



Supplement of

Detection of ship plumes from residual fuel operation in emission control areas using single-particle mass spectrometry

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Chemical signatures of main particle classes

The mass spectral signatures of all 300 inspected clusters as well as time series of their particle counts are provided in Figs. S5 and S6, the attribution to general classes is documented by Table S1.

Six general particle types were identified. Such groups have variations among their clusters, but similar signatures of the same overall chemical species as well as comparable temporal trends and size distributions. The labelling scheme reflects the most intense peaks and characteristic species for the respective general particle type and is frequently used in the literature (Ault et al., 2010; Decesari et al., 2014; Dall'Osto et al., 2016; Arndt et al., 2017). Their particle number and aerodynamic size can be found in Fig. S2(a), whereas average mass spectra of anions and cations are shown in Fig. S2(b) and (c), respectively. Carbon-containing particles contributed the majority to total particle numbers. Their spectra are either dominated by strong carbon cluster peaks from elemental carbon (EC), or by molecular fragments from organic carbon (OC). The balance between these signatures indicate the EC/OC ratio (Ferge et al., 2006; Spencer and Prather, 2006), however, in the present study this ratio is a continuum, broken into several small clusters by ART-2a (Zhou et al., 2006) and manually merged according to their dominant signals.

The EC-OC particle class shows no distinct K^+ peak, indicating fossil fuel combustion (oil burning or traffic) as most likely source. It has been shown that secondary material increases the particle's hygroscopicity (Moffet et al., 2008)(Ault et al., 2010; Decesari et al., 2014), suppressing the formation of negative ions (Neubauer et al., 1998; Hatch et al., 2014). The strong sulfate signal and the frequent absence of negative carbon clusters indicate condensation of sulfate during atmospheric transport, while its dominance over nitrate can result from processing in marine environment (Ault et al., 2010; Köllner et al., 2017), but also from the summer weather conditions due to the higher volatility of ammonium nitrate compared to ammonium sulfate (Querol et al., 2009; Arndt et al., 2017).

Particles with molecular fragment signals dominating over the carbon clusters were assigned to the OC-EC-nit class. They also show a pronounced K^+ peak and nitrogen-containing signals; both can be attributed to a higher contribution of biomass combustion (Silva et al., 1999; Pagels et al., 2013).

In the K-CN class, K^+ signals dominate the cation mass spectra, a well-documented signature for aerosols from biomass burning and wood combustion (Silva et al., 1999; Zhang et al., 2013). Potassium has a low ionization energy and the ion is energetically preferred compared to ions of other species, thus it survives collisions in the particle plume, when other ions are neutralized (Reinard and Johnston, 2008). The mass spectra of negative ions show CN^- and CNO^- signals from nitrogen-containing organic compounds (Silva et al., 1999; Köllner et al., 2017). A particle class similar to the K-CN particles, but with higher peaks from NO_2^- and NO_3^- indicate a strong contribution of secondary material, including nitrate in addition to the sulfate.

Sea salt particles are typically larger and produce characteristic signatures. Fresh sea salt particles are characterized by sodium ions (e.g. $^{23}Na^+$, $^{46}NaC^+$, $^{62}Na_2O^+$, $^{63}Na_2OH^+$), K^+ , $^{16}O^-$ and $^{35,37}Cl^-$. (Dall'Osto et al., 2004; Murphy et al., 2019). Chlorine is replaced by nitrate during atmospheric processing (Gard et al., 1998), thus the strong nitrate signals and weak chlorine peaks ($^{35,37}Cl^-$) suggest that these particles are not fresh but have been transported over some distance.

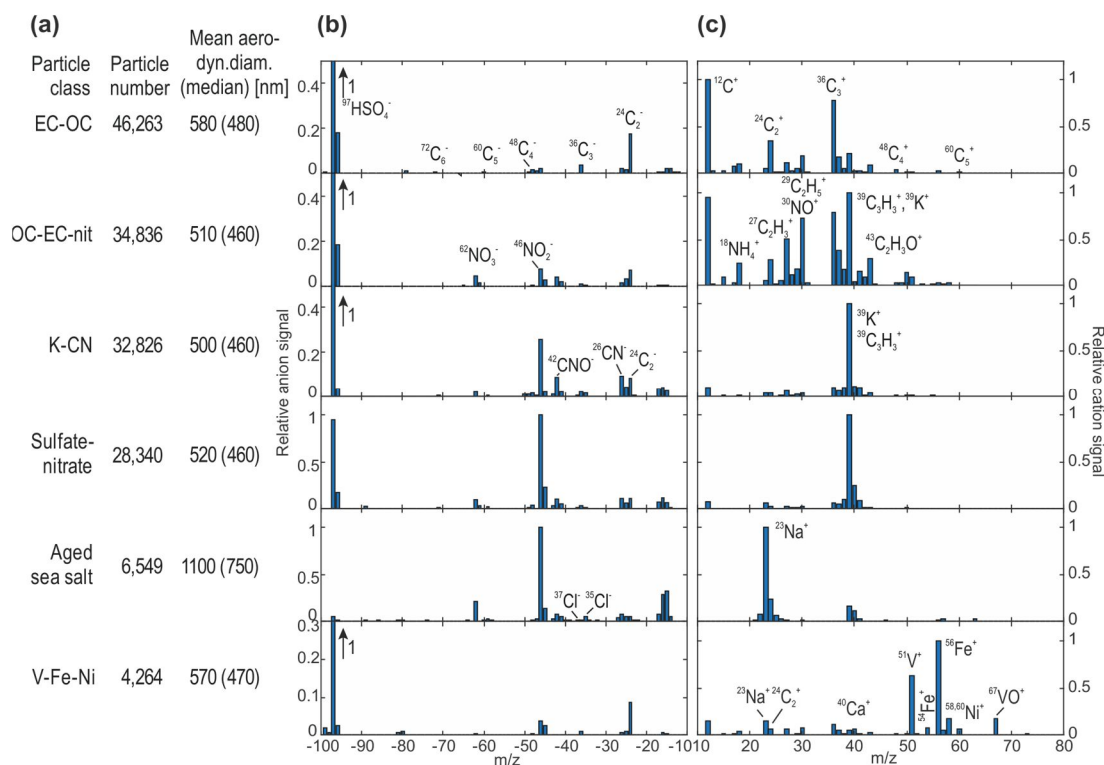


Figure S1: (a) General particle classes, their respective particle numbers and aerodynamic size. (b) Average anion mass spectra and (c) cation mass spectra of the particle classes. The mass spectra of the top 300 particle clusters from ART-2a analysis are shown in the Supplement and their assignment to the general classes is documented by Table S1.

Temporal signatures of main particle classes

Figure S2 (Fig. 3 in the manuscript) shows the time evolution of the six main particle classes with 10 minutes resolution.

The EC-OC particle numbers (dark grey area) exhibit a weak diurnal oscillation, however, not with enhanced levels during the morning and afternoon as expected from increased local traffic and human activity. In contrast, they follow the changes in wind direction and speed. It is conceivable that the northern winds transport local emissions from the city center to the sampling site, however, the strong sulfate peaks in the EC-OC group and the lack of negative ions point on aged particles that might have been transported over larger distances, see Fig. S1. Also the particle size is larger than typical for urban traffic emissions (Dall'Osto et al., 2016) which is in agreement with the assumption that these freshly emitted particles are underrepresented compared to the larger ones that are enriched by the aerosol concentrator.

The OC-EC-nit, K-Cn and Sulfate-nitrate particles roughly follow the trend of the EC-OC particles, but show an additional diurnal variation that is especially pronounced during the last two days of the measurements. Their particle numbers increase after sunset and drop in the morning, reflecting condensation of semi-volatile components at night. This temporal behavior is comparable for OC-EC-nit, K-CN and Sulfate-Nitrate particles, because: (I) They are all composed of organic matter to some extent. (II) They can simultaneously grow into the efficiently detected size mode by condensation of secondary material, and (III) all possible diurnal features of aerosol chemistry, such as daytime photochemical formation of SOA or increases in nitrate with relative humidity (Salcedo et al., 2006; Dall'Osto et al., 2009; Healy et al., 2012) are interfered by the strong land/onshore circulation. This dominance of air circulation over local atmospheric chemistry is also reflected by the pronounced particle number maxima during the last nights of the measurement period, where local terrestrial air masses contribute. In contrast, during the second and third night, the air trajectories passed over less land before entering the site and the maxima are substantially smaller. Also the OC-EC-nit and K-CN particle numbers follow the diurnal trend, probably because they grow by night-time condensation into the size mode that is enriched by the aerosol concentrator, while local emissions of smaller particles are barely detected. However, a small transient feature of K-CN particles can be noticed at the 29th June at 8 pm, before the general increase after sunset around 10 pm. It might be associated with local emissions, e.g. from barbecuing. The enhanced contributions of aged sea salt at the 30th June can be attributed to stronger winds in the central Baltic Sea.

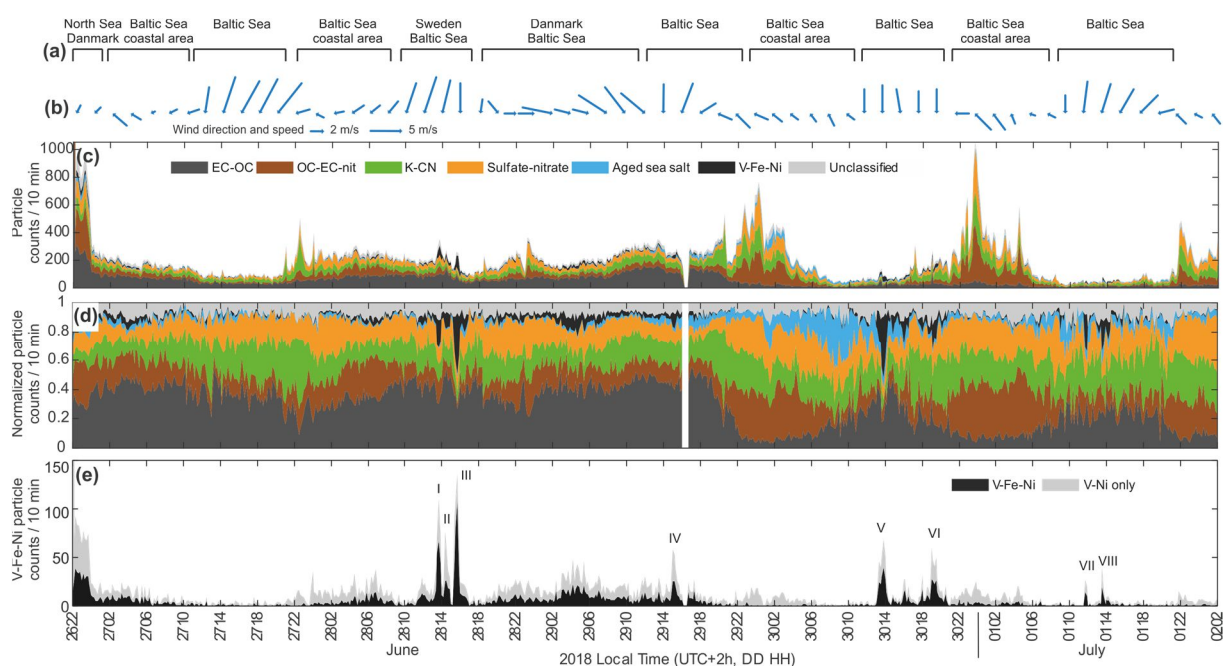


Figure S2 (Fig. 3 in the manuscript): (a) Air mass origin (top row: >12 h, bottom row <12 h) according to the HYSPLIT back trajectory analysis (Fig. S2 and S3). (b) Measured wind data from the meteorological station at the harbor exit, 12 km north of the measurement site. (c) The time series of particle counts from the general particle classes shows regional/long-range transported air pollution (26–29 June) and night-time secondary organic aerosol formation (29 June – 02 July). (d) The same data as (c), but normalized to total particle counts illustrate the contribution of each particle type as well as increased sea-salt levels during the 30 June. (e) The temporal behavior of V-Fe-Ni particles from residual fuel combustion reveals transient events and smooth background signals, predominantly during onshore winds. Apart from the short events, their contribution to total particle numbers is low.

Experiments on the research ship engine

To provide an estimate of the Fe resonance effect on particle detection in a ship plume, we conducted on-line experiments using a one-cylinder four-stroke, common rail injection diesel research ship engine with 75 kW maximum power, installed at the “Institute of Piston Machines and Internal Combustion Engines”, University of Rostock, Germany (Streibel et al., 2017). Aerosols were sampled from the hot flue gas and mixed into a stream of 5 m³/min outdoor ambient air in a home-built dilution tunnel of 3 m length and 15 cm diameter. 1 l/min of the diluted and cooled aerosol was guided to the SPMS instrument. We compared our KrF-excimer laser at 248 nm inducing the Fe resonance with the most common laser system in SPMS, the Nd:YAG at 266 nm, both operated with a pulse duration of about 5 ns, the same pulse energy of 1.6 mJ. The focal lens (f=200 mm) was adjusted to maintain a comparable spot area, respective intensity for both wavelengths.

Key idea to mimic ambient aerosols containing contributions from ships after atmospheric transport is the use of diesel fuel during measurements and previous engine operation with bunker fuel to generate particles with small impurities from bunker fuel by carryover effects.

The on-line experiments were conducted at 75 kW (100%) load after a sufficient warm-up time and a 30 min run-in period at full load. Measurements with the Nd:YAG laser were performed 15 total operation hours after switching to diesel fuel, in which the engine consumed 65 kg diesel fuel for an energy output of 225 kWh. For the KrF-excimer laser, the experiment was repeated on the next day without switching back to bunker oil. At this time, the engine was run for 22 hours on diesel, with a total fuel consumption of 123 kg and 441 kWh energy output since switching from bunker oil to diesel.

A single-particle investigation reveals that many more particles show clear signatures of the residuals V, Ni, and Fe if ionized with 248 nm, despite the longer engine operation time (22h for 248 nm vs. 15h for 266 nm) since switching from bunker fuel to diesel (Fig. S3). One explanation for the increased detection efficiency for V and Ni lies in the complex UV absorption spectra of these metals, with possibly more and stronger lines overlapping with the rather broad KrF laser spectrum compared to the Nd:YAG laser (Passig et al., 2020).

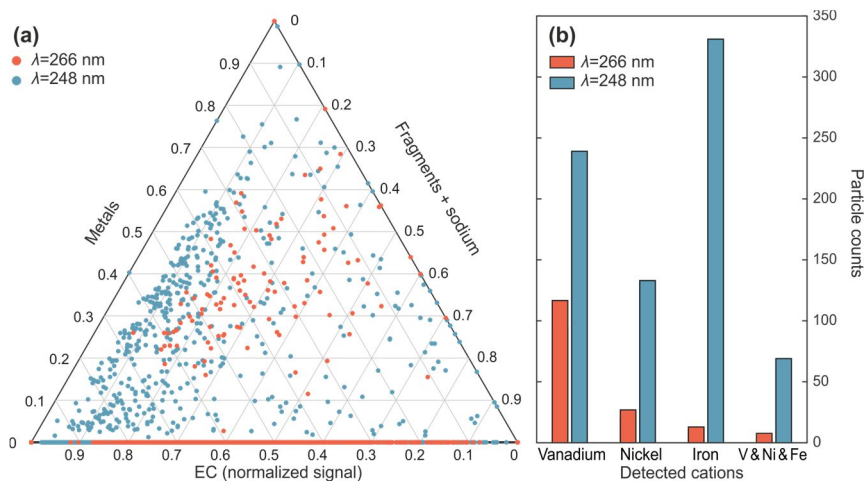
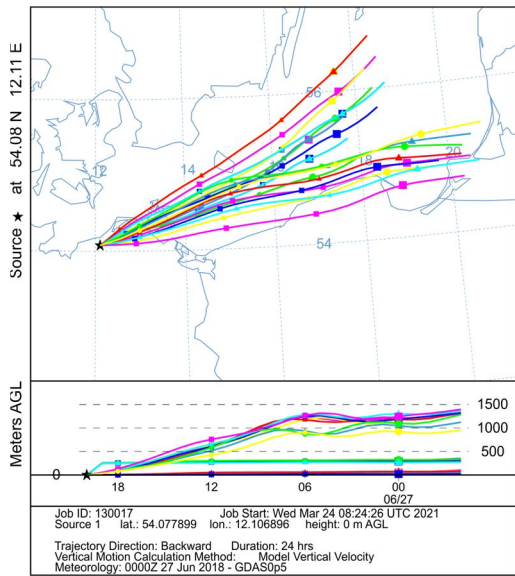
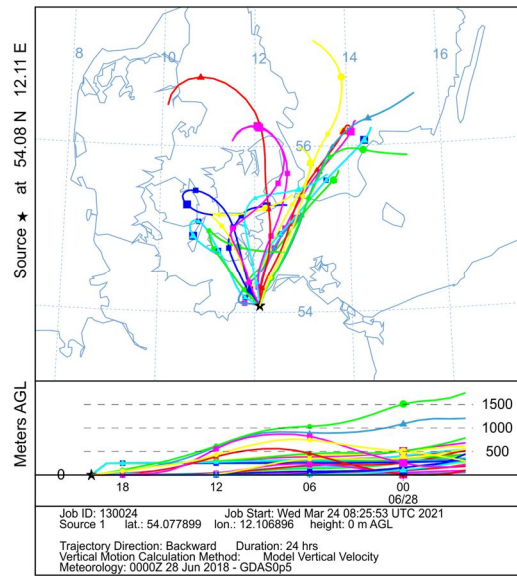


Fig. S3 Metal residues from previous bunker fuel use in ship engine particles, emitted during diesel fuel operation. (A) Ternary plot of single particle composition (each 2000 particles) indicates a minor particle fraction with high metal content (predominantly V^+) for ionization with 266 nm (red dots). Ionization with 248 nm reveals metal signatures for many more particles, also with lower concentrations (blue dots). Particles without metal signals result in dots at the bottom line. (EC signal: sum of C_3^+ , C_4^+ , C_5^+ ; fragments/sodium: sum of C^+ , Na^+ , $C_3H_3O^+$, $C_3H_7^+$; metals: $3 \times$ sum of V^+ , VO^+ , $^{54}Fe^+$, $^{58}Ni^+$). (B) From each 2000 particles, many more show clear metal signatures if ionized with 248 nm. The largest difference can be noticed for iron, a consequence of direct resonance. To reduce ambiguity, particles are counted to the respective group only if the following signals are above noise level: both V^+ and VO^+ for V, $^{54}Fe^+$ and $^{56}Fe^+$ for Fe, $^{58}Ni^+$ for Ni.

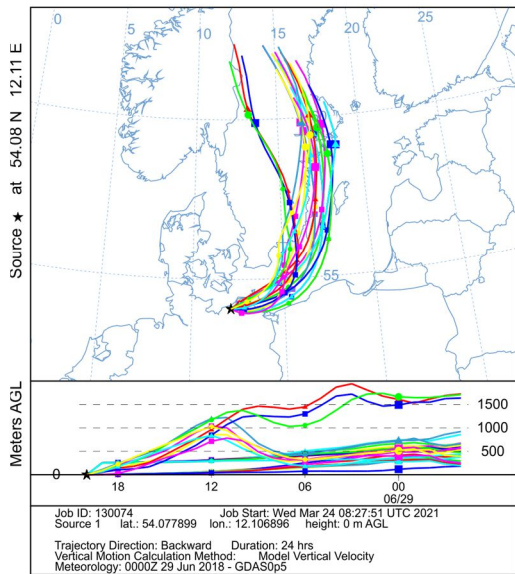
NOAA HYSPLIT MODEL
Backward trajectories ending at 2000 UTC 27 Jun 18
GFSG Meteorological Data



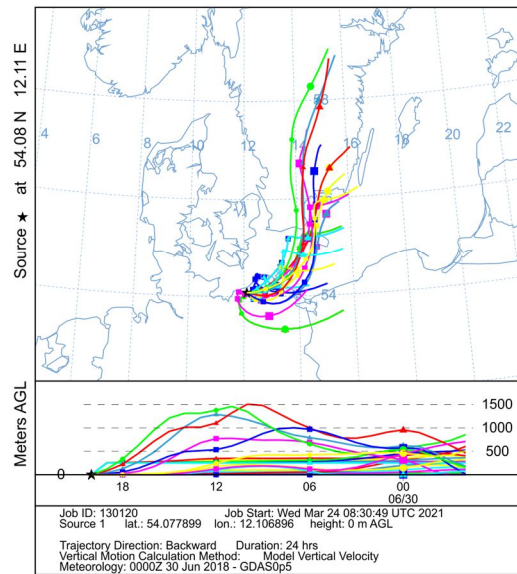
NOAA HYSPLIT MODEL
Backward trajectories ending at 2000 UTC 28 Jun 18
GFSG Meteorological Data



NOAA HYSPLIT MODEL
Backward trajectories ending at 2000 UTC 29 Jun 18
GFSG Meteorological Data



NOAA HYSPLIT MODEL
Backward trajectories ending at 2000 UTC 30 Jun 18
GFSG Meteorological Data



NOAA HYSPLIT MODEL
Backward trajectories ending at 2000 UTC 01 Jul 18
GFSG Meteorological Data

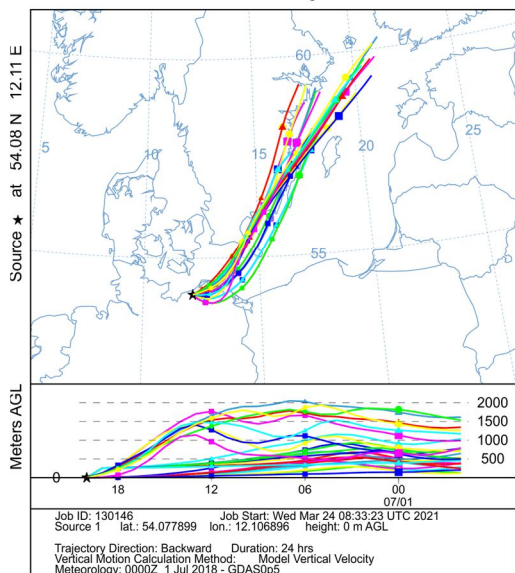


Figure S4: HYSPLIT back trajectories for 24h run time, ending at the measurement site during the experiments.

Table S1: Assignment of the top-300 clusters of the ART-2a analysis to main particle classes.

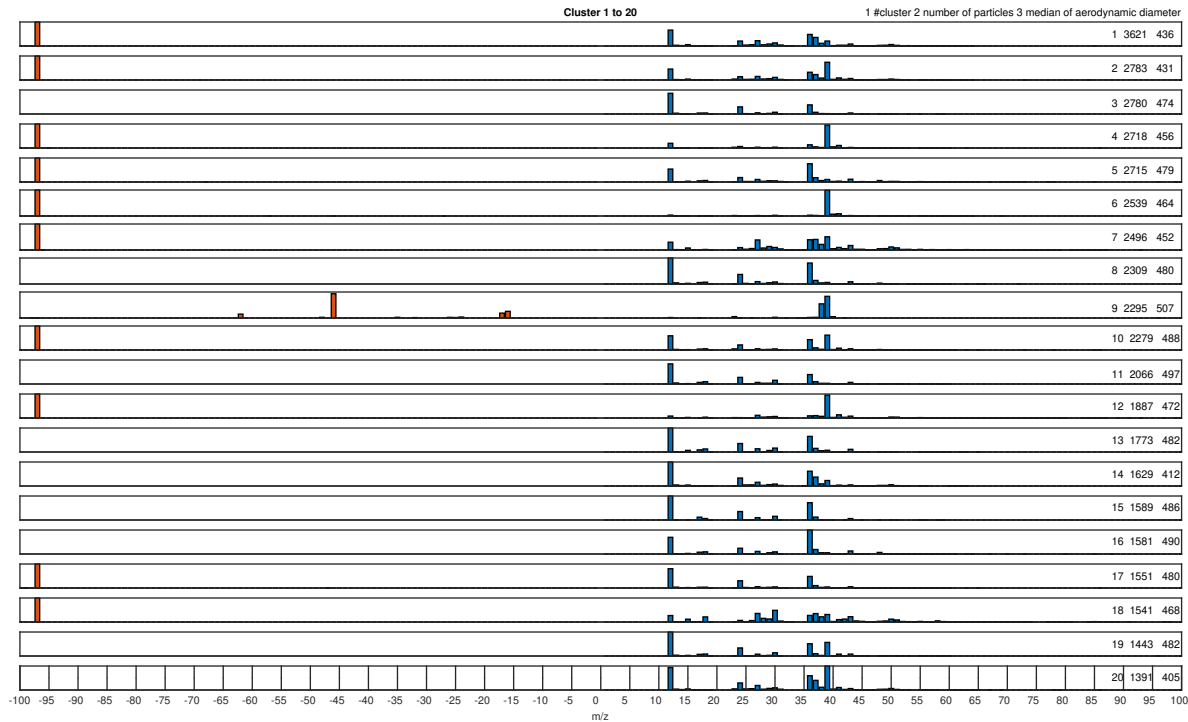
EC-OC	OC-EC-nit	Sulfate-nitrate	K-CN	aged sea salt	V-Fe-Ni	other	remarks
3	1	24	2	57	72		
5	7	30	4	80	97	43	NO+
8	14	31	6	82	110	44	NO+
10	18	32	9	106	111	45	NO+
11	20	39	12	120	127	89	Al+ Fe+ org
13	26	41	21	132	138	152	low quality
15	27	51	23	146	150	174	Ca+, large
16	28	52	34	155	151	184	Al+ org.
17	33	62	35	159	161	206	Fe+43
19	40	64	42 local	177	164	212	EC-S
22	43	69	47	180	183	222	MGO?
25	44	70	49	193	196	236	Fe+ Ni+
29	45	73	51	225	226	256	low quality
36	53	75	56	234	230	287	Fe+
37	54	95	65 local	246	250	296	Fe+
38	59	96	67 local	252	265		
44	66	100	84	253			
46	68	101	86	254			
48	74	104	90	257			
50	77	105	83	259			
55	78	112	85	260			
58	87	115	91	266			
60	88	117	92	267			
63	102	121	98	287			
71	107	124	114				
76	109	126	118				
79	113	128	122				
81	116	130	125				
93	123	131	129				
94	135	136	133				
99	139	137	140				
103	141	143	149				
108	182	147	153				
119	187	148	154				
134	199	156	157				
142	200	158	192				
144	205	160	210				
145	215	162	211				
163	216	165	213				
170	219	166	224				
171	223	167	235				
178	228	168	261				
185	229	169	282				
186	231	172	281				
190	232	173					
194	236	175					
201	238	176					
202	242	179					
204	243	181					
207	244	188					
208	247	189					
214	249	191					
218	251	195					
221	258	197					
222	264	198					
227	268	203					
233	275	209					
239	279	217					
245	283	220					
248	284	240					
255	280	241					
262	286	263					
269	289	270					
273	290	272					
276	292	274					
277	293	291					
278	297	295					
285							

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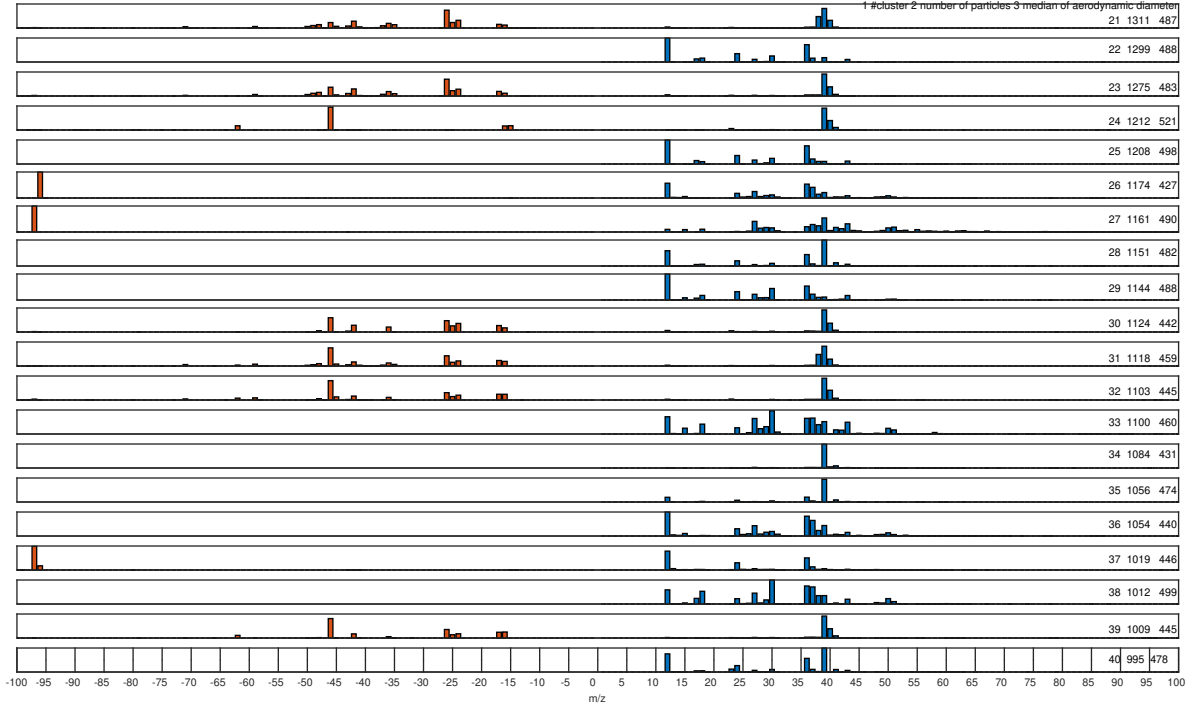
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Figure S4: Weight matrices (neg. and pos. mass spectra) of the top 300 cluster centers from the ART-2a analysis.

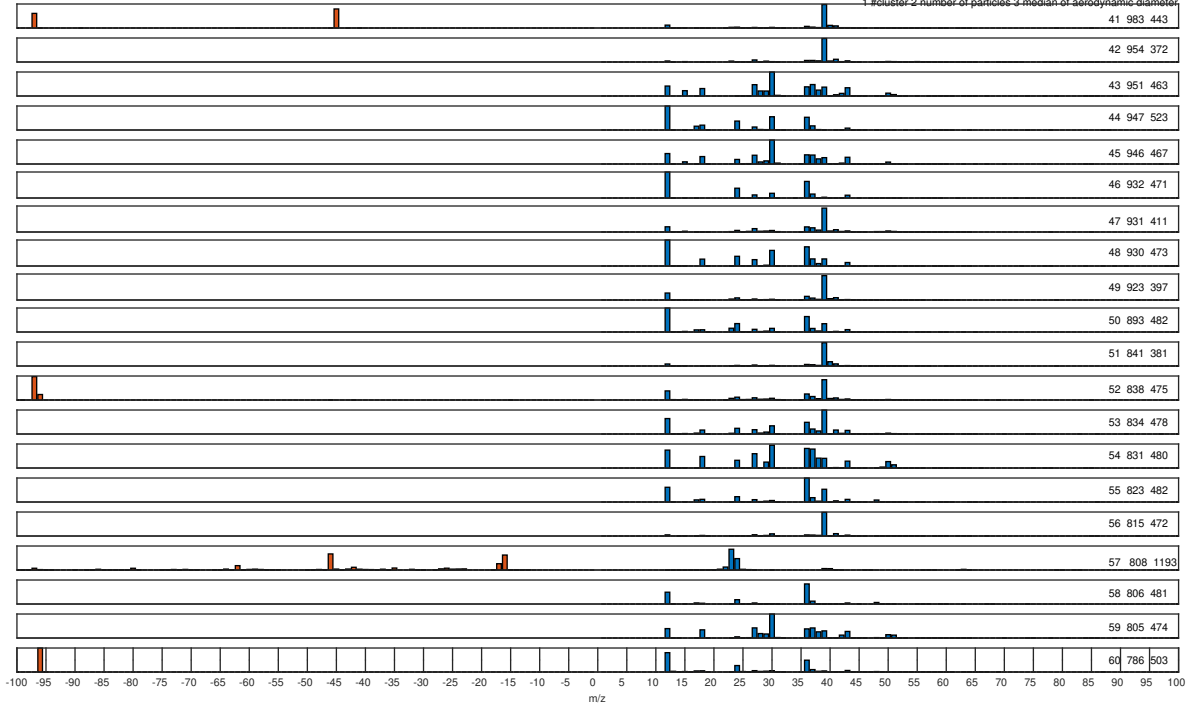


Cluster 21 to 40

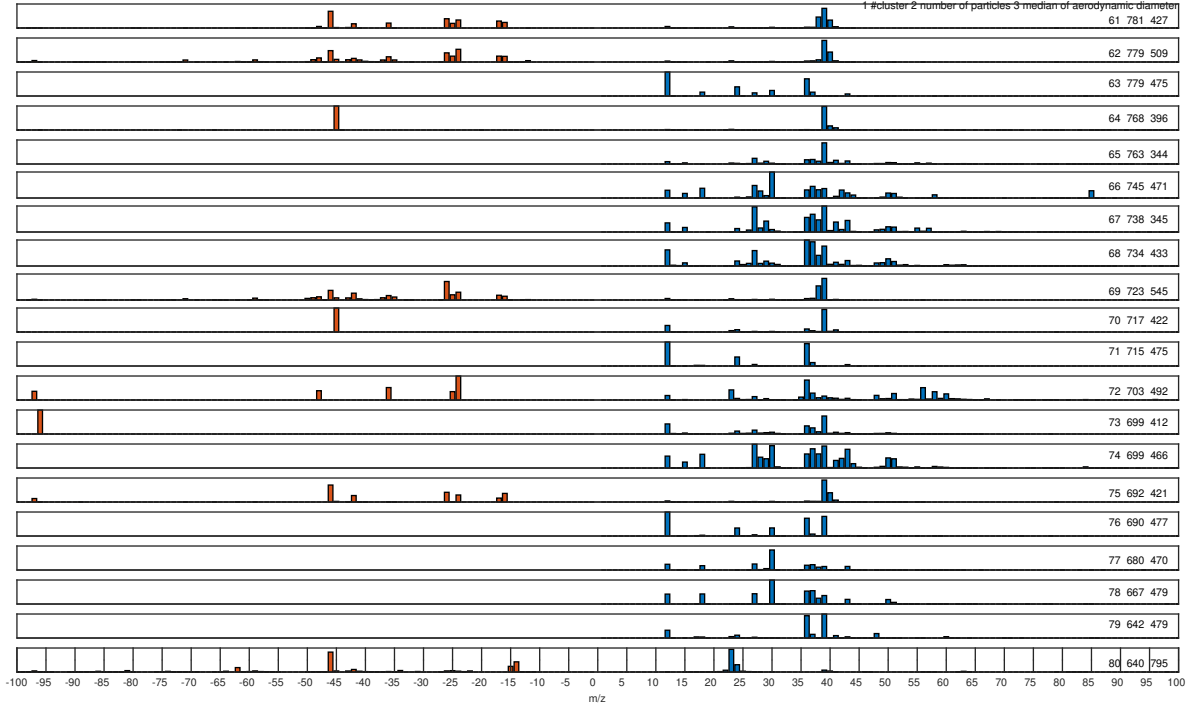
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21 1311 487

Cluster 41 to 60

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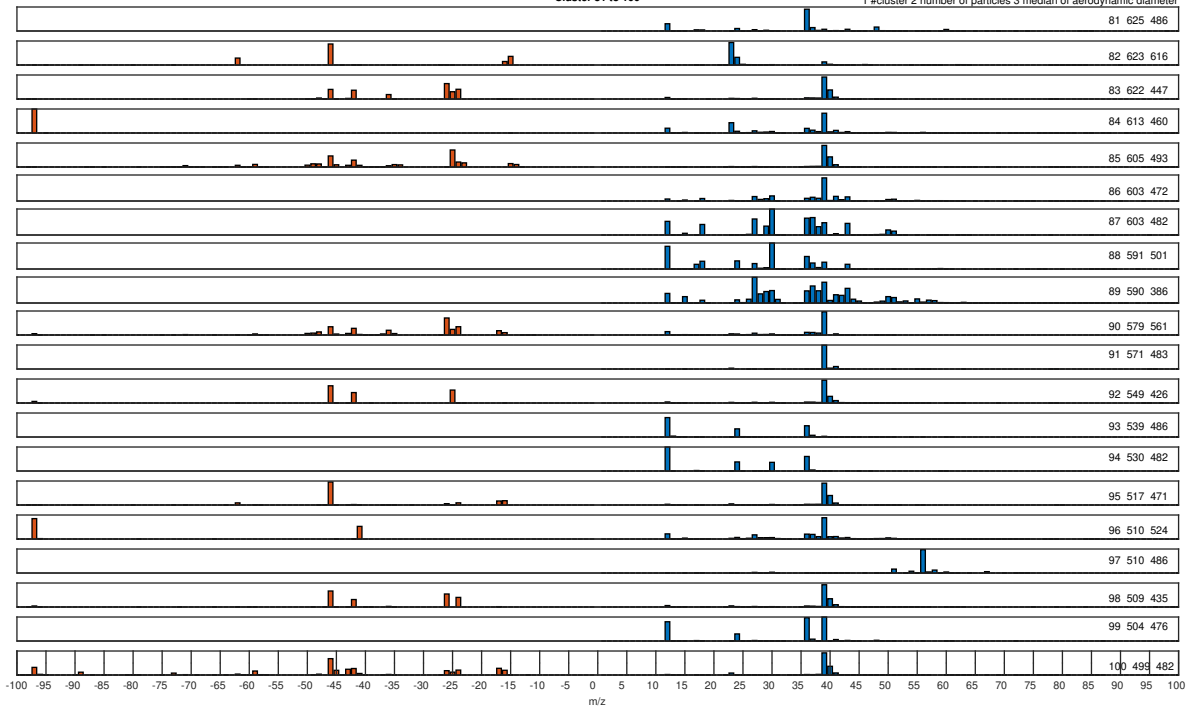


Cluster 61 to 80

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61 781 427

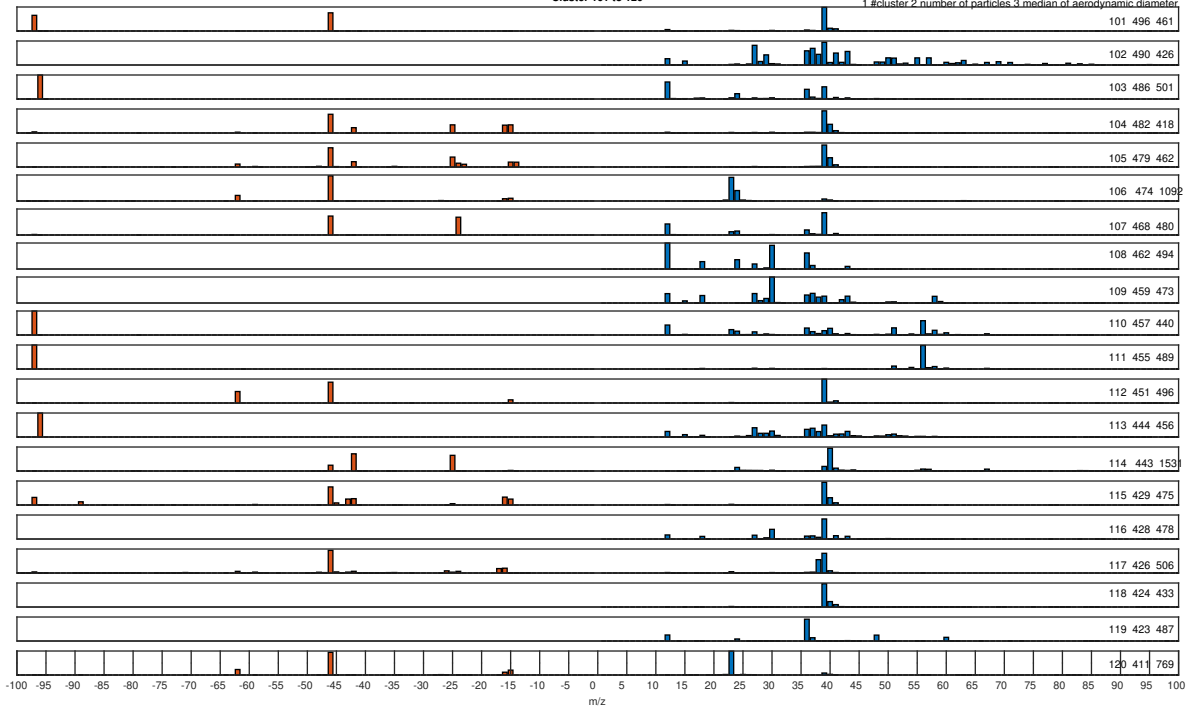
Cluster 81 to 100

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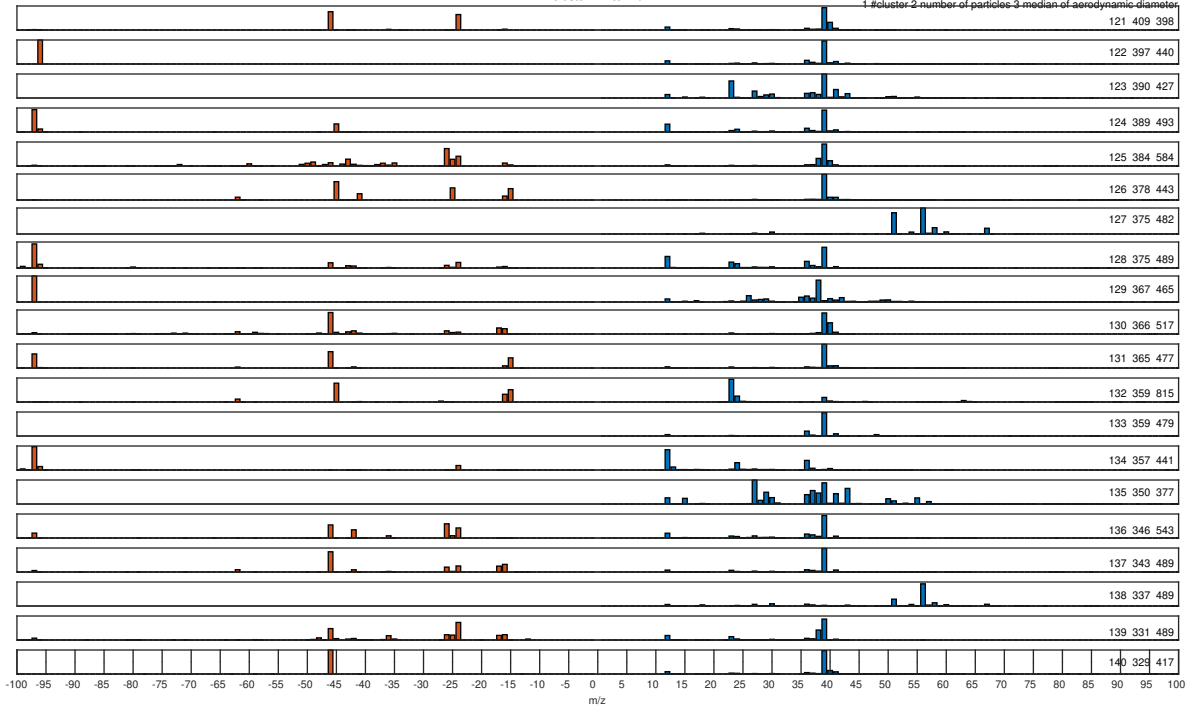
Cluster 101 to 120

1.#cluster 2.number of particles 3.median of aerodynamic diameter



Cluster 121 to 140

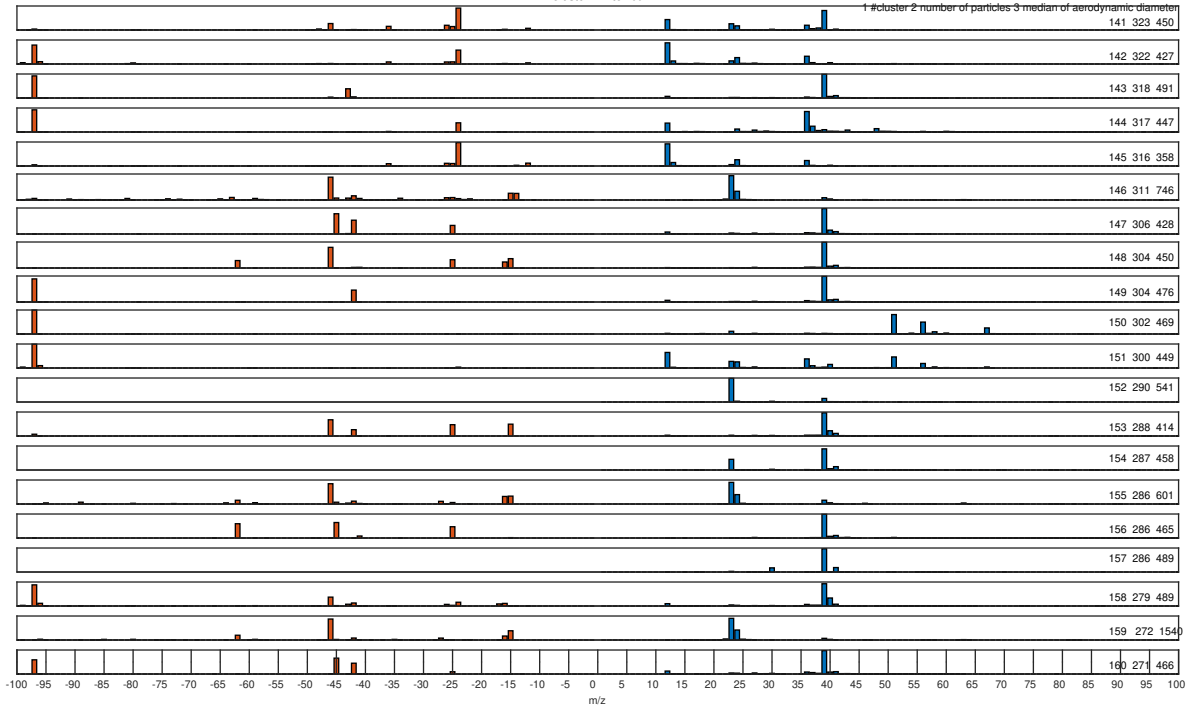
1.#cluster 2.number of particles 3.median of aerodynamic diameter



m/z

Cluster 141 to 160

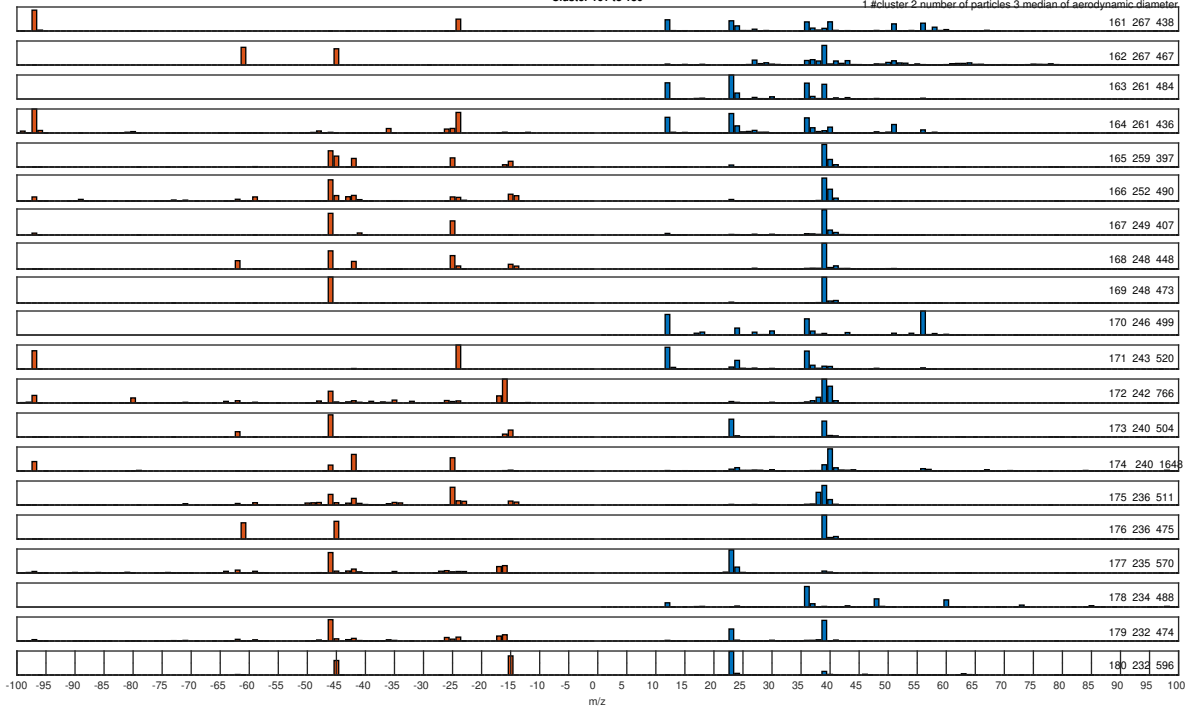
1 #cluster 2 number of particles 3 median of aerodynamic diameter



m/z

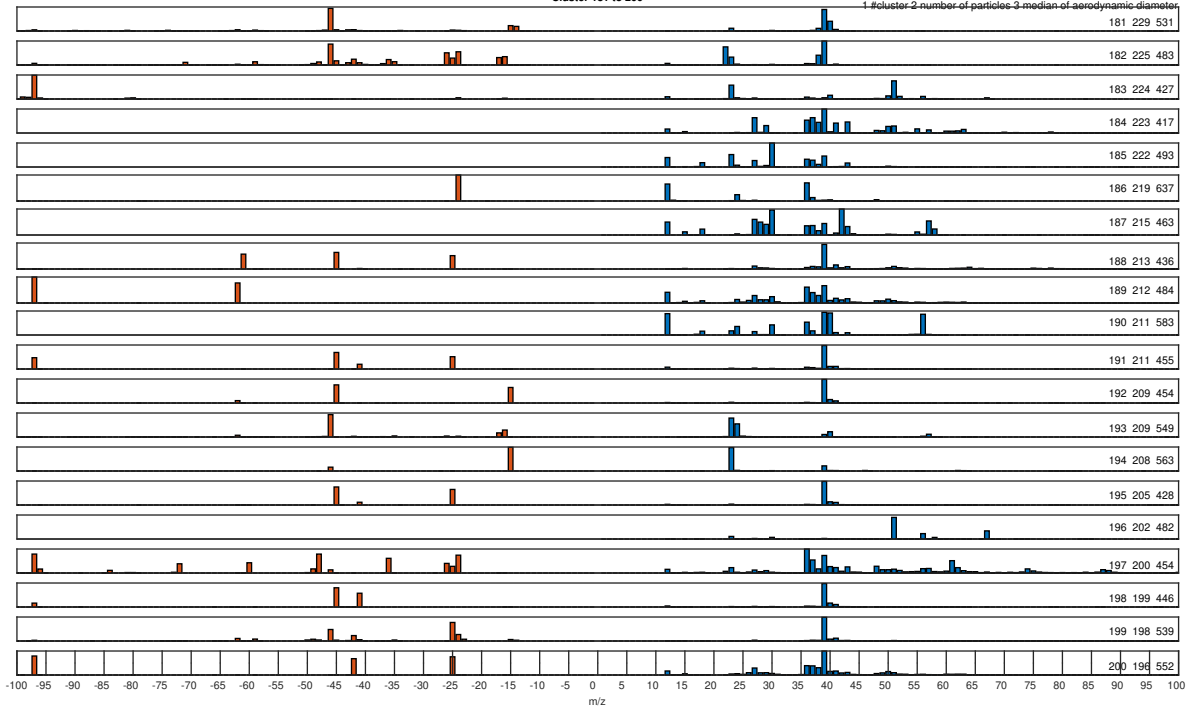
Cluster 161 to 180

1.#cluster 2.number of particles 3.median of aerodynamic diameter

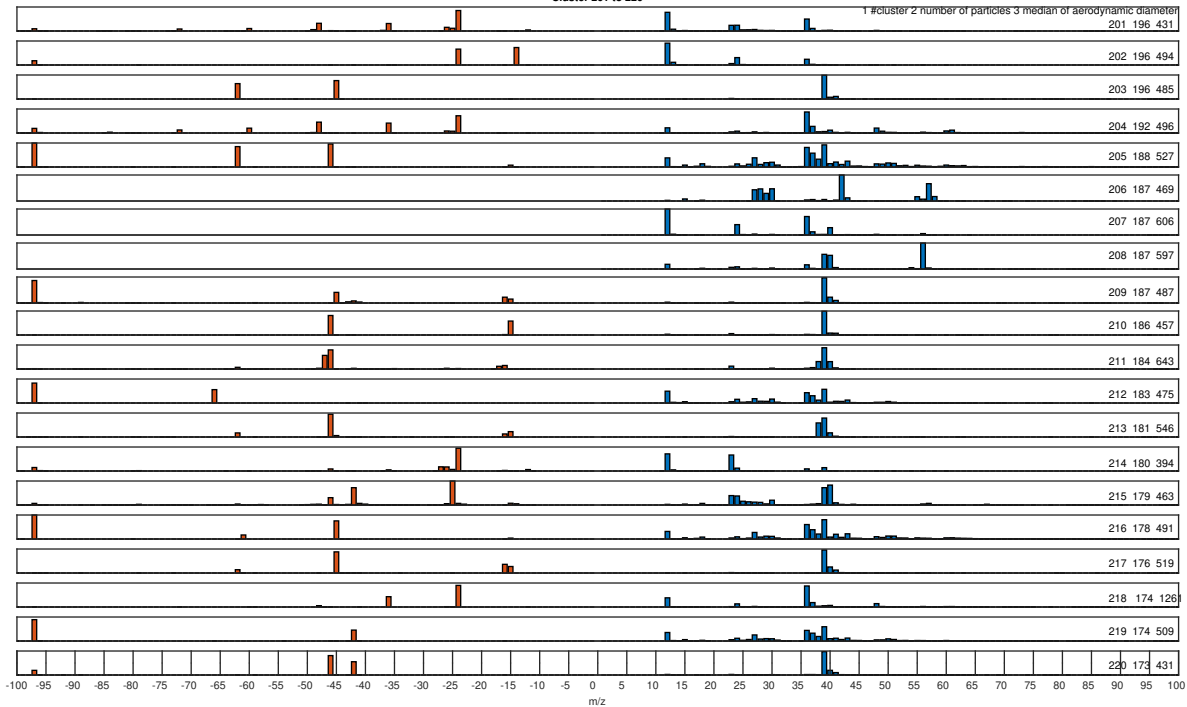


Cluster 181 to 200

1.#cluster 2.number of particles 3.median of aerodynamic diameter

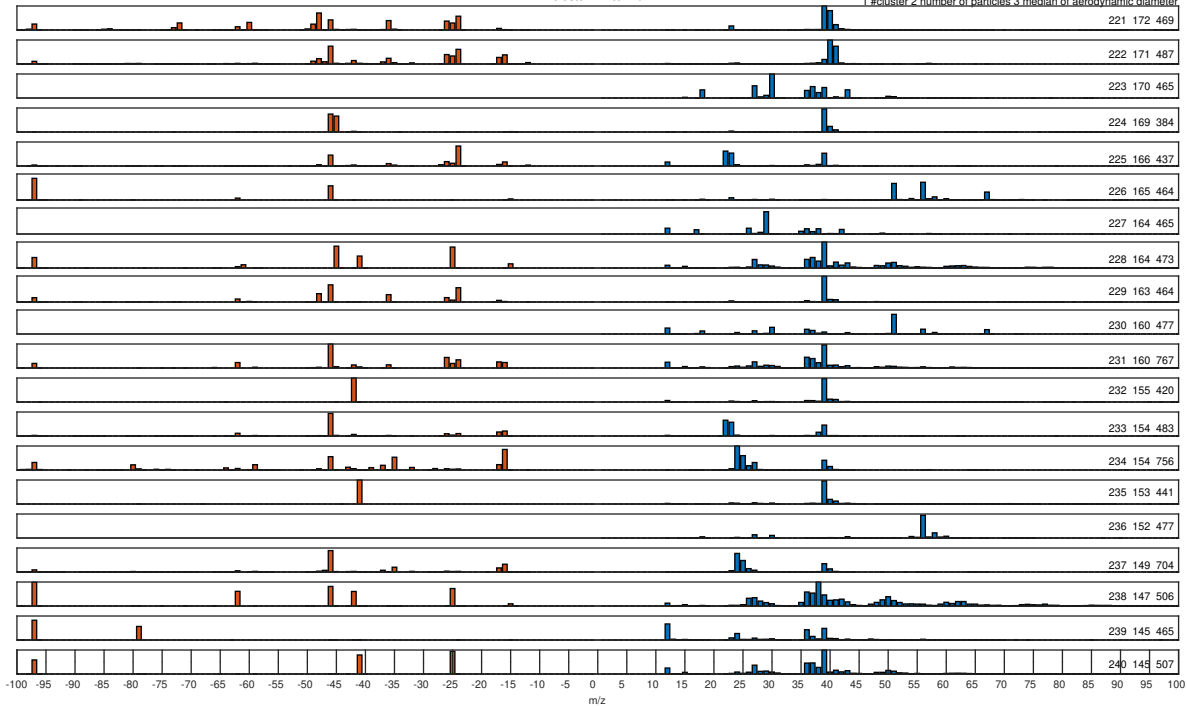


Cluster 201 to 220

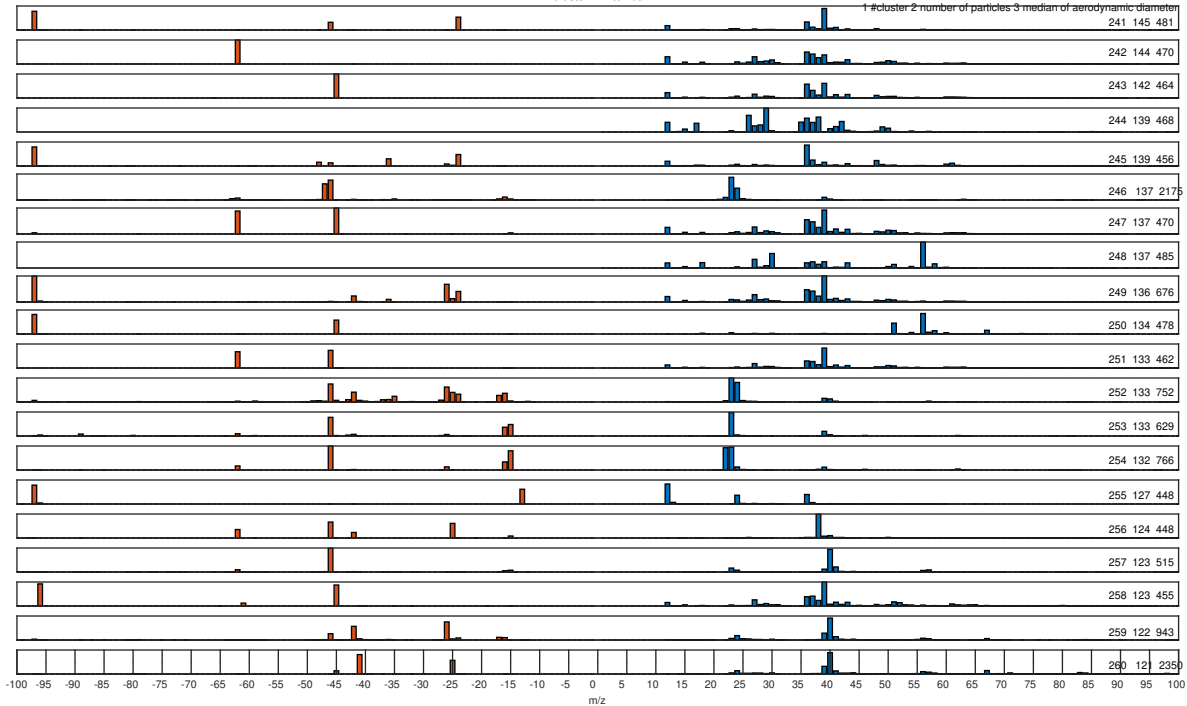


Cluster 221 to 240

1 #cluster 2 number of particles 3 median of aerodynamic diameter



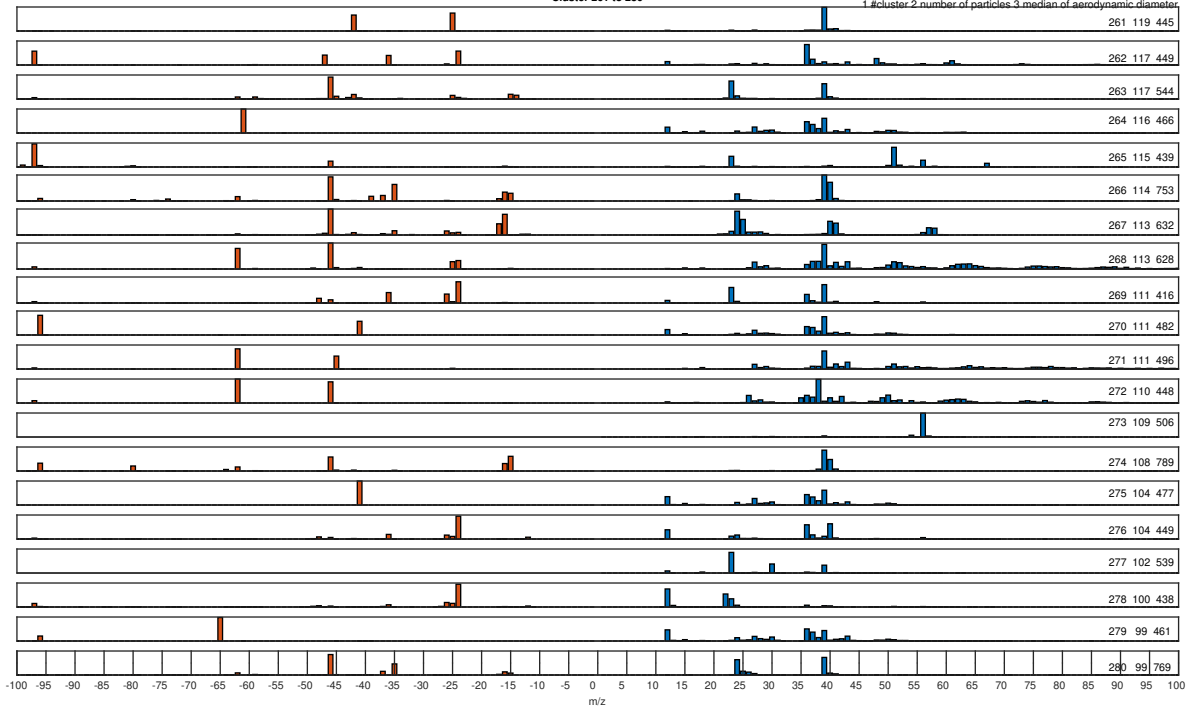
Cluster 241 to 260

1 #cluster 2 number of particles 3 median of aerodynamic diameter
241 145 481

m/z

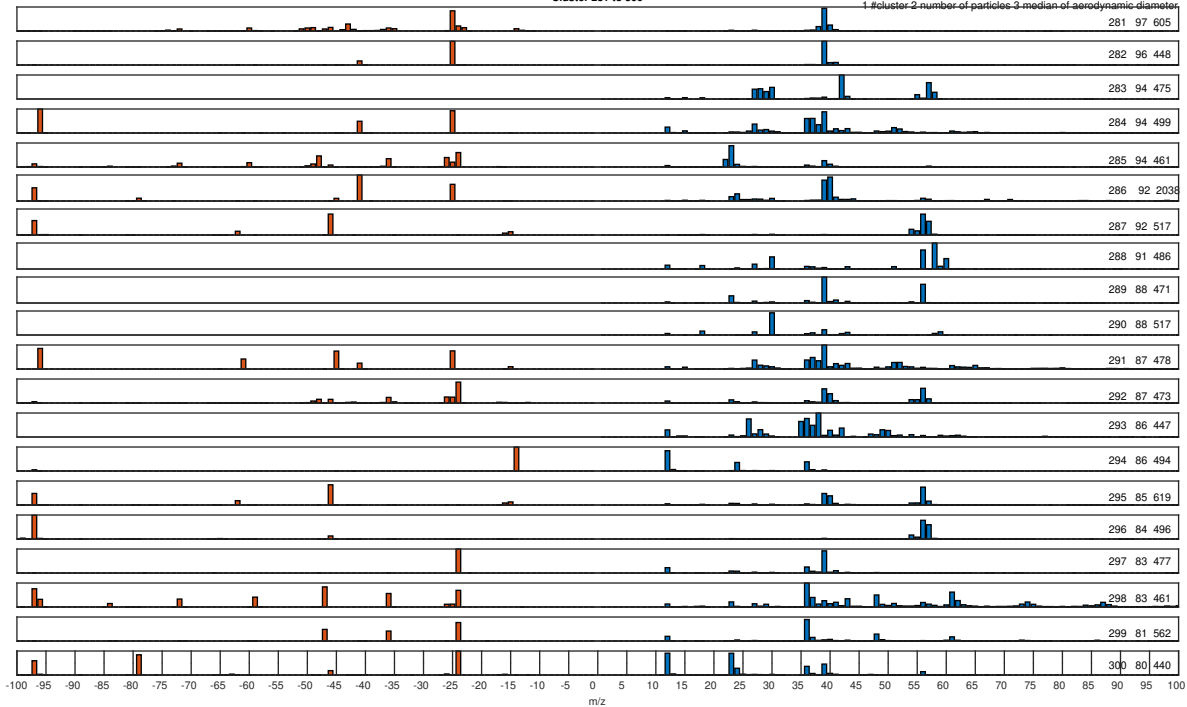
Cluster 261 to 280

1.#cluster 2.number of particles 3.median of aerodynamic diameter



Cluster 281 to 300

1.#cluster 2.number of particles 3.median of aerodynamic diameter



Cluster 301 to 320

1 #cluster 2 number of particles 3 median of aerodynamic diameter

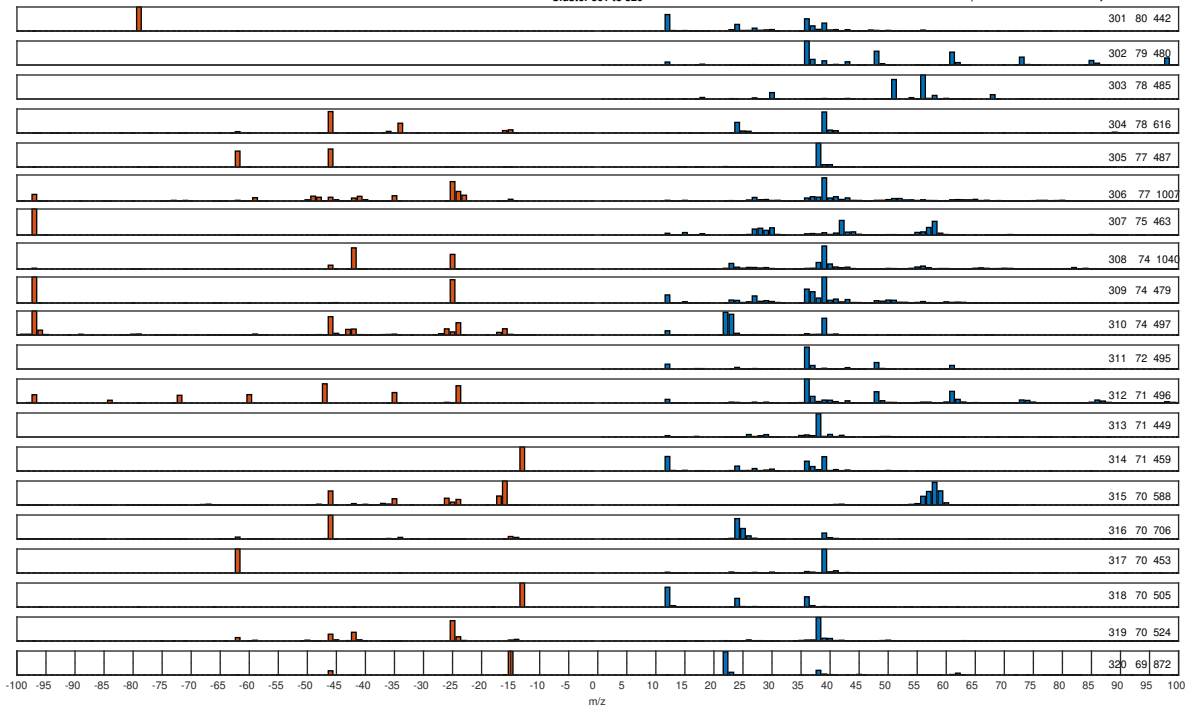
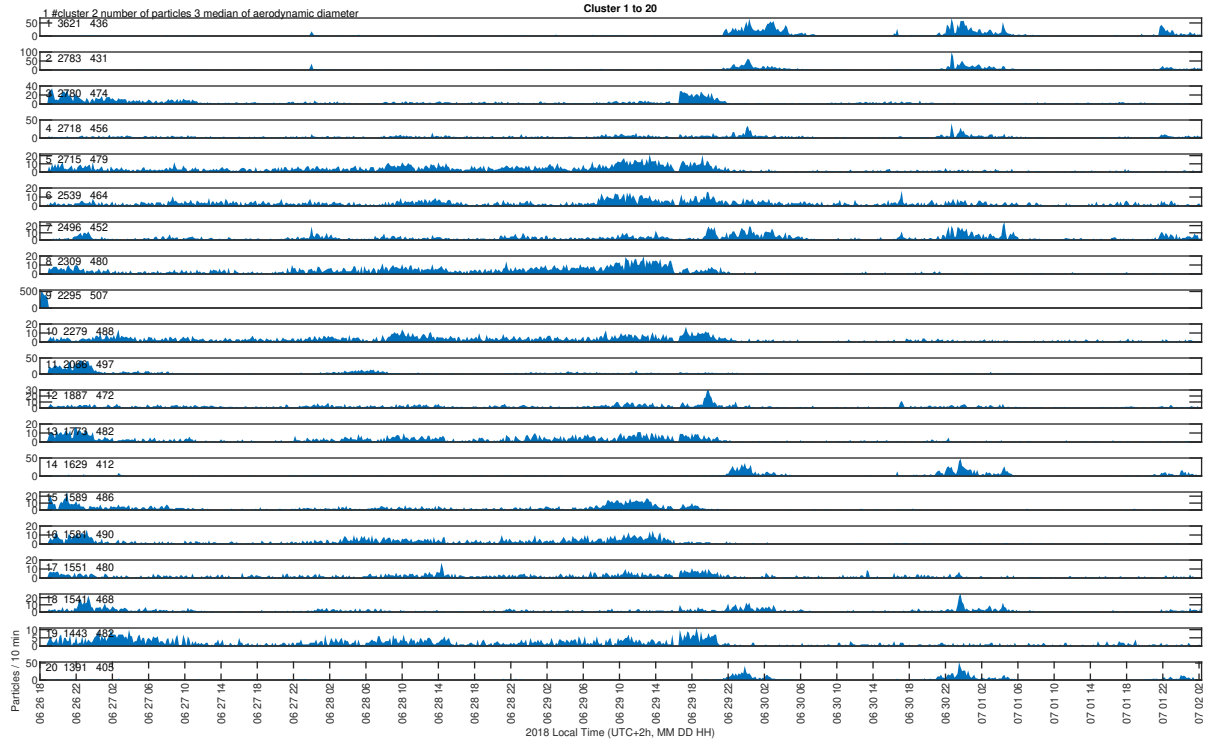
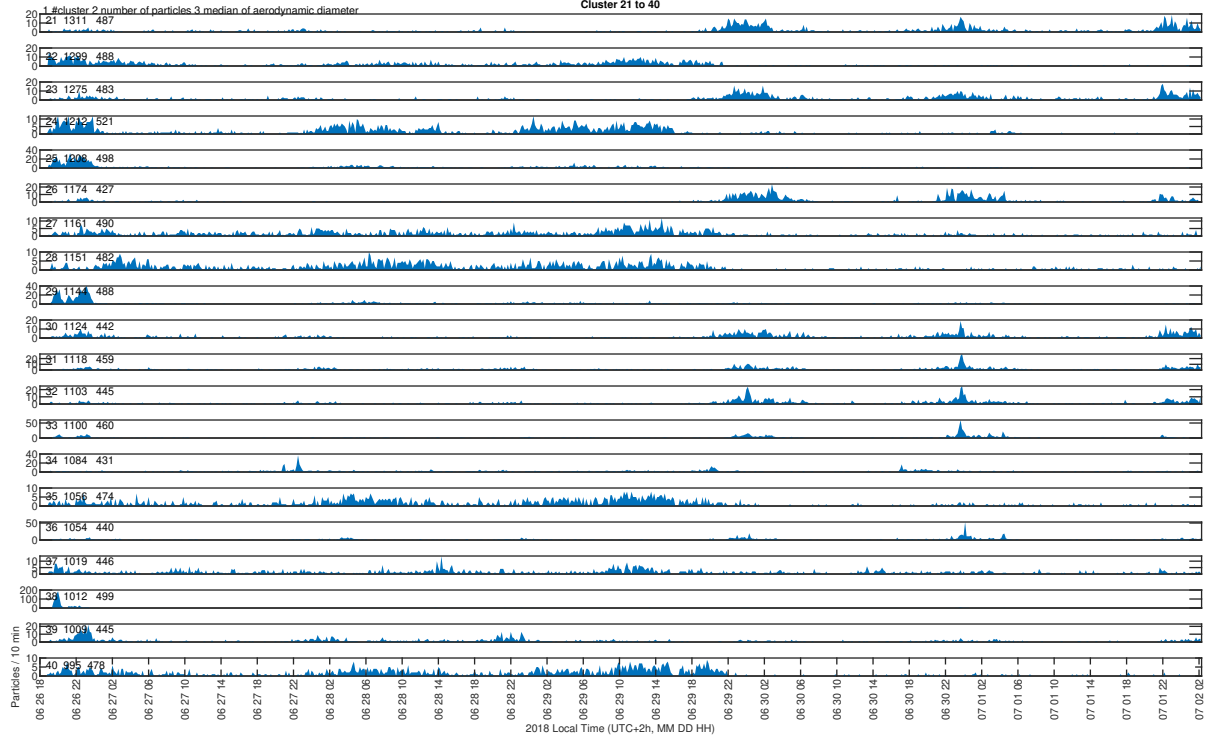
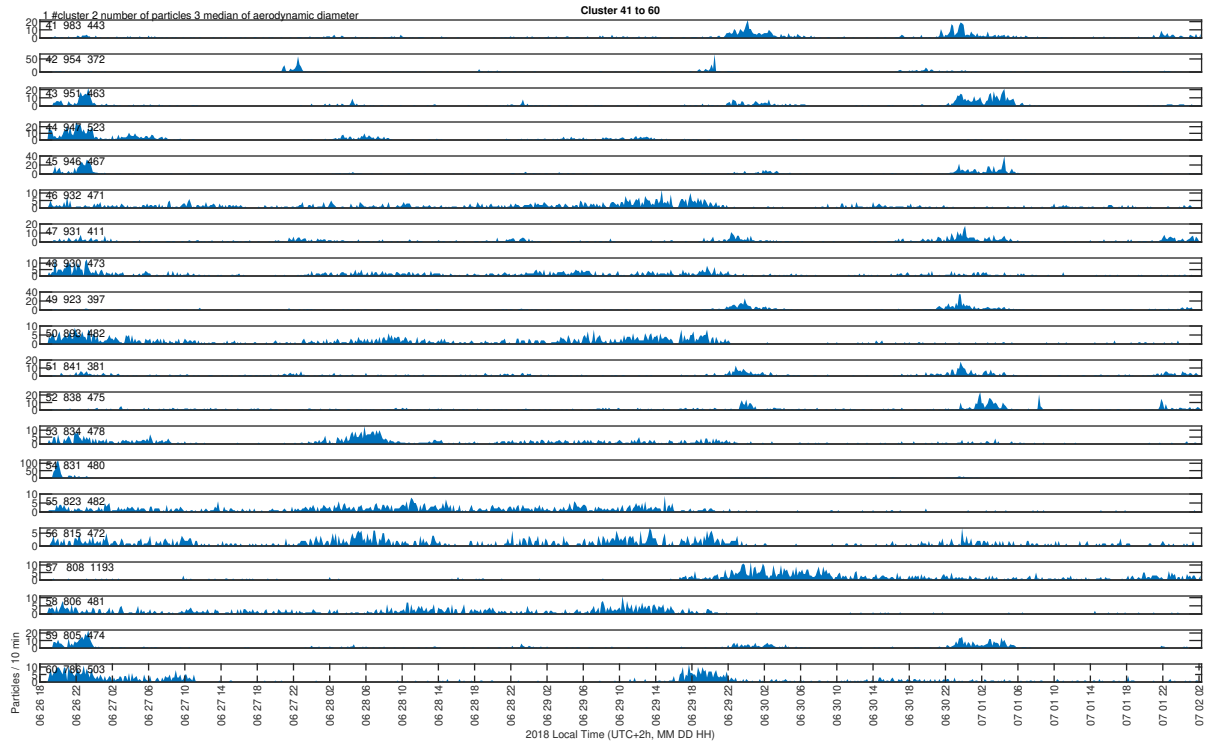


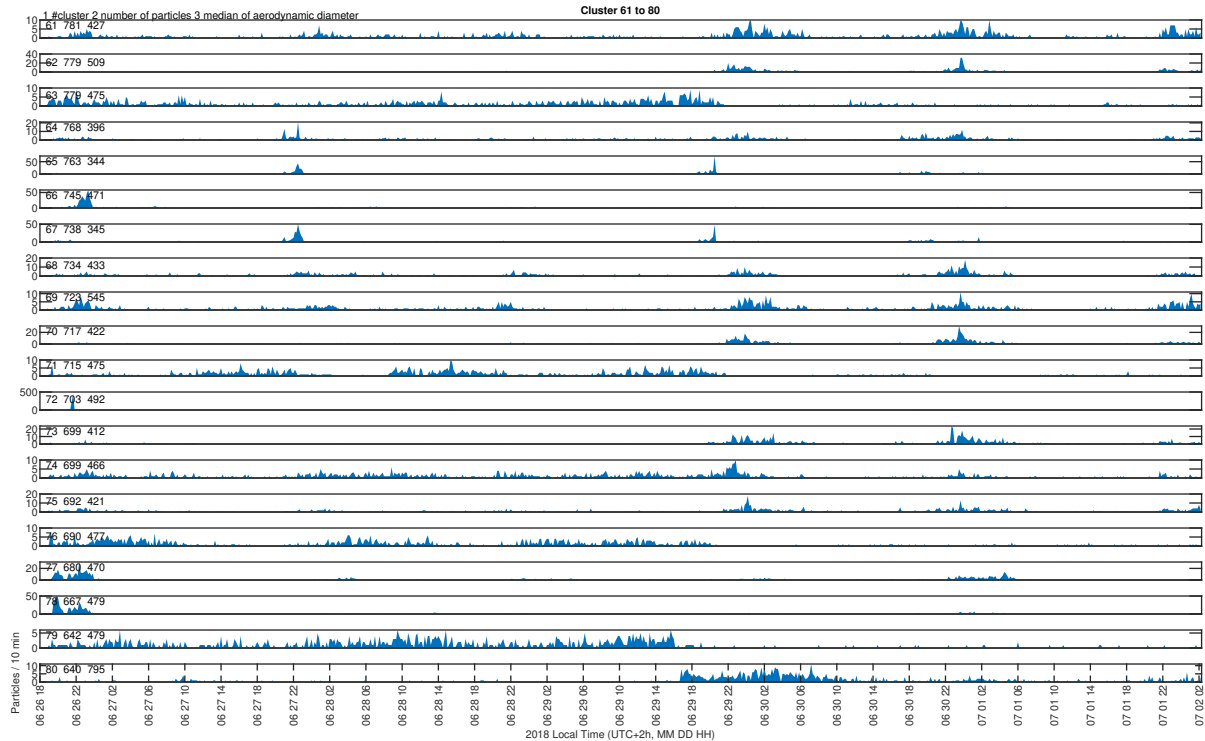
Figure S5: Time series of the top 300 clusters from the ART-2a analysis.

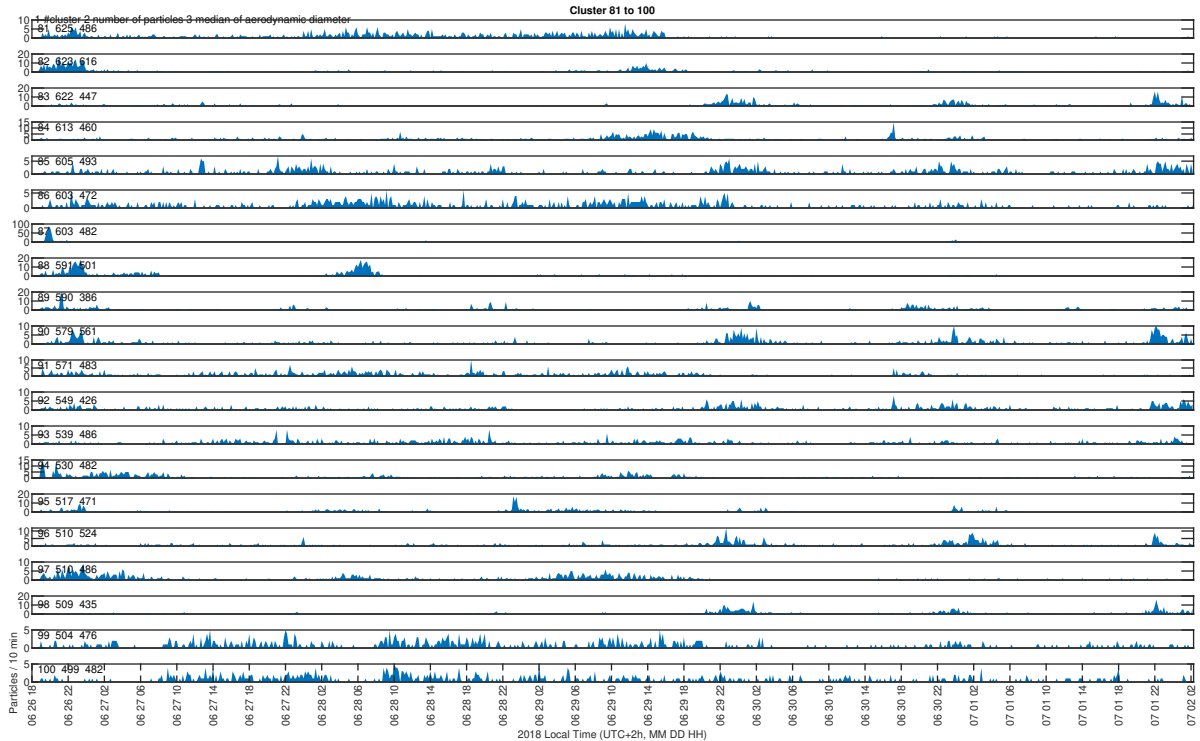


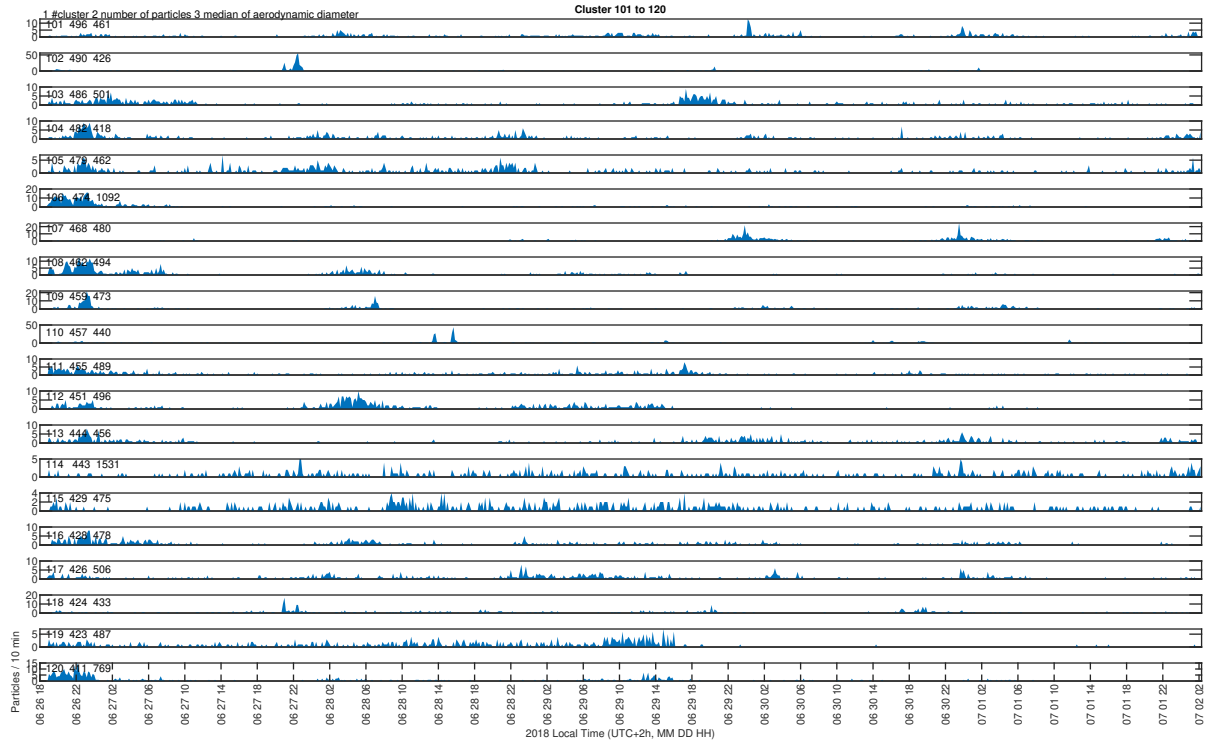
Cluster 21 to 40

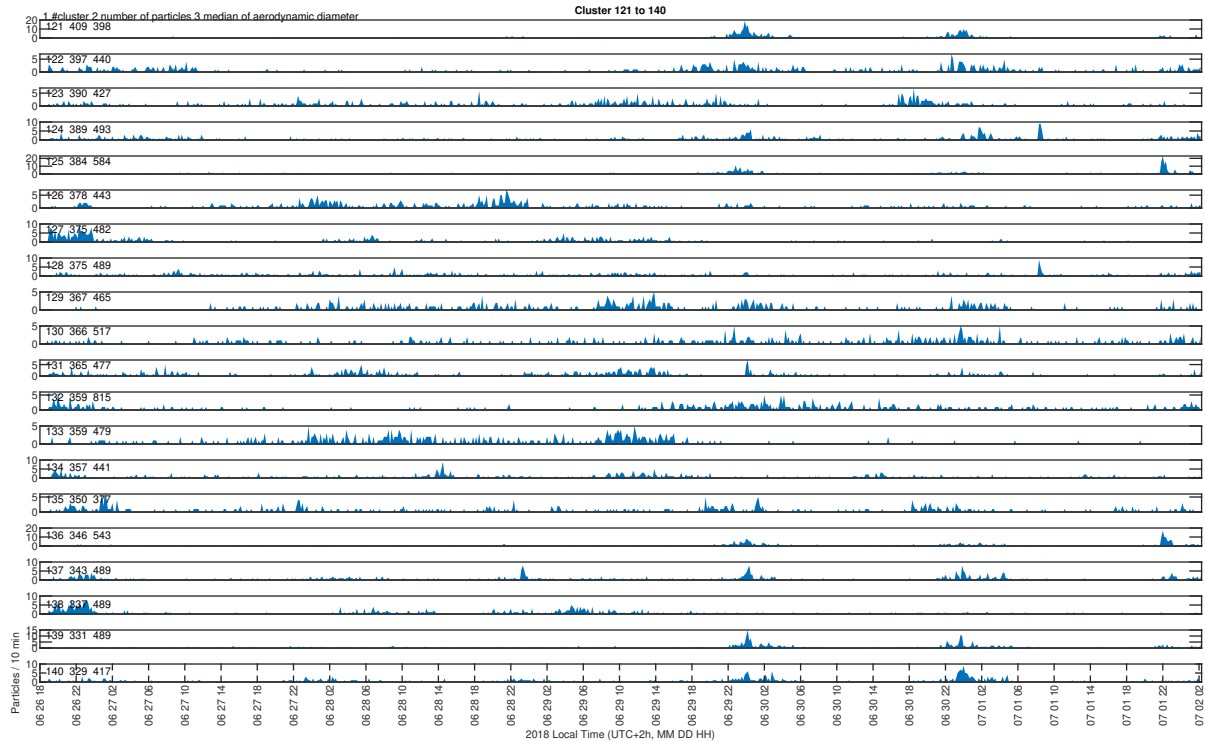


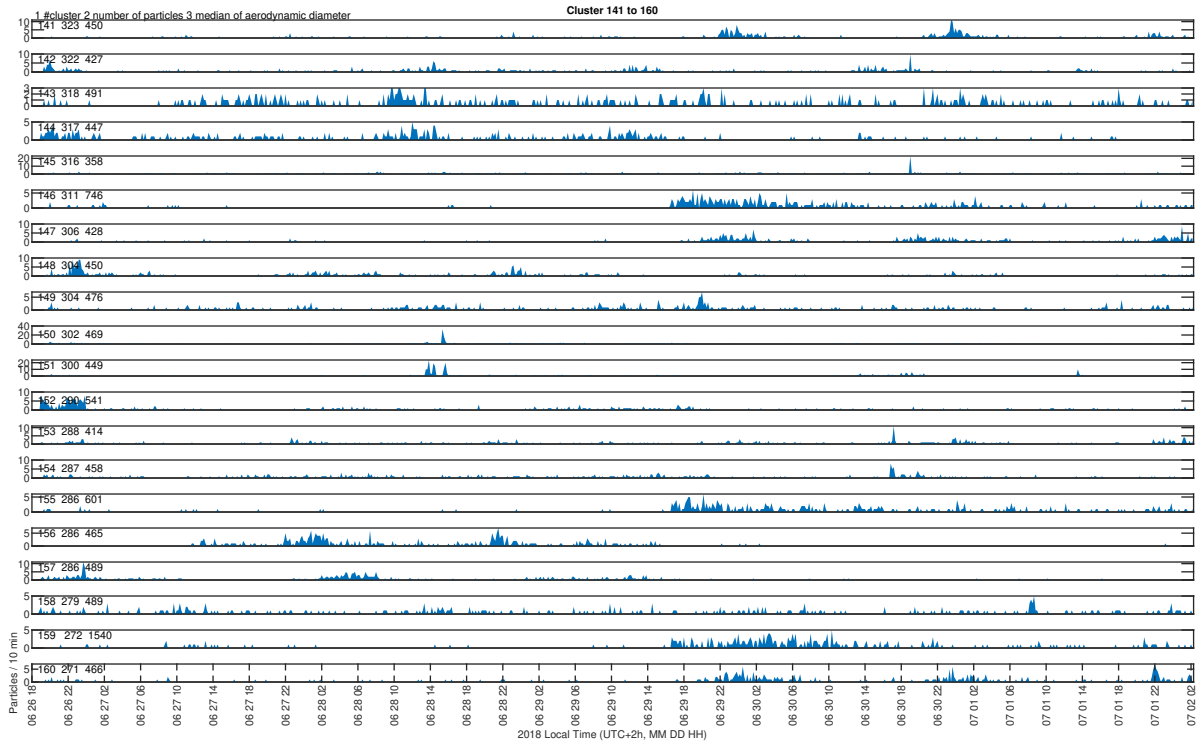




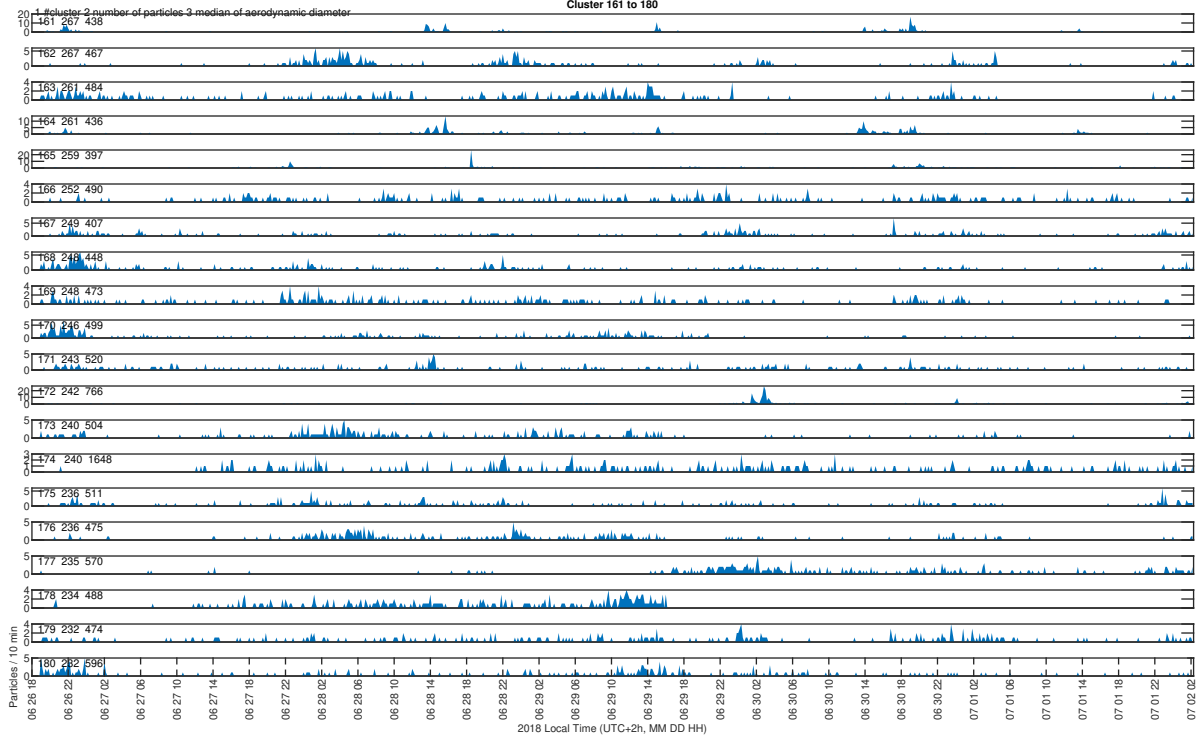


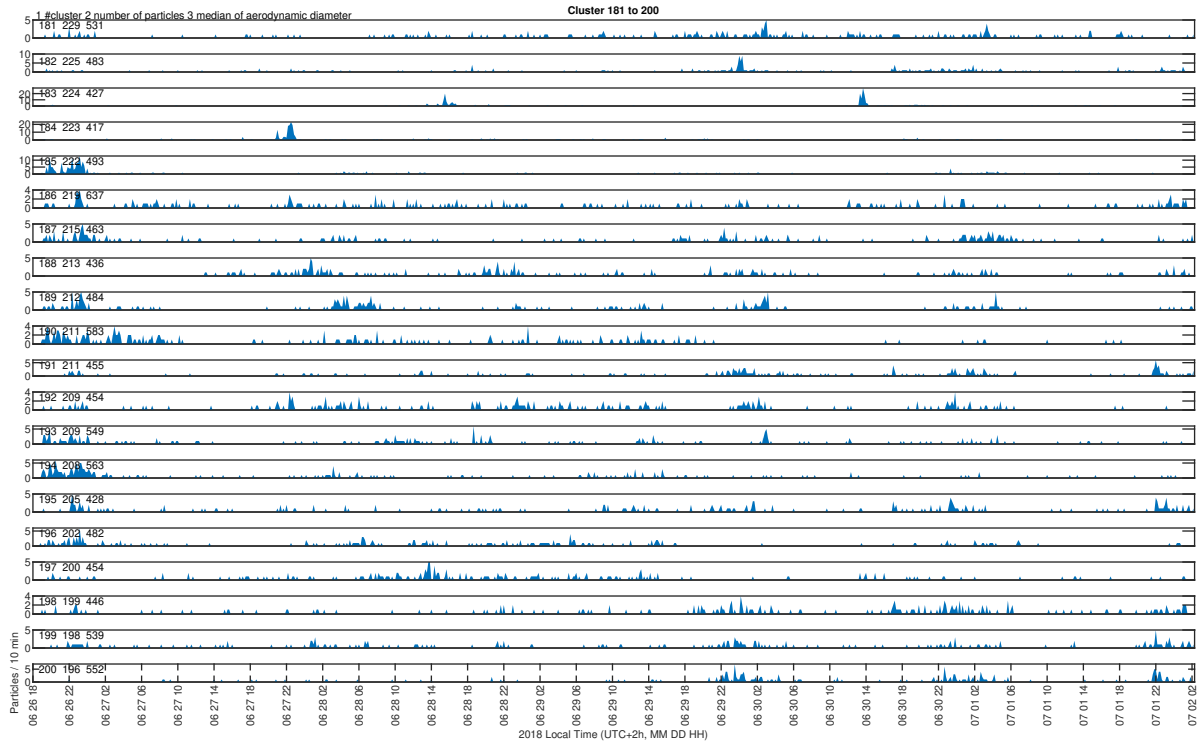


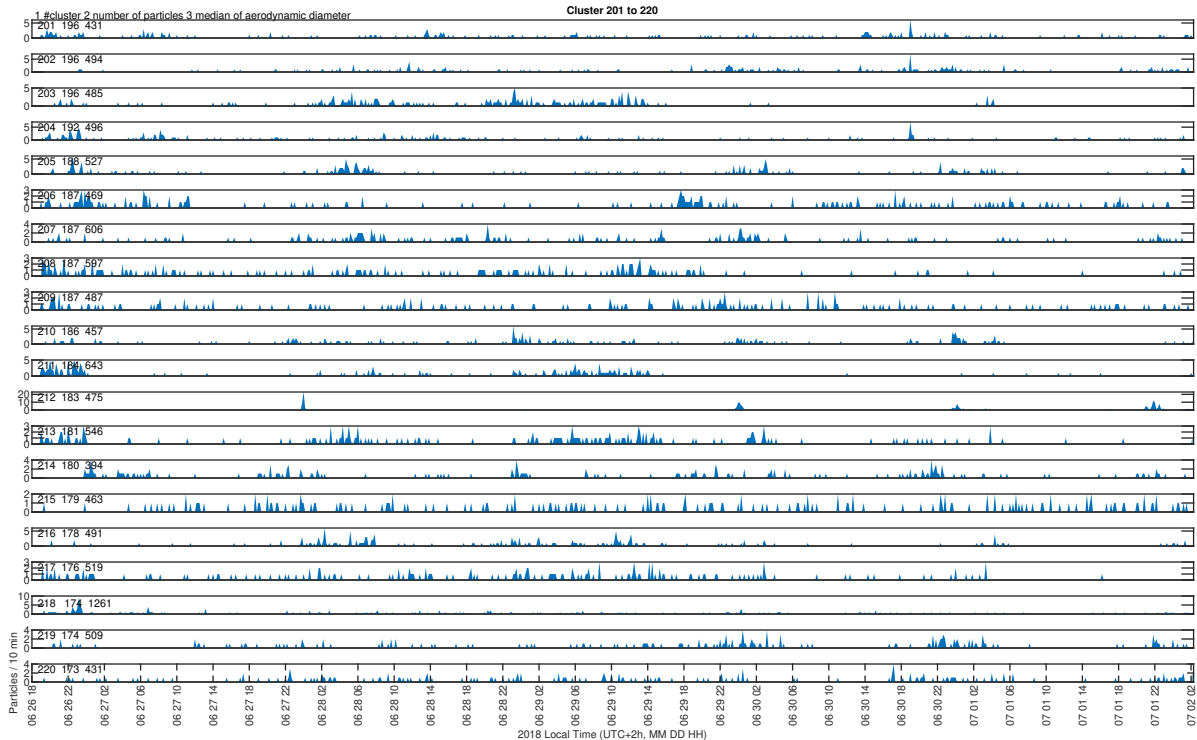


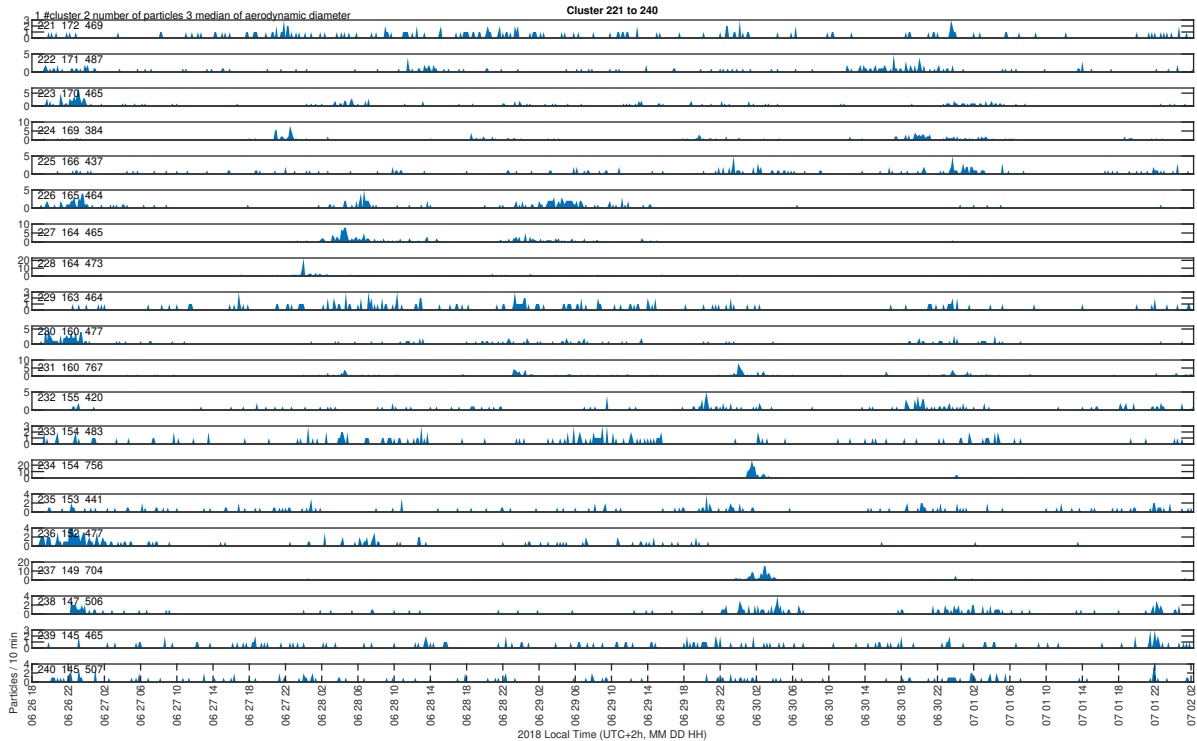


Cluster 161 to 180









Cluster 241 to 260

