



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 extend. (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-build 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 runin 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 C₃⁺, C₄⁺, C₅⁺; fragments/sodium: sum of C⁺, Na⁺, C₃H₃O⁺, C₃H₇⁺; metals: 3x sum of V⁺, VO⁺, ⁵⁴Fe⁺, ⁵⁸Ni⁺). (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, ⁵⁴Fe⁺ and ⁵⁶Fe⁺ for Fe, ⁵⁸Ni⁺ for Ni.



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



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

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+
2	14	21	6		82		110	14	NO+
8	14	51	0		02		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
10	40	64	42	local	177		164	212	FC-S
19	40	04	42	local	1//		104	212	EC-3
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	55	100	84	local	252		200		
44	00	100	84		255				
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				
60	100	101	00		207				
03	102	121	56		207				
/1	107	124	114						
76	109	126	118						
79	113	128	122						
81	116	130	125						
93	123	131	129						
94	135	136	133						
00	120	127	140						
33	135	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	205	160	210						
145	215	102	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	222	173							
170	232	1/3							
194	236	1/5							
201	238	176							
202	242	179							
204	243	181							
207	244	188							
208	247	189							
214	249	191							
214	243	105							
210	251	155							
221	258	197							
222	264	198							
227	268	203							
233	275	209							
239	279	217							
245	283	220							
245	200	220							
248	284	240							
255	280	241							
262	286	263							
269	289	270							
273	290	272							
276	292	274							
277	202	201							
277	295	201							
2/8	297	295							
285									

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

- 283 284 288 294
- 298
- 299 300

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





















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Cluster	301	10 320



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

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