



1 **Assessing the sources of particles at an urban background site using**
2 **both regulatory instruments and low-cost sensors – A comparative**
3 **study**

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18 **Abstract**

19 Measurement and source apportionment of atmospheric pollutants is crucial for the
20 assessment of air quality and the implementation of policies for its improvement. In most
21 cases, such measurements use expensive regulatory grade instruments, which makes it
22 difficult to achieve wide spatial coverage. Low-cost sensors may provide a more affordable
23 alternative, but their capability and reliability in separating distinct sources of particles have
24 not yet been tested extensively. The present study examines the ability of a low-cost Optical
25 Particle Counter (OPC) to identify the sources of particles and conditions that affect particle
26 concentrations at an urban background site in Birmingham, UK. To help evaluate the results,
27 the same analysis is performed on data from a regulatory-grade instrument (SMPS) and
28 compared to the outcomes from the OPC analysis. The analysis of the low-cost sensor data
29 manages to separate time periods and atmospheric conditions according to the level of
30 pollution at the site. It also successfully identifies a number of particle sources, which were
31 also identified using the regulatory-grade instruments. The low-cost sensor, due to the
32 particle size range measured (0.35 to 40 μm), performed rather well in differentiating sources
33 of particles with sizes greater than 1 μm . However, the ability of the low cost sensor to
34 distinguish diurnal variations and separate sources of smaller particles was more limited. This
35 study highlights the current capability of low-cost sensors in source identification and
36 differentiation using clustering approaches. The current level of source identification
37 demonstrated in this paper indicates the combination of hardware and analytical technique
38 is useful for background site studies, where larger particles with smaller temporal variations
39 are of significant importance. Future directions towards particulate matter source
40 apportionment using low cost OPCs are highlighted.

41



42 **1. Introduction**

43

44 Particulate matter (PM) plays a dominant role in air quality and is known to cause adverse
45 health effects (Dockery et al., 1993; Pascal et al., 2013; Wu et al., 2016; Zeger et al., 2008). As
46 a result, regulatory limits are set for its concentrations, especially in urban areas (US EPA,
47 2012; WHO, 2006). For the implementation of such regulations, the identification of the
48 sources of PM is required. To accomplish this, measurements of the concentrations of PM,
49 typically alongside PM composition, in the area of study are conducted. Until recent years
50 these measurements were usually made using regulatory-grade instruments which, while
51 providing high quality data, are rather expensive thereby limiting the number that could be
52 deployed and consequently the spatial resolution of any measurement network. This
53 increases the spatial interpolation uncertainty (Kanaroglou et al., 2005) and can result in
54 inadequate connection between the levels of air pollution exposures and health effects
55 (Holstius et al., 2014), especially in complex urban environments (Harrison, 2017; Mueller et
56 al., 2016). Additionally, many low and middle income countries are unable to invest the large
57 economic assets currently required for source apportionment, even though in many of these
58 countries, the air quality is poor (Ghosh and Parida, 2015; Kan et al., 2009; Petkova et al.,
59 2013; Pope et al., 2018; Singh et al., 2020).

60 In the past decade, the development of new and cheaper sensors for air quality monitoring
61 has intensified. Many different sensors were introduced measuring either the number
62 concentration or surface area of PM, or the gas phase species (Jovašević-Stojanović et al.,
63 2015; Lewis et al., 2018; Popoola et al., 2018). Overall, low-cost PM sensors currently offer
64 better comparison with regulatory grade equipment compared to their gas phase
65 counterparts (Lewis et al., 2018). However, many shortcomings have been identified in their
66 application, with the most common being the loss of measurement accuracy due to
67 environmental conditions such as relative humidity (RH) variations or high PM concentrations
68 (Castell et al., 2017; Crilley et al., 2018; 2020; Di Antonio et al., 2018; Miskell et al., 2017;
69 Zheng et al., 2018). Measurements in ambient conditions also lead to discrepancies with
70 research-grade instruments, which often measure in controlled environments that are air
71 conditioned (U.S. Environmental Protection Agency, 2016). The reproducibility and variability
72 of the outputs from sensors of the same type can also be problematic (Austin et al., 2015;



73 Sousan et al., 2016; Wang et al., 2015). Therefore, the need for constant and careful
74 calibration is repeatedly highlighted in many studies that evaluate the potential of low cost
75 sensors (Rai et al., 2017; Spinelle et al., 2015, 2017). When these calibration steps are
76 implemented, low-cost sensors have been shown to provide reliable near-real time
77 measurements, maintaining high correlations with research-grade instruments (Kelly et al.,
78 2017; Malings et al., 2020; Sayahi et al., 2019) with the added advantages of lower cost and
79 portability.

80 Consequently, low-cost sensors have been successfully deployed in many studies for which
81 the use of more expensive instruments was not feasible. There is a number of applications in
82 low and middle income countries (e.g. Nagendra et al., 2019; Pope et al., 2018), in studies
83 which included mobile measurements within the urban environment (Ionascu et al., 2018;
84 Jerrett et al., 2017; Miskell et al., 2018), or studies of indoor air quality from multiple sites,
85 such as the SKOMOBO project conducted in New Zealand, in which the air quality in schools
86 was assessed (Weyers et al., 2018). The greatest advantage though is likely, as their name
87 implies, their lower cost which made possible the formation of a network of measuring
88 stations (Feinberg et al., 2019; Kotsev et al., 2016; Moltchanov et al., 2015), increasing the
89 spatial resolution and through new data analysis methods improve the mapping of air
90 pollution up to a sub-neighbourhood level (Schneider et al., 2017). Therefore, it is suggested
91 that the development and use of low-cost sensors, either used individually or in conjunction
92 with research-grade instruments (Snyder et al., 2013), have the potential to radically change
93 the conventional approach of both pollution measuring and policy making (Borrego et al.,
94 2018; Kumar et al., 2015; Lagerspetz et al., 2019), providing a more effective general public
95 information and enhanced environmental awareness (Penza et al., 2014), even for countries
96 with smaller budgets (Amegah, 2018).

97 As yet, studying the different sources of particles at a site with the use of data from low-cost
98 sensors has not been widely attempted yet. Pope et al., (2018) managed to identify major
99 pollution sources studying the ratios of PM of different sizes provided by low-cost sensors,
100 while Popoola et al., (2018) using a network of sensors identified the sources of pollution near
101 Heathrow airport in London, UK. Hagan et al., (2019) applying a statistical method (Non-
102 negative Matrix Factorisation) on low-cost sensor data, identified a combustion factor in a
103 three-factor solution in New Delhi, India. The present study investigates the ability of low-
104 cost sensors to provide measurements that can be used to identify the sources of pollution



105 at a background site in Birmingham, UK, using clustering of particle composition profiles. This
106 method was successfully used in a number of previous studies, though with the use of
107 measurements from research-grade instruments (Beddows et al., 2009, 2015; Von Bismarck-
108 Osten and Weber, 2014; Dall’Osto et al., 2011; Sabaliauskas et al., 2013). To support the
109 clustering method, chemical composition data from both research-grade and low-cost sensor
110 instruments were used, as well as meteorological data from a closely located measurement
111 station. Apart from attempting the source differentiation with low-cost sensor data, a direct
112 comparison with the results from a similar analysis using research-grade instruments is also
113 conducted to not only validate the results but find the strengths and weaknesses of such an
114 application.

115

116 **2. Methods**

117 **2.1 Location of the site and instruments**

118 The measurement site (fig. 1), characterised as an urban background, is the Birmingham Air
119 Quality Supersite (BAQS) located at the grounds of the University of Birmingham (52.45°N;
120 1.93°W), about 3 km southwest from the city centre (Alam et al., 2015). In the present study,
121 measurements from the following instruments for the period 24/01/2020 to 12/3/2020 (the
122 date range was chosen to avoid the effect of the lockdown due to COVID-19) were used (Table
123 1):

124 The Alphasense OPC-N3, which is an optical particle counter, measuring particle number
125 concentrations in the size range between 0.35 to 40 μm at rates up to about 10000 particles
126 per second. As the sample air stream enters the instrument, it passes through a laser beam
127 and the particle size and number concentrations are derived from the light scattered by the
128 particles, based on the Mie scattering theory. It can also provide data for particle mass
129 loadings (PM_1 to PM_{10}) assuming a particle density, shape and refractive index. The OPC is
130 located within the air conditioned station, so measurements represent PM dry mass.

131 The AethLabs MA200 (microAeth MA200) which provides black carbon (BC) information (0-1
132 mg BC/m^3). The sample is deposited onto an internal filter, and an IR light (880 nm) is directed
133 through the sample on the filter and into a detector on the other side of the sample. The
134 amount of light absorbed from the sample is proportional to the BC concentration.



135 Two Naneos Partectors (Naneos Particle Solutions GmbH) which provide the lung deposited
136 surface area metric (LDSA, $\mu\text{m}^2/\text{cm}^3$) in the particle diameter range 10 nm to 10 μm . In
137 general, the provided data is dictated by the particle number concentration and diameter
138 ($Nd^{1.1}$) for both semi-volatile and solid particles. A catalytic stripper (Catalytic Instruments
139 CS015) was used to remove the semi-volatile particles entering one of the two Naneos
140 Partectors. The other Naneos Partector was not subject to the catalytic stripper and therefore
141 measured the surface of all particles. In the present study, apart from the values provided
142 directly from the sensors, the ratio between the measurements of the two Naneos Partectors
143 was also considered according to:

144

$$145 \quad LDSA_{ratio} = \frac{LDSA \text{ after the catalytic stripper}}{LDSA \text{ before the catalytic stripper}}$$

146

147 This was done to resolve whether such a configuration can also provide information such as
148 the level of pollution or the age of the incoming air masses, as increased concentrations of
149 semi-volatile compounds are usually associated with anthropogenic sources, especially in the
150 urban environment (Harkov, 1989; Schnelle-Kreis et al., 2007). Thus, a high $LDSA_{ratio}$ is
151 expected to be associated with fresher pollution (i.e., pollution sources at a close distance
152 from the site), while lower ratios are probably associated with either cleaner conditions or
153 more regional and aged pollution, usually associated with sources at a greater distance from
154 the measuring site. The specific metric though should be considered with caution, as it can be
155 biased by the absolute surface areas measured.

156 The sensors monitoring nitrogen dioxide (NO_2) and ozone (O_3) concentrations are part of a
157 Box Of Clustered Sensors (BOCS) (Smith et al., 2019), which is a low-power instrument based
158 on the clustering of multiple low-cost air pollution sensors allocated in two independent
159 circuits to redundantly measure concentrations and other airflow parameters. The air is
160 driven by a pump through the cell that hosts the electrochemical sensors (EC) and the
161 nondispersive infrared sensors (NDIR). While the EC sensors redundantly (6 sensors per gas)
162 measure carbon monoxide, NO_2 , nitrogen monoxide, oxidizing gases (O_x), the NDIR sensors
163 measure carbon dioxide. EC sensors are based on recording the current generated by redox
164 reactions that occur at the electrode-electrolyte interface in an electrochemical cell
165 composed of three electrodes (working electrode (WE), counter electrode (CE) and reference



166 electrode (RE)). While the gas of interest reacts on the WE surface, the CE completes the
167 redox reaction and the RE ensures that the WE potential remains in the proper range. In the
168 present study, measurements of O_x and NO_2 were only used from the specific sensor.
169 For the same period data from regulatory-grade instruments were also available. Thus,
170 particle size composition data from a Scanning Mobility Particle Sizer (SMPS) in the size range
171 12 – 552 nm, along with PM data for the sizes of 1, 2.5, 4 and 10 μm acquired using a Fidas
172 200E were used. Additionally, chemical composition data for NO_2 , O_3 , as well as SO_4^{2-} , NO_3^-
173 and organic content (size range 40 nm to 1 μm) from an Aerosol Chemical Speciation Monitor
174 (ACSM) were also available. Meteorological data (wind speed and direction, temperature, RH
175 and rain level) from the Birmingham Air Quality Supersite were also used in the
176 characterisation of the clusters formed from both methods.
177 Back trajectory data calculated using the HYSPLIT model (Draxler and Hess, 1998), were
178 extracted by the NOAA Air Resources Laboratory
179 (<https://ready.arl.noaa.gov/READYtransp.php>). Data was processed using the Openair
180 package for R (Carslaw and Ropkins, 2012).

181

182 **2.2 k-means clustering**

183 In this study, two size spectra are considered, one deriving from the OPC and one from the
184 regulatory-grade SMPS. It is noted that the size spectra from the two instruments only briefly
185 overlap in the size range 350 – 552 nm, with the SMPS mostly measuring smaller particles and
186 the OPC mostly measuring larger particles. For the period studied (24/1/2020 – 12/3/2020),
187 874 hours of available data (averaged from 10 second intervals - 76% coverage) from the OPC
188 and 732 hours from the SMPS (66% coverage) were exposed to k-means clustering. k-means
189 clustering is a method successfully used in many studies of particle source differentiation
190 (Beddows et al., 2015; Von Bismarck-Osten and Weber, 2014; Giorio et al., 2015; Wegner et
191 al., 2012) and was proven to have better performance compared to other clustering
192 techniques (Beddows et al., 2009; Salimi et al., 2014), as it was found to produce clusters with
193 the highest similarity between their elements and the highest separation against the other
194 clusters formed (Hennig, 2007). The optimal number of clusters was chosen using two
195 metrics, the Dunn Index and the Silhouette width as proposed by Beddows et al., 2009. The
196 Dunn Index provides a measure of the ratio of the minimum and the maximum cluster. The



197 larger the Dunn Index the better separated are the clusters formed. The Silhouette width is a
198 measure of the similarity of the spectra within each cluster. In the present study the best
199 statistically fitted solution was chosen, though in source differentiation studies such a
200 solution may not always provide with the best separation of all the available sources. Using
201 the aforementioned statistical tests, a six-cluster solution was independently suggested for
202 both the OPC and SMPS datasets.

203

204

205 **3. Results**

206 **3.1 General conditions, sources of particles and pollution at the site**

207 Being an urban background, the site studied presents relatively low concentrations of most
208 pollutants (Table 2), without the effect of direct sources of pollution, such as traffic. Wind
209 rose and polar plots illustrating the conditions in the period studied are found in figure S1.
210 The main source of pollution lies on the north and northeast sectors, where the city centre is
211 located, as well as in the southern and eastern sectors where a populous residential area is
212 located. As a result, the main sources of NO₂ and BC as well as the smaller sized PM are
213 associated with easterly winds (this though is not reflected in particles observed in the SMPS
214 size range). For the PM₁₀ apart from the aforementioned, increased concentrations are also
215 found with southwestern winds likely associated with marine sources. Typical for the UK, the
216 average wind profile for the period consists mainly of western and southwestern winds
217 (McIntosh and Thom, 1969), reducing the effect of the pollution sources in the east of the
218 site. Finally, the secondary pollutants NO₃⁻ and SO₄²⁻ which are in most cases associated aged
219 pollution and long-distance transport, have less consistent profiles, though they both seem
220 to be mainly associated with southern wind directions.

221

222 **3.2 Clustering of the OPC data**

223 Due to the larger particle sizes measured by the OPC-N3, the differences in the cluster profiles
224 are mainly associated with the particle number concentrations and to a lesser extent on the
225 different peaks, which are less distinct due to the smaller variation found as particle diameter
226 increases. The frequency of the clusters formed, and their diurnal occurrence is shown in



227 figure 2. The average particle size distribution spectra and wind roses for the clusters formed
228 are found in figures S2 and S3.

229 The six clusters formed from the OPC data are:

230 **OPC.1:** A rather polluted group with the highest NO₂ concentrations and average secondary
231 pollutants, PM and LDSA ratio. Its fresher polluted character is further confirmed using the
232 SMPS data which showed higher than average particle concentrations for particles with
233 diameter smaller than 50 nm. This group presents low average temperature, RH and slower
234 than average southwestern winds, which is explained, to an extent, by the cluster being more
235 frequent during night-time.

236 **OPC.2:** The second group refers mainly to a single midday event on 12/3/2020 with high-
237 speed southwestern winds, high temperature and very low RH. On this day the concentrations
238 of all the pollutants were rather low, though due to the high wind speeds (an increase in the
239 wind speed is observed at the start of the occurrence of this cluster – at 10:00 AM - which
240 affects the particle distribution profile as can be seen in Figure S4) the PM₁₀ were close to
241 average (when PM₁ and PM_{2.5} were rather low) indicating the stronger presence of coarser
242 particles, possible of marine origin as shown by the back trajectories, a source with an
243 increasing importance at larger size PM at this area (Harrison et al., 2004; Taiwo et al., 2014).
244 This group presents the highest LDSA ratio, which is in agreement with the low concentrations
245 of the secondary pollutants.

246 **OPC.3:** A group occurring mainly during some of the midday periods in January, with the
247 lowest temperature and wind speed averages, as well as the highest average RH, containing
248 both southwestern and southern winds. While the concentrations of the measured pollutants
249 are close to average, high sulphate and ozone concentrations were found, with the former
250 pointing to air masses with higher concentrations of aged pollutants. The LDSA ratio though,
251 was found to be very high despite the higher concentrations of sulphate and nitrate. The near
252 average NO₂ concentrations may point to the effect of a nearby pollution source that may
253 resulted to the increased LDSA ratio found.

254 **OPC.4:** A group with low concentrations of NO₂, BC and PM, but close to average secondary
255 pollutants' concentrations. It is associated with close to average temperature, RH and wind
256 speed of mainly southwestern directions. It is slightly more frequent during daytime and has
257 lower than average concentrations of particles in the SMPS range.



258 **OPC.5:** This group includes the most polluted conditions in the area throughout the day. It is
259 associated with western and southwestern winds of average speed, high temperature and
260 lower than average RH. Most pollutant concentrations, including PM, are rather high while
261 O₃ is low. Similarly, it presents the highest concentrations of particles in all SMPS size ranges.
262 This cluster also includes the more polluted conditions found with north-eastern winds.

263 **OPC.6:** A group associated with rather clean conditions, presenting the lowest concentrations
264 of NO₂, BC, NO₃⁻ and organic content. It is associated with higher than average temperature
265 and wind speed and lower than average RH, and has low concentrations of PM₁ and PM_{2.5},
266 while PM₁₀ concentration is close to average. It is more frequent during daytime, which
267 probably explains the highest O₃ concentrations. The fast-moving southwestern air masses,
268 which this group is associated with, are probably of marine origin that have not passed
269 through any significant pollution sources, which can be further suggested by both the low
270 LDSA values and the highest LDSA ratio.

271

272 **3.3 Clustering of the SMPS data**

273 In the past, a number of studies on the sources of particles were conducted for both the
274 greater area of Birmingham and specifically the site in the University (Harrison et al., 1997;
275 Taiwo, 2016; Yin et al., 2010). As, these studies mainly focused on the chemical composition
276 of coarser particles, to the authors' knowledge this is the first study that uses ultrafine particle
277 size distribution data to study the sources of particles in Birmingham, UK. The frequency and
278 hourly occurrence of the six clusters formed from the SMPS data is found in figure 3. The
279 average particle size distributions and wind roses for the clusters formed are found in figures
280 S5 and S6.

281 **SMPS.1:** This group contains averagely polluted hours and is associated with fresher
282 pollutants (such as NO₂ or NO) and PM, while secondary pollutants such as NH₄⁺, NO₃⁻ and
283 SO₄²⁻ are relatively low. Due to being associated with fresher emissions this group presents
284 higher than average concentrations of particles below 50 nm and a low LDSA ratio. It is
285 associated with average speed southwestern winds (it also includes the small portion of
286 north-eastern winds) and temperature, higher than average RH and occurs more frequently
287 during late night and early morning hours.



288 **SMPS.2:** Similar to the first group, average pollutants' concentrations are found in this group
289 with low concentrations of secondary pollutants. It is associated with slow western and
290 southwestern winds, lower than average temperatures and RH and is more frequent during
291 early morning hours. It has the highest concentrations of particles with diameter smaller than
292 20 nm, but the particle concentrations become relatively smaller as their size increase.

293 **SMPS.3:** This is a small group containing very clean night hours mainly in February, with higher
294 than average temperature, lower than average RH and strong western and southwestern
295 winds. It has low concentrations of pollutants and PM apart from O₃ (despite the time of day),
296 though PM₁₀ concentration is enhanced, probably associating this group with stronger marine
297 origins. The particle concentrations of all size ranges below 500 nm are the lowest among the
298 groups formed and along with the high LDSA ratio are in agreement with the very clean
299 conditions associated with this cluster. This cluster, contrary to all other, presents two peaks:
300 one peaking just below 30 nm and another one just over 100 nm, which indicates that it is
301 probably associated with at least two different sources.

302 **SMPS.4:** This group presents near average concentrations of all the pollutants studied. PM₁
303 average concentration is rather low while PM₁₀ is higher than the average. It is associated
304 with average speed southwestern winds, higher average temperature and low RH. It is more
305 frequent during midday and evening hours and it appears to represent the most common
306 conditions in the area, hence having the highest frequency of all clusters.

307 **SMPS.5:** This is a unique group associated with southern winds, the side at which the central
308 part of the University resides. This is the most polluted group, probably affected by emissions
309 from the University and the residential area found in that direction, with very high
310 concentrations of all the pollutants (apart from O₃), PM and ultrafine particles with available
311 data. The LDSA ratio is very high and this is probably due to the great surface area of the
312 involatile component found. It is associated with very slow wind speeds, low temperature,
313 very high RH and occurred evenly throughout the day, mainly on the first weeks of the
314 campaign when pollution levels were rather high, probably due to increased heating
315 emissions.

316 **SMPS.6:** This group presents low concentrations of all pollutants (apart from O₃), PM and
317 ultrafine particles with available data and is associated with western winds with higher than
318 average speed, near average temperatures and low RH. It occurred more frequently during



319 evening hours and almost equally frequently throughout the whole study period apart from
320 the first 2 weeks when pollution levels were rather high.

321

322 **3.4 Direct comparison between the methods**

323

324 Due to the difference in the size ranges measured by the SMPS and OPC instruments, it is
325 evident that a direct comparison between the two methods would provide mixed results as
326 some clusters found using the SMPS data are not detectable with the OPC, and vice versa.
327 The particle size range that is common between the two instruments lies at about 350 – 550
328 nm. Therefore, many particle sources associated with particles in the size range below the
329 minimum detectable size of the OPC are not expected to be found using its data and vice
330 versa. At a background site though, many of the sources of smaller sized particles play a less
331 important role as they are usually associated with fresher emissions, which are not common
332 to such sites.

333 The clustering process attempts to separate the particle size distributions into groups with as
334 similar spectral profiles as possible, while being as different to the other groups as possible.
335 As expected, the SMPS is more capable in separating different cluster profiles at the size range
336 smaller than 500 nm, a size range in which the cluster profiles formed by the OPC are almost
337 uniform (fig. 4). This shows the limitation of the OPC data to distinguish ultrafine particle
338 variations and thus it does not provide insight for the sources of particles within this size
339 range. On the other hand, the OPC performs much better in identifying different sources
340 when considering larger particles in the range between 1 – 10 μm , for which it manages to
341 clearly distinguish variations between the groups formed (fig. 5). The clusters formed using
342 the OPC data were also better associated with different sources of PM_{10} (fig. 6), compared to
343 those deriving from the SMPS data (fig. S7).

344 Table 3 contains the cluster relationships between the two methods, while Table S1 contains
345 the conditions observed when pairs of clusters from both methods are considered. The OPC.2
346 and OPC.3 clusters appear infrequently, and it would be nonsensical to directly associate
347 them with SMPS groups, as they appear under very specific conditions, that either are not
348 detected or are not identified as separate cases by the SMPS. As a result, they will be
349 separately studied later in this study.



350 The OPC.1 was mainly associated with SMPS.4 and SMPS.6 and to a lesser extend with
351 SMPS.1. OPC.1 has higher frequency during night times and it shares many of these hours
352 with groups SMPS.4 and SMPS.6, while with SMPS.1 it mainly shares early morning hours. It
353 includes the more polluted portion of the rather clean SMPS.6 and a portion with lower PM₁₀
354 (though not much difference from average pollutants' concentrations) from the more
355 polluted SMPS.4. It is interesting that the variation between the subgroups (in relation to
356 SMPS clusters) of the OPC.1 is very small for the NO₂ concentrations, a pollutant for which its
357 variations are not expected to be directly "visible" at the size range of the OPC as it is mainly
358 associated to fresher emissions. No great variation was found for the wind direction in the
359 subgroups of OPC.1, though it includes the lower temperature and higher RH conditions of
360 the SMPS clusters it is associated with. The OPC.1 includes the relatively clean part of the
361 more polluted SMPS.1 and the more polluted portion of the cleaner SMPS.6. While this does
362 not provide a clear connection between the OPC and SMPS results, it shows that there is
363 consistency in the results provided by the former in identifying particle sources of specific
364 qualities.

365 Similarly, OPC.4 was mainly associated with SMPS.4 and SMPS.6. As the OPC.4 occurs under
366 cleaner conditions, it includes the less polluted hours of both the SMPS clusters it is mainly
367 associated with, though the concentrations of the secondary pollutants such as NO₃⁻ and SO₄²⁻
368 are closer to the average. The OPC.4 is associated with the cleaner portion of the
369 aforementioned SMPS clusters with higher average temperature and RH though with variable
370 wind speeds.

371 OPC.5 represents a polluted group of hours associated mainly with SMPS.4, SMPS.5 and
372 SMPS.6. Being a group of hours associated with higher concentrations of pollutants, it
373 includes the more polluted portions of SMPS.4 and SMPS.6 with average meteorological
374 conditions, though lower wind speeds. It also coincides with the largest portion of SMPS.5,
375 mainly in the sixth week when the temperature was the lowest, including the portion with
376 the higher concentrations organic content and NO₃⁻. SMPS.5 is the group that is associated
377 with southern wind directions, a side from which a source of secondary pollutants (NO₃⁻, SO₄²⁻
378 , NH₄⁺), organic content and particles of diameter greater than 100 nm occurs. The OPC.5 is
379 associated with the part of SMPS.5 which is more burdened from secondary pollutants, hence
380 very large concentrations are observed for them.



381 Finally, OPC.6 is mainly associated with SMPS.2, SMPS.4 and SMPS.6. Being a cleaner group
382 of hours, it includes the portion of these SMPS clusters with lower pollutant concentrations
383 but higher PM₁₀ concentrations (though with lower PM₁ concentrations). These rather clean
384 conditions, along with the western and southwestern high-speed winds in average and the
385 large PM₁₀ concentrations, further enhance the possible marine character of this cluster. Due
386 to the size range of these particles such variation is not clearly identified by the SMPS,
387 resulting to them not being clearly separated when its data is considered.

388 The weekly contribution of each cluster group from the analysis of either dataset is found in
389 Figure 7 and the conditions on each week studied in Table S2. It is evident that the variation
390 from the SMPS is greater than that of the OPC, as the latter is less affected by the diurnal
391 variations. It is apparent that it is easier to comprehend the clusters' variation in association
392 with the levels of pollution in the site (the more polluted weeks have a greater portion of
393 SMPS.1 and SMPS.5), while for those with lower concentrations of pollutants the SMPS.4 and
394 SMPS.6 are more enhanced. These variations are harder to distinguish using the OPC data, as
395 they are less apparent in the size range measured by the sensor. To further understand the
396 possible sources using the latter, information from other instrument which provide chemical
397 composition data are needed, though it is still hard to pinpoint exact sources, due to the OPC's
398 weakness in explaining distinct particle sources within the day.

399

400 **3.5 Case studies**

401

402 **OPC.2**

403 OPC.2 occurs mainly on a single day in May (12th) with higher than average temperature and
404 strong western winds. This was the cluster with the lowest concentrations of NH₄⁺, NO₃ (about
405 an order of magnitude compared to average conditions) and SO₄²⁻, rather low concentrations
406 of NO₂, BC and high O₃. Using the SMPS data, this group of hours seems to follow the trends
407 of BC, associating it with SMPS.6 for low, SMPS.1 and SMPS.2 for medium and SMPS.4 for
408 higher concentrations of BC. This cluster has very low PM₁ and PM_{2.5} and near average PM₁₀
409 concentrations, probably associating it with marine sources (due to the high wind speed). Due
410 to this, it is not clearly separated using the SMPS data, which does so for the hours of this
411 group according to the level of fresher pollutants, the variation of which is smaller in this type



412 of environments. This cluster seems to be the result of the change in the wind profile which
413 greatly affected the coarser particles at the site (figure S4).

414

415 **OPC.3**

416 The third cluster formed using the OPC data, was a rather small group of hours in late January
417 (25,27 and 28th), with the lowest average temperature and wind speed compared to the rest
418 of the clusters. The wind direction profile for this group contains both western and southern
419 winds, the latter being associated with high concentrations of pollutants (as found by the
420 study of the SMPS data). The majority of the hours in this group (65%) were characterised as
421 freshly polluted when using the SMPS data, mainly associated with SMPS.2. Unfortunately,
422 data of NO₂, BC, O₃ and PM for this group were very scarce from regulatory-grade instruments
423 (due to instrument error). The ACSM data, which were available for the hours of this cluster
424 pointed to marginally lower than average values of organic content, nitrate and ammonium,
425 while the sulphate concentrations were rather high. Using the low-cost sensor data, it is found
426 that this group has the highest BC, O₃ and involatile component of LDSA while NO₂, and CO
427 were the lowest among the groups. This group also had the highest average particle
428 concentration in the size range of the OPC, which is in agreement with the highest PM
429 concentrations in all ranges (PM₁, PM_{2.5}, PM₁₀). As this is not visible from the SMPS, the cluster
430 associated with this group has nearly average particle concentrations in the SMPS particle
431 ranges. This group was not distinctively detected by the SMPS due to presenting variation in
432 larger sized particles, which can be one of the weaknesses of studying the sources of such
433 particles using SMPS data alone. The OPC.3 appears to contain the more polluted slow-
434 moving portion of SMPS.2 with enhanced SO₄²⁻, BC and PM concentrations.

435

436 **SMPS.3**

437 The third cluster from the analysis of SMPS data presented a unique profile with two peaks,
438 one below 30 nm and one a bit over 100 nm. This unique group was associated with very
439 clean conditions, with very low concentrations for all the pollutants with available data (apart
440 from O₃), as well as low particle concentrations for all the ranges in the SMPS and OPC range
441 as well as PM₁ and PM_{2.5}. The concentrations of PM₁₀ and SO₄²⁻ were somehow higher but
442 still lower than the average in the area for the period of the study. This group is associated



443 with high average temperature and wind speed and rather low RH, with wind directions being
444 mainly southwestern and western. This group occurred solely at night hours during a number
445 of relatively warm nights mainly in February and to a lesser extend in March. Even with very
446 low particle concentrations (as found by both the SMPS and OPC) the presence of two
447 separate peaks in the size range of the ultrafine particles is indicative of more than one
448 simultaneous source. Due to these sources of particles occurring at the ultrafine particle
449 range, the OPC was not able to distinguish this special condition and grouped the hours of
450 this cluster to a number of clusters (mainly OPC.5 and to a lesser extend OPC.1 and OPC.6),
451 occurring either during night-time or throughout the day. The inability of the OPC to
452 distinguish complicated conditions in the ultrafine range is a weakness of the OPC that should
453 be considered when such conditions are anticipated.

454

455 **4. Discussion**

456

457 As the SMPS measures smaller particle sizes and with better accuracy, compared to the OPC,
458 it managed to better separate the different sources of fresher pollution with the main
459 differentiating factor being the time of the day, for which the variability of such sources is
460 more prominent. The differences in NO₂ concentrations, which are mainly associated with
461 fresher emissions are more distinct between the groups and using this data better separation
462 of very clean (SMPS.3) and very polluted conditions from a distinct source (SMPS.5) was
463 achieved, while the other groups described mostly average conditions with lesser variability
464 (as expected in this range at a background site). Additionally, using the SMPS data it is possible
465 to distinguish multiple sources of ultrafine particles (SMPS.3), as they can appear as multiple
466 peaks within the SMPS spectra. This is not possible using the OPC data as the size range
467 measured by this instrument cannot identify such cases.

468 Contrary to the SMPS, using the OPC data provided less distinct separation of fresher
469 emissions (as expected due to the lack of data of small sized particles). Additionally, the OPC
470 data is less sensitive to diurnal variations due to the range of particles covered, which are in
471 a size range that does not vary significantly through the day but between days. This results in
472 the less distinct diurnal variations found between the groups formed. The analysis of the OPC
473 data though managed to adequately separate conditions and/or sources associated with



474 larger particles, such as aged pollution (for which it also managed to separate a small time-
475 window with very strong sulphate presence – OPC.3) which has the greatest contribution in
476 the particle chemical composition for the study area (Harrison et al., 2003; Taiwo, 2016; Yin
477 et al., 2010), RH variations or air masses of marine origin. To an extent, this might be due to
478 the number of clusters chosen as there is a possibility that a larger number of clusters from
479 the SMPS may separate sources of larger particles better, though with the risk of also
480 separating similar sources.

481 To sum up, the study of SMPS data with k-means clustering is far superior at separating
482 complex pollution sources within urban environments in which the variation of very small
483 particles is crucial for identifying particle and emission sources. This advantage of the SMPS
484 will not be overcome even with a denser measuring network of OPCs that could be acquired
485 for the same cost of the SMPS. However, clustering of the OPC data can provide useful
486 information to assess the sources of air pollution at background sites in which the direct
487 (smaller) particle sources are few. The method managed to find sources of greater pollution
488 associated with higher concentrations of particles of greater sizes (which are mainly
489 associated with aged pollution though), showing that the footprint of pollution in the ultrafine
490 particle range can have a detectable effect in coarser particle distributions as well. While not
491 as precise as the SMPS, a denser network of such instruments in background sites can be
492 more beneficial and more cost efficient in studying multiple pollution sources or “hot spots”
493 within the urban environment.

494 The current inability of low-cost PM sensors in measuring particle size spectra at small sizes
495 (<300 nm) is the greatest drawback in their application for separating particle sources, since
496 much information is contained in these smaller sizes. OPCs using shorter wavelength light
497 sources and hence smaller particle detection could be beneficial here. Also, there are several
498 low-cost sensors that provide insight for the surface area or the total number of particles in
499 the ultrafine particle size range (such as the LDSA sensor used in this study). The combined
500 use of the OPC with these instruments, along with sophisticated statistical techniques, may
501 provide possibilities for more precise source differentiation than shown in the present study.
502 It is noted that while clustering of particle number size distributions is one approach in the
503 study of the source assessment of particles, other alternative methods, such as the Positive
504 Matrix Factorisation (PMF), may also provide useful results.



505

506 **5. Conclusions**

507

508 The present study investigates the capabilities of a low-cost OPC sensor for source
509 differentiation at an urban background site in Birmingham, UK. It is used alongside a
510 regulatory-grade SMPS instrument, which has previously been used successfully for source
511 differentiation. The clustering approach identified optimal solutions of six clusters for both
512 the SMPS and OPC data. There were similarities between the SMPS and OPC solutions, which
513 provide insights into periods of low and high pollution. However, large differences were also
514 observed. A more distinct separation of direct emission sources was achieved using the SMPS
515 data, which identified sources with time windows that correlated with extreme NO₂
516 concentrations (either high or low), as well as periods with more complex sources. The OPC
517 was able to distinguish time periods with greater variation of super micron sized particle
518 sources (e.g. marine sources). There seems to be a clearer distinction of the diurnal variability
519 of sources using the SMPS data, while the OPC seems to be able to only distinguish the
520 variability within periods of days rather than hours, as found by the less variable diurnal and
521 weekly variation. This though might not be a great drawback when considering background
522 sites, as this variability is smaller in such environments which are mainly affected by regional
523 pollution, while the local emissions are less and more distinct. Low-cost sensors can be a
524 reliable alternative for source identification studies in environments with less complex
525 sources, which present smaller alterations within the span of the day. Still, such instruments
526 cannot be used for scientific analyses which require greater precision. Their application will
527 probably be adequate when studying the sources of particles with a more regional character
528 (e.g. marine sources) rather than direct and variable sources (e.g. traffic or cooking emissions)
529 and can provide enough information for the air quality levels, sources and conditions these
530 are anticipated from. Such studies may include the analysis of mineral dust events resulting
531 from either anthropogenic activities or meteorological events (e.g. dust storms), bioaerosol
532 events in forested areas and other sources which affect mainly the composition of coarser
533 particles.

534 This study demonstrates that single low-cost sensor PM units can provide sensible source
535 differentiation of large sized PM pollution sources. This allows for the prospect of source



536 apportionment via networks of low-cost sensors in the near future, thereby allowing
537 triangulation of sources. The development of more sophisticated low-cost sensors in
538 conjunction with their low cost ensures the prospect of the application of a denser
539 measurement network, making better air quality monitoring and control feasible in the near
540 future. This though, requires more similar studies which can further elucidate the strengths
541 and weaknesses of those sensors compared to the regulatory-grade ones, as they develop.

542

543 **Author Contributions**

544 The study was conceived and planned by FDP who also contributed to the final manuscript,
545 and DB who also carried out the analysis and prepared the first draft. AS, MH, DCSB and SD
546 have provided with the data for the analysis. DCSB provided help with the analysis of the
547 data. RMH provided advice on the analysis. PME and AB contributed to the final manuscript.

548

549 **Competing Interests**

550 The authors have no conflict of interests.

551

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782 **TABLE LEGENDS**

783

784 **Table 1:** List of the measuring instrument used in the present study.

785

786 **Table 2:** Cluster conditions for both methods.

787

788 **Table 3:** Cluster relationships between OPC and SMPS clusters.

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790

791 **FIGURE LEGENDS**

792

793 **Figure 1:** Map of the site (Map by ©HERE).

794

795 **Figure 2:** Frequency and diurnal occurrence of the clusters formed by the OPC data.

796

797 **Figure 3:** Frequency and diurnal occurrence of the clusters formed by the SMPS data.

798

799 **Figure 4:** Particle contributions in the range 12 – 550 nm, for the clusters formed using
800 the OPC data (top) and the SMPS data (bottom).

801

802 **Figure 5:** Particle contributions in the range up to 10 μm , for the clusters formed using
803 the OPC data (top) and the SMPS data (bottom).

804

805 **Figure 6:** PM_{10} polar plots of the clusters formed by the OPC data.

806

807 **Figure 7:** Cluster percentage contribution per week (week number refers to week of
808 year 2020).

809



810 **Table 1: List of the measuring instrument used in the present study.**

811

Monitoring	Model	Manufacturer	Regulatory grade	Approximate cost (£)
NO ₂	NO2-B43F	Alphasense	No	250
O _x	Ox-B43I	Alphasense	No	160
Black Carbon	MA200	Aethlabs	No	5,700
Lung Deposited Surface Area		Naneos	No	8,500
OPC	OPC-N3	Alphasense	No	250
SMPS	TSi3082	TSi	Yes	80,000
ACSM	Quad - ACSM	Aerodyne	Yes	170,000
PM	Fidas 200E	Palas	Yes	25,000
NO ₂	T500U	Teledyne	Yes	15,000
Black Carbon	AE33 Aethalometer	Magee Scientific	Yes	25,000
O ₃	49i	Thermo	Yes	3,000

812

813



Table 2: Cluster conditions for both methods.

	NO ₂ (ppb)	BC (ng m ⁻³)	PM ₁ (µg m ⁻³)	PM _{2.5} (µg m ⁻³)	PM ₁₀ (µg m ⁻³)	O ₃ (ppb)	Org (µg m ⁻³)	SO ₄ ²⁻ (µg m ⁻³)	NO ₃ ⁻ (µg m ⁻³)	LDSA ratio	RH (%)	WS (m s ⁻¹)	T (°C)
OPC.1	18.6±13.9	555±630	4.32±4.08	6.53±4.62	9.97±5.81	31.9±9.81	0.254±0.231	4.12E-02±5.42E-02	8.90E-02±1.15E-01	0.443	83.9±13.1	4.16±2.50	5.20±3.11
OPC.2	9.64±1.90	233±32.8	2.56±0.72	5.61±1.58	10.7±2.97	38.6±1.34	0.142±0.082	2.98E-02±5.67E-02	1.64E-02±5.53E-03	0.847	65.1±10.5	7.1±1.01	7.16±1.53
OPC.3	13.1±8.20	278±153	2.95±0.78	5.80±1.98	9.70±2.69	37.6±6.79	0.241±0.254	6.73E-02±6.25E-02	8.41E-02±1.54E-01	0.830	91.8±8.73	3.47±1.11	4.60±1.95
OPC.4	11.5±7.15	281±191	2.51±1.55	4.84±3.20	8.33±5.35	36.5±5.17	0.192±0.235	4.53E-02±6.62E-02	1.08E-01±2.53E-01	0.536	83.5±11.5	4.37±2.09	6.26±2.73
OPC.5	18.3±16.3	659±879	6.27±6.56	9.10±7.18	13.3±8.37	31.5±11.9	0.338±0.558	4.10E-02±6.49E-02	1.31E-01±2.62E-01	0.417	82.6±11.5	4.38±2.50	6.68±3.31
OPC.6	8.58±6.72	197±155	2.85±1.12	5.96±2.51	10.3±4.30	40.0±4.69	0.116±0.152	3.50E-02±5.08E-02	3.50E-02±1.18E-01	0.588	81.2±12.3	4.87±2.07	6.42±2.89
Average	15.9±13.7	498±673	4.53±4.93	7.11±5.61	11.0±6.94	33.6±9.95	0.252±0.403	4.19E-02±6.05E-02	1.00E-01±2.08E-01	0.499	83.1±12.3	4.37±2.37	6.05±3.11

	NO ₂ (ppb)	BC (ng m ⁻³)	PM ₁ (µg m ⁻³)	PM _{2.5} (µg m ⁻³)	PM ₁₀ (µg m ⁻³)	O ₃ (ppb)	Org (µg m ⁻³)	SO ₄ ²⁻ (µg m ⁻³)	NO ₃ ⁻ (µg m ⁻³)	LDSA ratio	RH (%)	WS (m s ⁻¹)	T (°C)
SMPS.1	16.0±14.9	485±852	3.35±2.64	5.70±3.89	9.52±6.05	32.2±10.3	0.215±0.300	3.06E-02±4.80E-02	5.47E-02±7.76E-02	0.331	85.1±10.7	4.1±2.70	5.53±3.06
SMPS.2	16.8±12.0	406±539	2.70±1.57	5.11±2.33	8.91±3.75	32.9±8.10	0.132±0.156	2.53E-02±4.11E-02	2.56E-02±4.31E-02	0.501	83.2±9.71	3.74±1.67	4.64±2.86
SMPS.3	4.38±2.91	88.1±62.2	2.64±1.62	5.57±3.62	9.26±5.87	41.6±3.24	0.062±0.063	3.74E-02±5.75E-02	2.07E-02±7.15E-02	0.555	80.1±8.93	7.19±2.48	7.43±2.72
SMPS.4	14.3±12.3	452±592	3.77±2.56	6.71±3.75	11.1±5.67	35.6±9.32	0.249±0.306	4.68E-02±6.27E-02	8.12E-02±1.53E-01	0.499	79.4±13.9	4.74±2.38	6.97±2.62
SMPS.5	29.8±17.2	1389±838	17.95±7.89	21.1±8.08	25.1±7.95	16.1±10.6	1.066±0.562	1.41E-01±7.58E-02	5.74E-01±3.60E-01	0.833	93.9±7.49	2.6±1.63	4.90±2.94
SMPS.6	13.2±10.8	340±395	2.68±1.58	5.23±3.12	9.12±5.42	36.0±6.54	0.164±0.189	2.93E-02±4.31E-02	3.86E-02±7.17E-02	0.467	81.0±12.7	4.73±2.11	6.1±3.11
Average	15.1±13.2	460±649	4.12±4.72	6.78±5.48	10.8±6.90	33.8±9.84	0.280±0.403	4.61E-02±6.40E-02	1.07E-02±2.23E-01	0.499	82.8±12.4	4.41±2.42	5.95±2.99



Table 3: Cluster relationships between OPC and SMPS clusters.

OPC/SMPS	SMPS.1	SMPS.2	SMPS.3	SMPS.4	SMPS.5	SMPS.6	Total OPC
OPC.1	48	30	9	71	13	66	237
OPC.2	1	3		5		3	12
OPC.3		15		2	4	2	23
OPC.4	25	27	6	52	19	50	179
OPC.5	24	26	17	39	40	38	184
OPC.6	7	25	9	28	3	25	97
Total SMPS	105	126	41	197	79	184	732

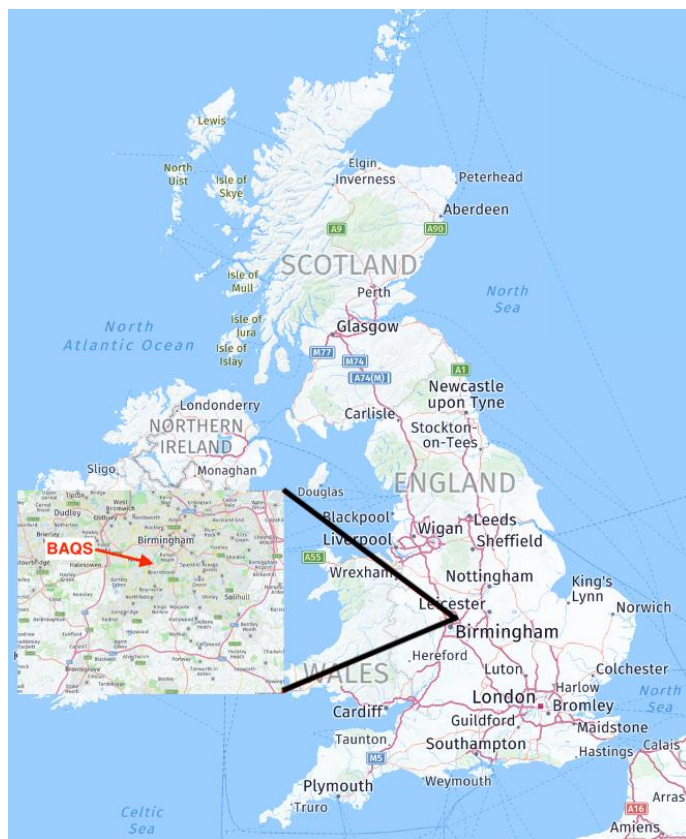


Figure 1: Map of the location of the Birmingham Air Quality Supersite (BAQS) site in the U.K. (Map by ©HERE).

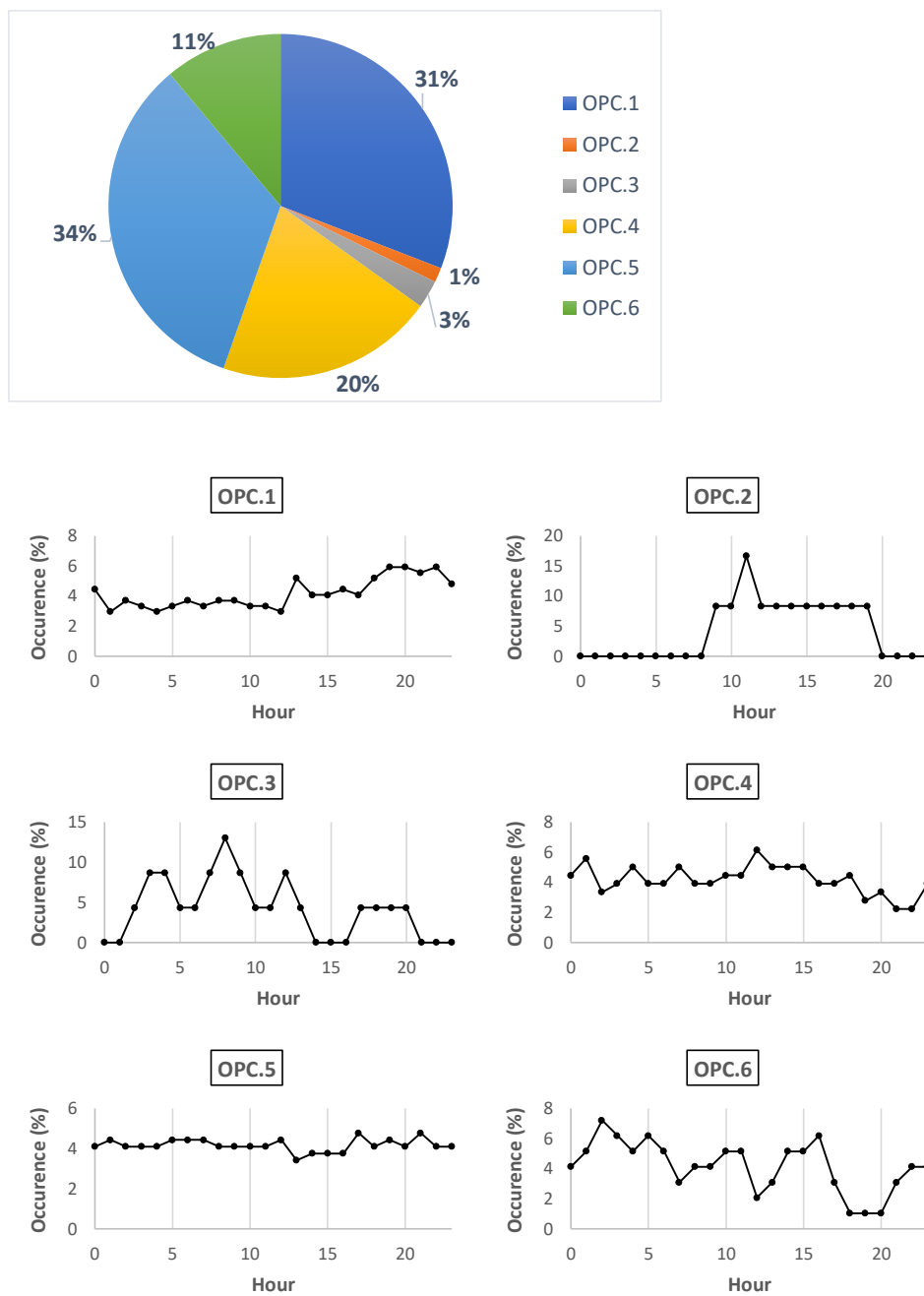


Figure 2: Frequency and diurnal occurrence of the clusters formed by the OPC data.

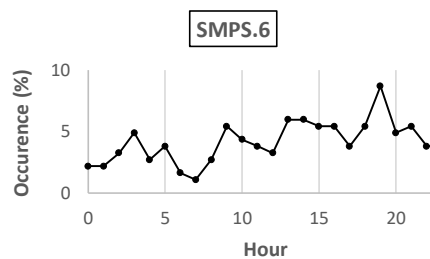
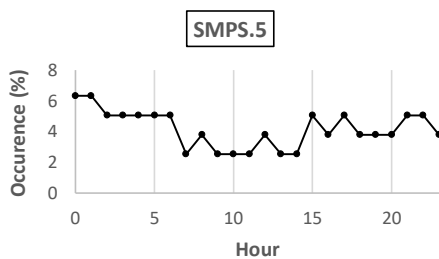
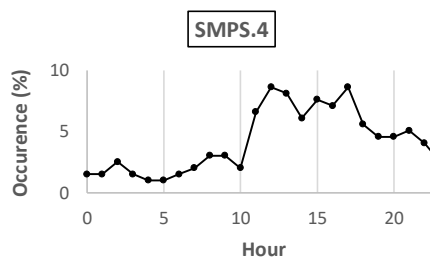
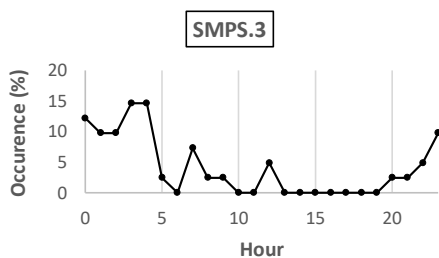
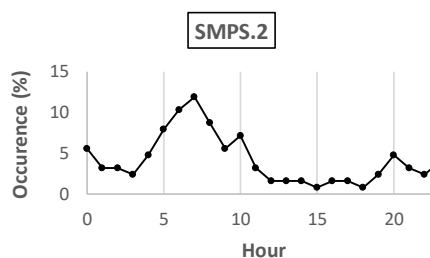
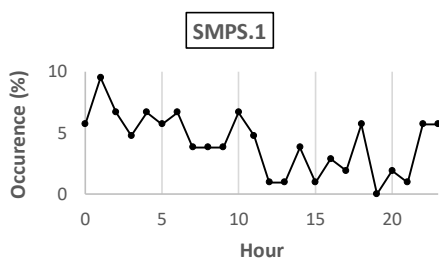
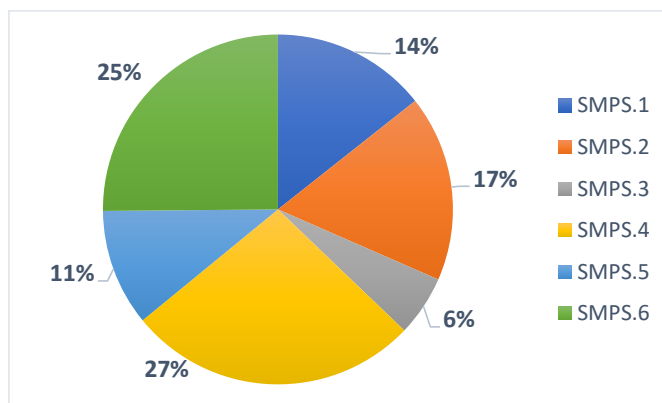


Figure 3: Frequency and diurnal occurrence of the clusters formed by the SMPS data.

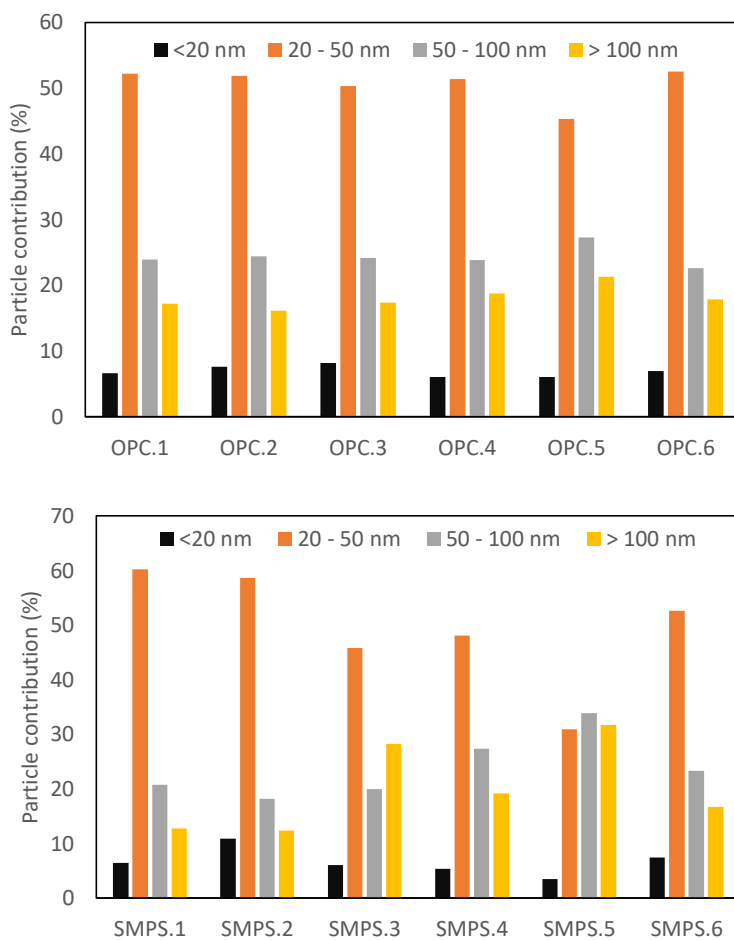


Figure 4: Particle contributions in the range 12 – 550 nm, for the clusters formed using the OPC data (top) and the SMPS data (bottom).

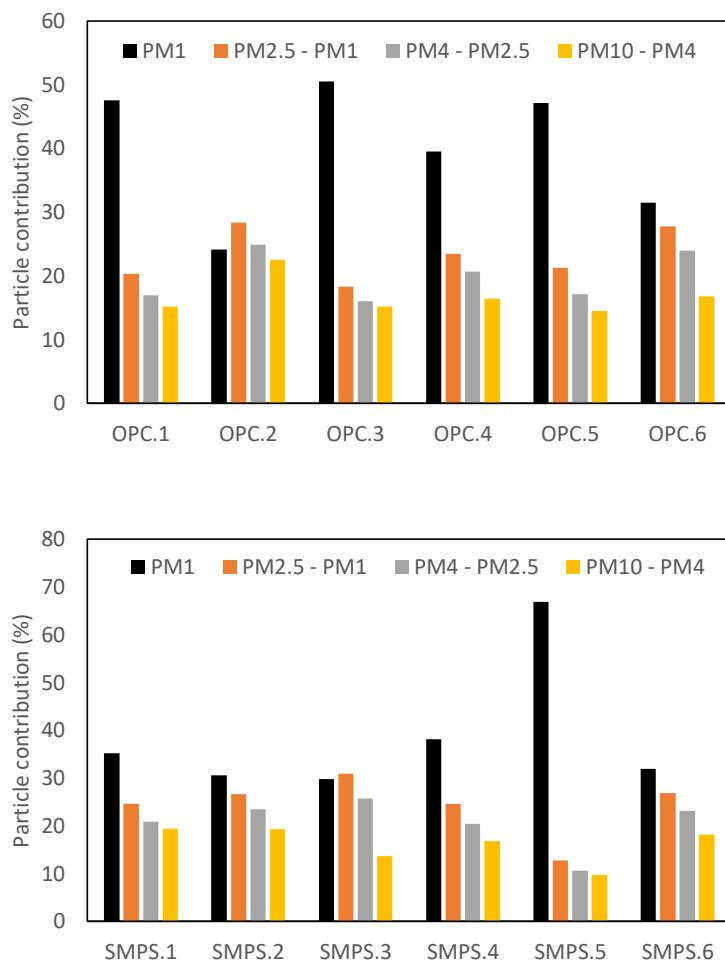


Figure 5: Particle contributions up to 10 μm , for the clusters formed using the OPC data (top) and the SMPS data (bottom).

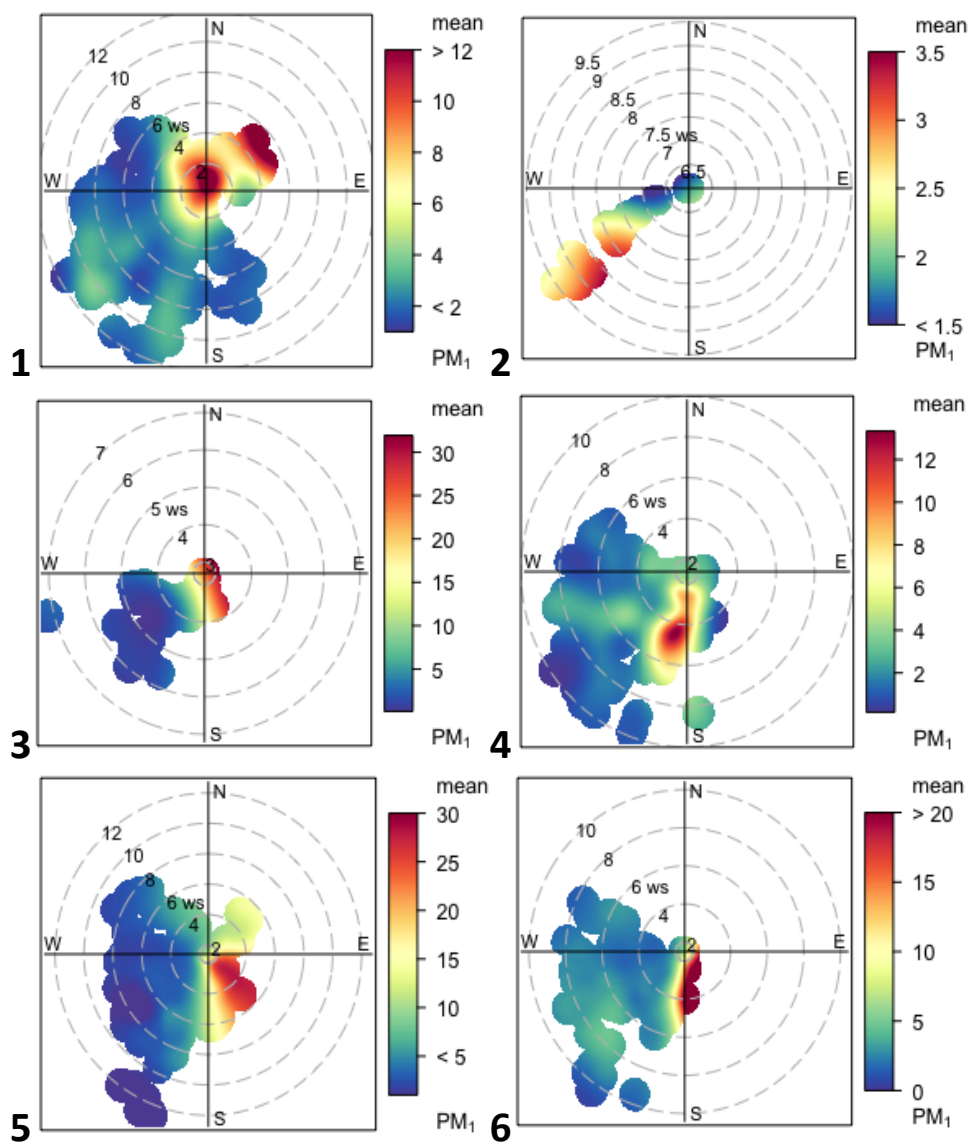


Figure 6: PM_{10} polar plots of the clusters formed by the OPC data.

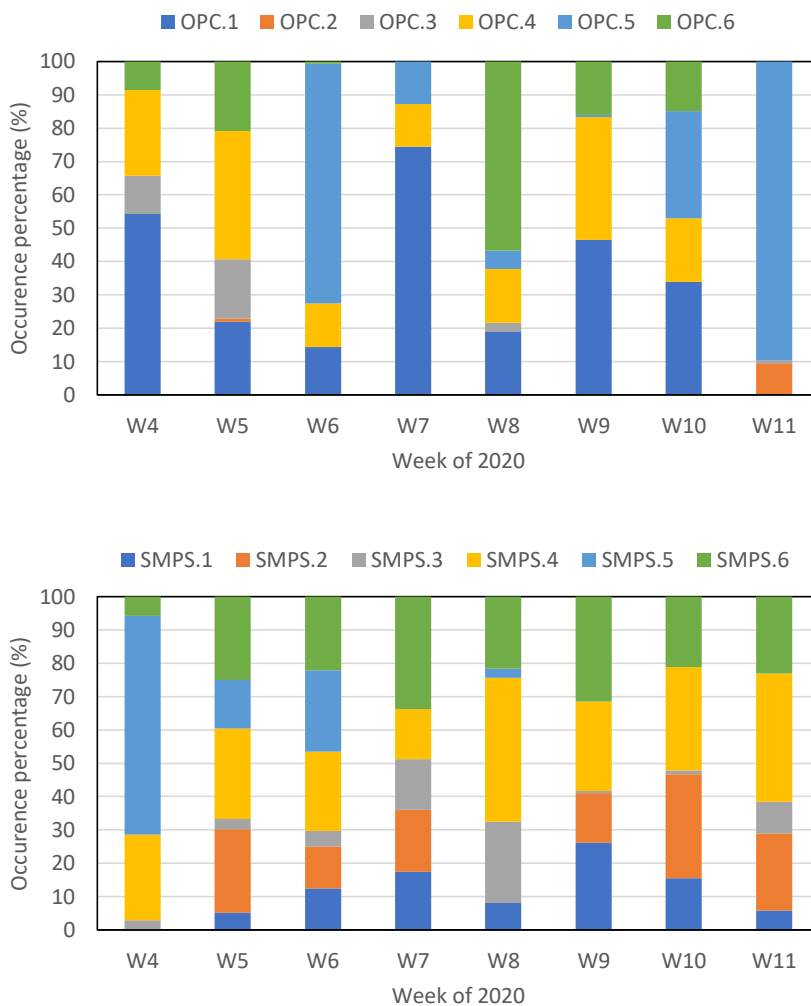


Figure 7: Cluster percentage contribution per week (week number refers to week of year 2020).