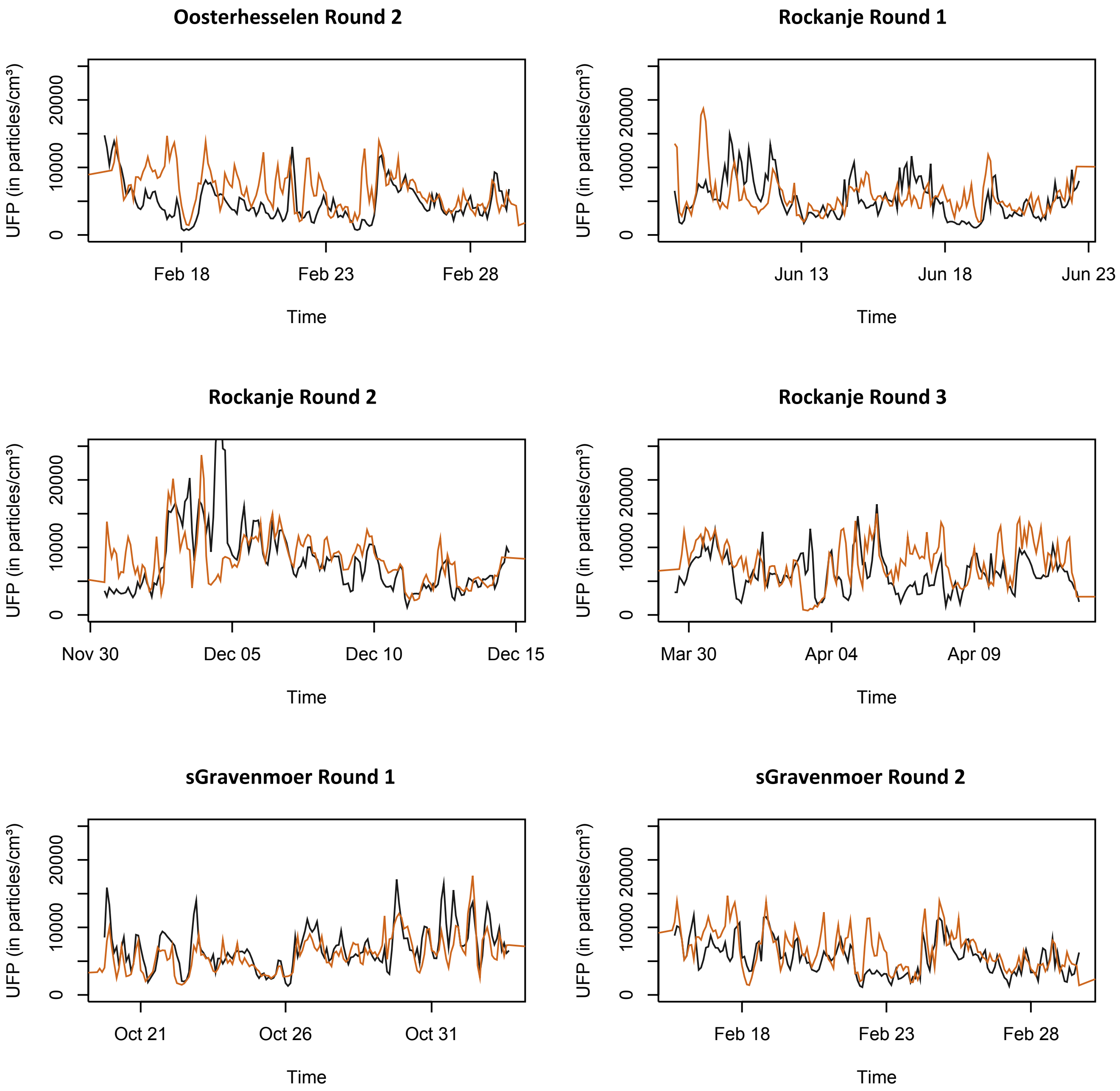

Maurik Round 1
UFP (in particles/cm³)
0
10000
20000
Sep 11
Sep 16
Sep 21
Time

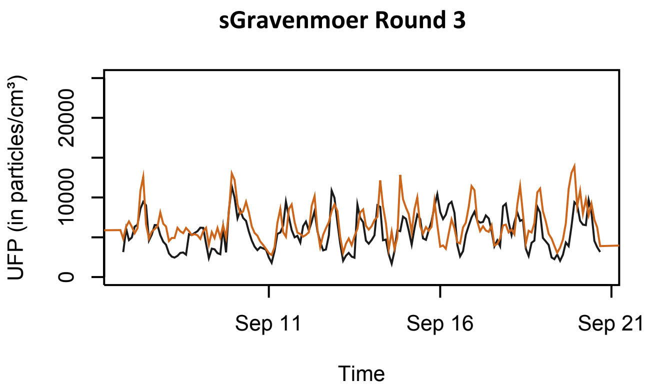

Maurik Round 2
UFP (in particles/cm³)
0
10000 20000
Jan 09
Jan 14
Jan 19
Time

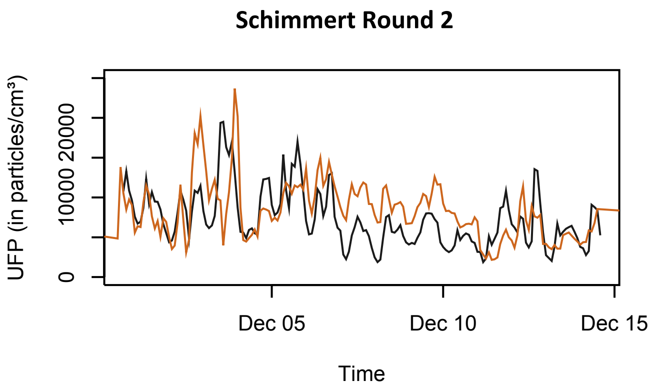

Maurik Round 3
UFP (in particles/cm³)
0
10000
20000
Oct 06
Oct 11
Oct 16
Time

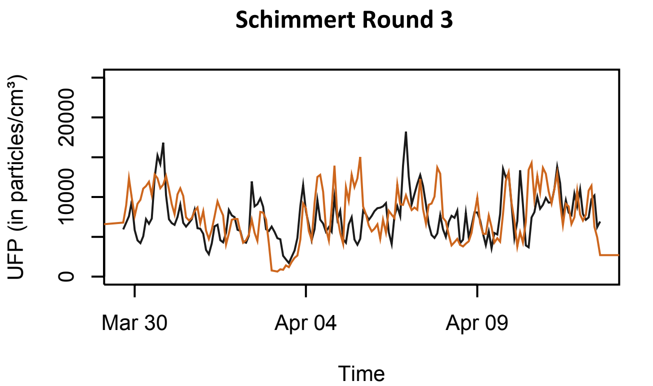

Monnickendam Round 1
UFP (in particles/cm³)
0
10000 20000
Jun 13
Jun 18
Time

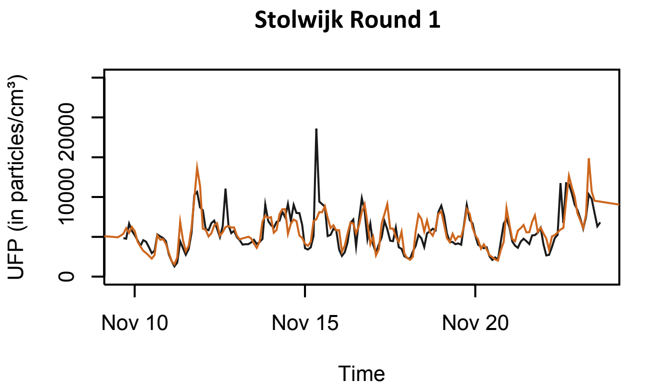

Monnickendam Round 2
UFP (in particles/cm³)
0
10000
20000
Nov 30
Dec 05
Dec 10
Time
Monnickendam Round 3
UFP (in particles/cm³)
0
10000 20000
Mar 30
Apr 04
Apr 09
Time
Odiliapeel Round 1
UFP (in particles/cm³)
0
10000
20000
Sep 11
Sep 16
Sep 21
Time
Odiliapeel Round 2
UFP (in particles/cm³)
0
10000 20000
Jan 09
Jan 14
Jan 19
Time
Odiliapeel Round 3
UFP (in particles/cm³)
0
10000
20000
Oct 06
Oct 11
Oct 16
Time
Oosterhesselen Round 1
UFP (in particles/cm³)
0
10000 20000
Oct 21
Oct 26
Oct 31
Time

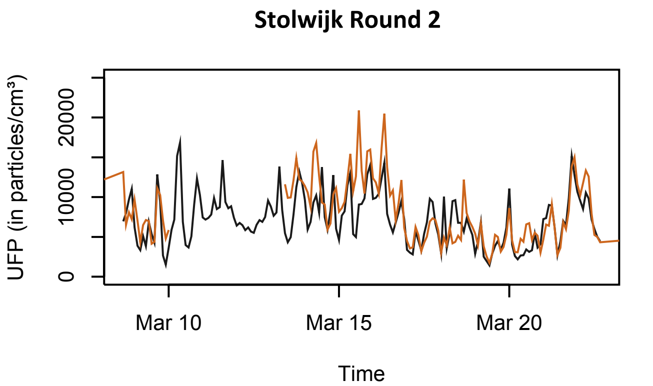

Oosterhesselen Round 2
UFP (in particles/cm³)
0
10000
20000
Feb 18
Feb 23
Feb 28
Time
Rockanje Round 1
UFP (in particles/cm³)
0
10000 20000
Jun 13
Jun 18
Jun 23
Time
Rockanje Round 2
UFP (in particles/cm³)
0
10000
20000
Nov 30
Dec 05
Dec 10
Dec 15
Time
Rockanje Round 3
UFP (in particles/cm³)
0
10000 20000
Mar 30
Apr 04
Apr 09
Time
sGravenmoer Round 1
UFP (in particles/cm³)
0
10000
20000
Oct 21
Oct 26
Oct 31
Time
sGravenmoer Round 2
UFP (in particles/cm³)
0
10000 20000
Feb 18
Feb 23
Feb 28
Time

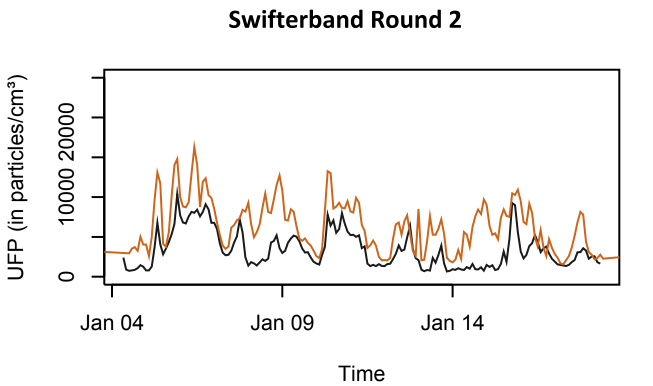

sGravenmoer Round 3
UFP (in particles/cm³)
0
10000
20000
Sep 11
Sep 16
Sep 21
Time

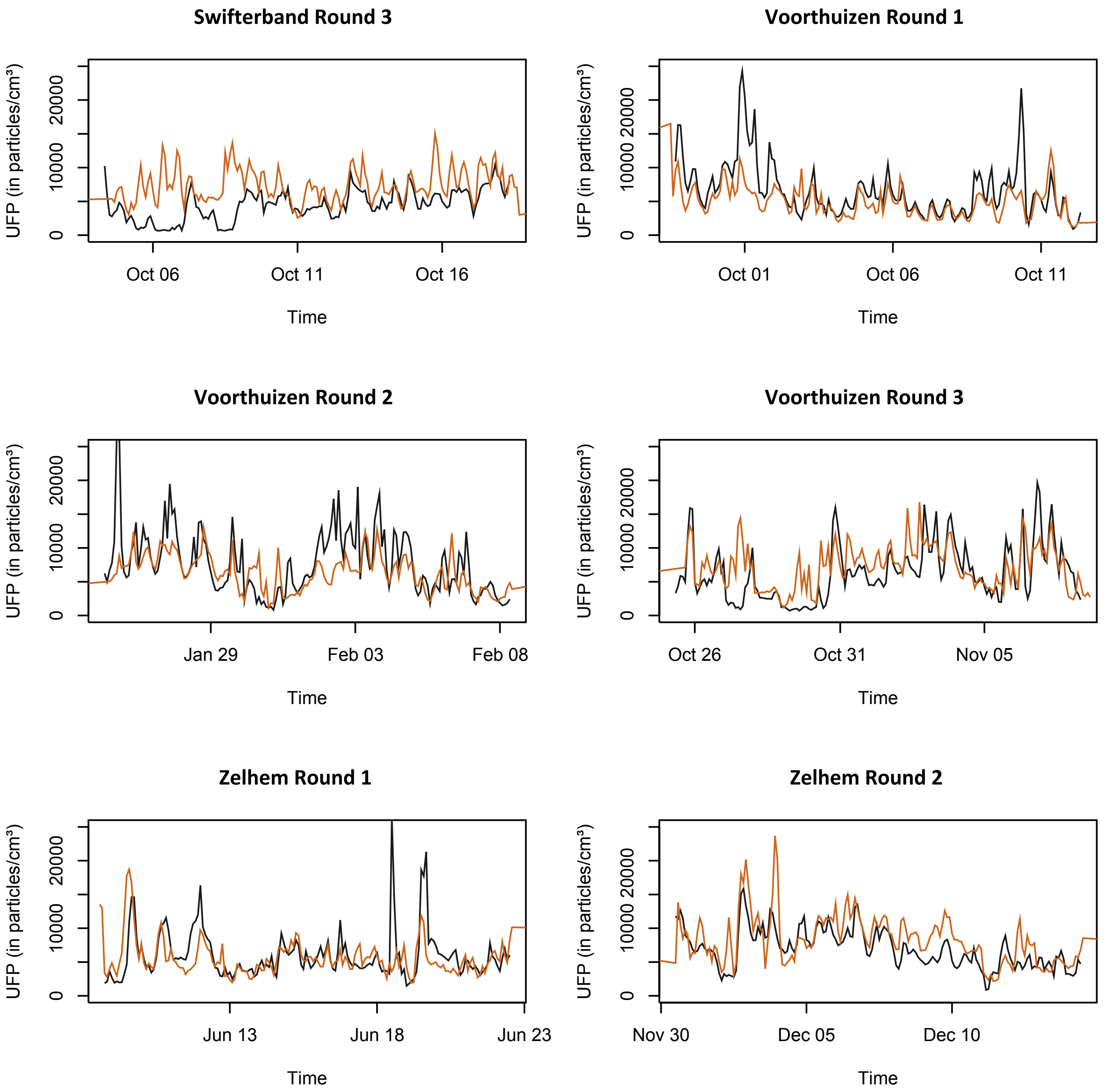

Schimmert Round 2
UFP (in particles/cm³)
0
10000 20000
Dec 05
Dec 10
Dec 15
Time


Schimmert Round 3
UFP (in particles/cm³)
0
10000
20000
Mar 30
Apr 04
Apr 09
Time


Stolwijk Round 1
UFP (in particles/cm³)
0
10000 20000
Nov 10
Nov 15
Nov 20
Time


Stolwijk Round 2
UFP (in particles/cm³)
0
10000
20000
Mar 10
Mar 15
Mar 20
Time


Swifterband Round 2
UFP (in particles/cm³)
0
10000 20000
Jan 04
Jan 09
Jan 14
Time


Swifterband Round 3
Voorthuizen Round 1
Voorthuizen Round 2
Voorthuizen Round 3
Zelhem Round 1
Zelhem Round 2
UFP (in particles/cm³)
0
10000
20000
Time
Oct 06
Oct 11
Oct 16
Oct 01
Oct 06
Oct 11
Jan 29
Feb 03
Feb 08
Oct 26
Oct 31
Nov 05
Jun 13
Jun 18
Jun 23
Nov 30
Dec 05
Dec 10

Zelhem Round 3

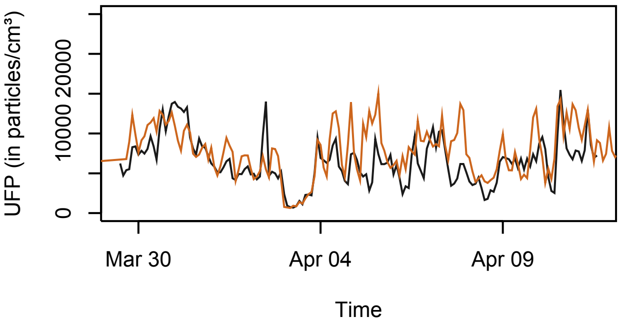


Figure C.2: Average diurnal pattern of UFP for all sites. Reference site in black.

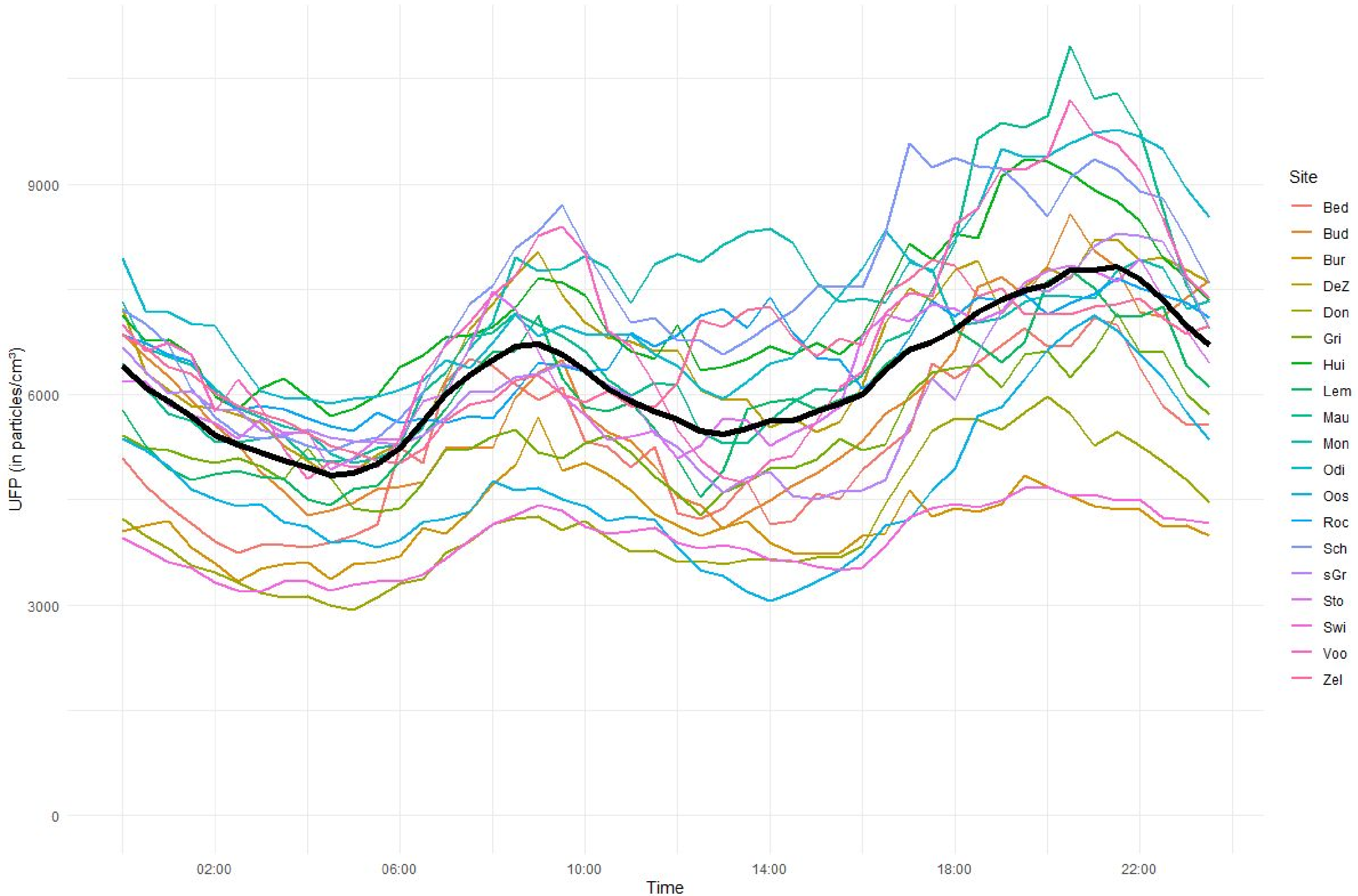


Table C.3: Difference in UFP (in particles/cm^3) per site between cold and warm season.

Site	Cold Season	Warm Season
Bed	5.524	2.920
Bud	5.840	*NA*
Bur	4.262	2.852
DeZ	6.619	*NA*
Don	4.168	*NA*
Gri	6.044	4.122
Hui	6.984	10.164
Lem	5.925	*NA*
Mau	5.807	8.924
Mon	8.562	6.060
Odi	7.216	7.650
Oos	4.669	*NA*
Roc	8.035	5.664
Sch	7.441	7.351
sGr	6.222	5.650
Sto	6.186	7.091
Swi	3.916	*NA*
Voo	6.792	9.357
Zel	7.328	5.930