



1 Characterization of the Particle Emission from Ships Operating at 2 Sea Using Unmanned Aerial Vehicles

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10 **Abstract.** This research demonstrates the use of an unmanned aerial vehicle (UAV) to characterize the gaseous (CO₂) and
11 particle (10 - 500 nm) emissions of a ship at sea. The field study was part of the research voyage “The Great Barrier Reef as
12 a significant source of climatically relevant aerosol particles” on-board the RV Investigator around the Australian Great
13 Barrier Reef. Measurements of the RV Investigator exhaust plume were carried out while the ship was operating at sea, at a
14 steady engine load of 30%.

15 The UAV system was flown autonomously using several different programmed paths. These incorporated different altitudes
16 and distances behind the ship in order to investigate the optimal position to capture the ship plume. Five flights were
17 performed, providing a total of 27 horizontal transects perpendicular to the ship exhaust plume. Results show that the most
18 appropriate altitude and distance to effectively capture the plume was 25 m above sea level and 20 m downwind.

19 Particle number (PN) emission factors (EF) were calculated in terms of number of particles emitted (#) per weight of fuel
20 consumed (Kg fuel). Fuel consumption was calculated using the simultaneous measurements of plume CO₂ concentration.

21 Calculated EF_{PN} were between 9.19×10^{14} and $5.15 \times 10^{15} \# \cdot (\text{Kg fuel})^{-1}$. These values are in line with those reported in the
22 literature for ship emissions ranging from $0.2 \text{ to } 6.2 \times 10^{16} \# \cdot (\text{Kg fuel})^{-1}$ to $6.2 \times 10^{16} \# \cdot (\text{Kg fuel})^{-1}$.

23 This UAV system successfully assessed ship emissions to derive emission factors (EFs) under real world conditions. This is
24 significant as, for the first time, it provides a reliable, inexpensive and accessible way to assess and potentially regulate ship
25 emissions.

26 1. Introduction

27 Shipping is the most significant contributor to international freight, with almost 80% of the worldwide merchandise trade by
28 volume transported by ships in 2015 (UNCTAD 2015). Emissions from this transportation mode are a significant contributor
29 to air pollution, both locally and globally. Ships are a major pollutant source in areas surrounding harbours (Viana et al.
30 2014), with over 70% of emissions reaching 400 km inland (Fuglestedt et al. 2009). In 2012 exhaust from diesel engines,
31 the predominant source of ship power, was classified as a group 1 carcinogen by the International Agency for Research on
32 Cancer (IARC). In 2007, pollution from ship exhaust was found to be responsible for approximately 60,000
33 cardiopulmonary and lung cancer deaths worldwide annually (Corbett et al. 2007a). Such emissions are also a strong climate
34 forcing agent, contributing to global warming through the absorbance of solar and terrestrial radiation (Hallquist et al. 2013a;
35 Lack et al. 2011; Winnes et al. 2016).

36 Despite these findings, emissions from shipping have consistently been subject to less regulation than those of land-based
37 transport with ship emissions in international waters remaining one of the least regulated parts of the global transportation
38 system (Cooper 2001; 2005; Corbett and Farrell 2002; Corbett and Koehler 2003; Eyring et al. 2005; Streets et al. 1997;
39 USEPA-OTAC 2012). Currently, no specific restrictions for ship-emitted particulate matter (PM) exist, with the only
40 regulated pollutants being NO_x and SO₂. The International Maritime Organization (IMO) recently revised the regulation of



41 these gaseous pollutants through the Annex VI of the International Convention for the Prevention of Pollution from Ships –
42 the Marine Pollution Convention (MARPOL). The IMO expected that these regulations would lead to an indirect decrease in
43 particle number (PN) concentration due to the reduction of NO_x emissions and the use of fuel with lower sulphur content
44 [14]. However, it has been found that the use of some low sulphur fuels lead to increased PN concentrations at lower engine
45 loads (Anderson et al., 2015), which stresses the importance for regulation specifically addressing particulate matter (PM).
46 The majority of emitted PM is in the ultrafine size range, < 0.1 μm, which have been demonstrated to have a particularly
47 significant impact on health and the environment (WHO 2013). However, due to the lack in regulation, ultrafine particles, in
48 terms of PN concentration, emitted from ships have remained unassessed in real world conditions. Quantifying PN
49 concentration is critical to improve our understanding of shipping’s impact on health and climate (Anderson et al. 2015;
50 Blasco et al. 2014; Chen et al. 2005; Cooper 2001; Corbett and Farrell 2002; Corbett et al. 2007b; Isakson et al. 2001;
51 Mueller et al. 2015; Reda et al. 2015; Ristovski et al. 2012; Williams et al. 2009). To achieve this, wide-scale evaluation of
52 ship emission factors (EFs) is necessary. EFs are commonly expressed as the amount of pollutant (x) emitted per unit mass
53 of fuel consumed $g(x) \cdot (Kg \text{ fuel})^{-1}$. Different methods have been used to investigate ship EFs, including laboratory test-bench
54 studies, on-board measurements, and measurement of ship emission plumes.
55 Test-bench studies (Anderson et al. 2015; Kasper et al. 2007; Mueller et al. 2015; Petzold et al. 2008; Petzold et al. 2010;
56 Reda et al. 2015) have been used to characterize emissions from different engines at various loads in laboratory conditions.
57 However, engine performance and emissions have been shown to be different in real world operations when compared to
58 laboratory studies. This calls for measurements of ship emissions in-situ to collect reliable data for EF calculations (Agrawal
59 et al. 2008; Blasco et al. 2014; Murphy et al. 2009). To date, only a few studies have been undertaken on-board ships to
60 calculate real emission factors (Hallquist et al. 2013b; Juwono et al. 2013). This is attributed to the prohibitive costs and time
61 commitments of setting up and maintaining on-board measurement equipment on commercial ships. Airborne ship plume
62 measurements (Balzani Lööv et al. 2014; Beecken et al. 2014a; Berg et al. 2012; Cappa et al. 2014; Lack et al. 2008; Lack et
63 al. 2009; Pirjola et al. 2014a; Schreier et al. 2015; Sinha et al. 2003; Westerlund et al. 2015) offer an alternative method of
64 in-situ measurements without requiring on-board monitoring stations. In the past the cost, the significant difficulties in
65 deployment of these systems, and the risk for manned aircrafts have limited their feasibility. However, this has recently
66 changed with the rapid advances being made in commercially available Unmanned Aerial Vehicle (UAV) technology.
67 Hexacopter UAVs have seen a wide scale increase in industry and research applications due to their ease of use and
68 comparatively low cost (Brady et al. 2016; Gonzalez et al. 2011; Malaver Rojas et al. 2015). Used in conjunction with air
69 monitoring equipment, these systems provide, for the first time, the ability to perform relatively simplistic and cost-effective
70 airborne measurements of ship emissions. However, to date no studies have reported the use of a UAV system capable of
71 collecting data to calculate the EF of PN concentration for ships at sea.
72 This research utilized a customized hexacopter UAV carrying instruments for PN concentration and CO₂ measurements to
73 derive EF_{PN} . The UAV system was deployed from the RV Investigator research vessel while at sea. Autonomous
74 measurements of the RV investigators exhaust plume were taken over several flights at various altitudes and distances from
75 the ship. Data collected was used to optimize the sampling flight path and successfully quantify the RV investigators EF for
76 PN concentration.

77 2. Methodology and Measurement system

78 Measurements were conducted as part of the research voyage “The Great Barrier Reef as a significant source of climatically
79 relevant aerosol particles” aboard the RV Investigator research vessel over a two day period of the 13 and 14 October 2016
80 (day 1 and day 2). Measurements of PN and CO₂ concentration emitted by the RV Investigator were taken using a PN and
81 CO₂ monitor mounted on a customized DJI EVO S800 hexacopter UAV (DJI 2014).



82 2.1. The RV Investigator and the voyage

83 The RV Investigator is a sophisticated ocean research vessel configured to enable a wide range of world class atmospheric,
84 biological, goescience and oceanographic research. The vessel is 94 m long, has a gross weight of 6,082 tons, a fuel capacity
85 of 700 tons of ultra-low sulphur diesel fuel. It is powered by three 9 cylinder 3000 kW MaK diesel engines, each coupled to
86 a 690V AC Generator. Ship propulsion is achieved using two 2600 kW L3 AC reversible propulsion motors powered by
87 these generators. The RV Investigator can host up to 30 crew members and 35 researchers for a maximum voyage period of
88 60 days with at a maximum cruising speed of 12 knots.

89 A suite of instrumentation for atmospheric research is available on the RV Investigator. This includes a radar system capable
90 of collecting weather information within a 150 km radius of the vessel, and instruments measuring: sunlight parameters;
91 aerosol composition, particle concentration and size distributions; cloud condensation nuclei; gas concentrations; and various
92 other components of the atmosphere. These instruments are housed inside two dedicated on-board laboratories for aerosol
93 and for atmospheric chemistry research. An atmospheric aerosol sample is continuously drawn into the laboratories for
94 analysis through a specialized inlet fitted to the foremast of the ship. Of particular interest to this study, the ship contains a
95 PICARRO (PICARRO Inc., Santa Clara, California, USA) G2401 analyser (Inc. 2017) that continuously measures CO₂, CO,
96 H₂O and CH₄. It has an operation range between 0-1000 ppm and a parts-per-billion sensitivity (ppb) for CO₂.
97 The two day UAV measurement study was possible as part of the RV Investigator voyage “The Great Barrier Reef as a
98 significant source of climatically relevant aerosol particles”, which started in Brisbane on the 28th of September 2016. The
99 ship was used as both: a floating platform to allow launch and recovery of the UAV system; and as the source of an exhaust
100 plume measured by the UAV system for EF calculation. During a several day stationary period on the Great Barrier Reef off
101 the coast of Australia, it was possible to measure the ship plume under stable real world conditions over two consecutive
102 days. One of the three ship engines was maintained at a steady engine load of 25 – 30 % of the maximum engine power
103 during all measurements.

104 2.2. UAV system

105 Measurements of PN and CO₂ concentrations in the ship plume were performed using two commercial sensors mounted on-
106 board a hexacopter UAV. The UAV used (Figure 1) is a composite material S800 EVO manufactured by DJI (DJI 2014).
107 The UAV is 800 mm wide and 320 mm in height, with an unloaded weight of 3.7 kg. Minimum and maximum take-off
108 weights are 6.7 kg and 8 kg, respectively. The UAV contains a 16000 mAh LiPo 6 cell battery, which provides a hover time
109 of approximately 20 min when operating at minimum take-off weight. The telemetry range of the UAV is 2 km, which was
110 adequate to cover the desired sampling area (See Figure 2).

111 The payload consisted of a PN concentration and a CO₂ monitor mounted on-board underneath the UAV. Careful placement
112 of the payload was required to prevent flight issues caused by an altered centre of gravity. Also included was a carbon fiber
113 rod, which extended outward horizontally from the UAV. The sampling lines for the monitors were attached to the end of
114 this rod to ensure that measurements were not affected by the downwash of the UAV rotors. The total weight of the payload
115 was (1.2 kg), which allowed the UAV system to fly for 12-15 min before landing at the home point (A) (See Figure 2).

116 The S800 was used in conjunction with the DJI Wookong autopilot. The software provides an intuitive and easy to use
117 interface where autonomous flight paths can be planned, saved, and uploaded into the UAV. In addition to this, the ground
118 station allows for continuous, real-time monitoring of the status of the UAV during operation; which includes its longitude,
119 latitude, altitude, waypoint tolerance and airspeed.

120 The DJI S800 was chosen for this study because it is designed to operate under the 20 kg all up weight (AUW) class of
121 UAV. This reduces operational costs and avoid subjection to the tighter regulations of larger platforms. Small UAV cannot
122 be operated above any person, or closer than 30 m of populated areas, houses and people. Furthermore, current Civil
123 Aviation Safety Australia (CASA) regulations restrict the use of small UAV (2 and 20 kg) to visual line-of-sight daylight



124 operation, with a maximum altitude of approximately 120 m and within a radius of 3 nmi of an airport. UAVs in this
125 category are not permitted for research unless the research institution has been granted a permit exception. These exceptions
126 can be granted if the institution in question has or collaborates with an UAV operation team who must have: an experienced
127 UAV pilot who is also radio controller specialist; a license for commercial UAV operation; and appropriate liability
128 insurance . Queensland University of Technology (QUT) has an unmanned operator certificate and four pilots who have
129 UAV controller licenses.

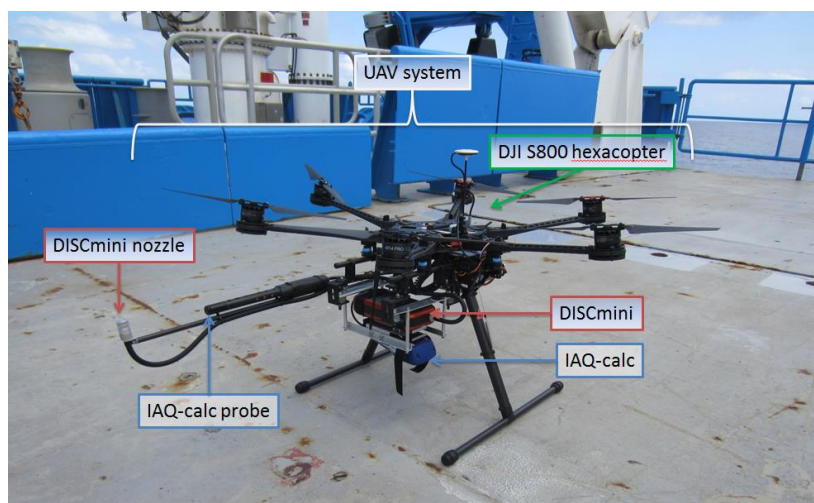
130 2.2.1. Instrumentation

131 2.2.1.1. Instrumentation for PN concentration

132 This study measured PN concentration using a Mini Diffusion Size Classifier (DISCmini), developed by the University of
133 Applied Sciences, Windisch, Switzerland (Fierz et al. 2008). The DISCmini is a portable monitor used to measure
134 concentration of particles in the 10-500 nm diameter size range, with a time resolution of up to 1 s (1 Hz). It can measure PN
135 concentrations between 10^3 and 10^6 N/cm³. Measurement accuracy is dependent upon the particle shape, size distribution,
136 and number concentration. The advantages of using the DISCmini are its relatively small dimensions (180 x 90 x 40 mm),
137 low weight (640 g, 780 g with the sampling probe, Figure 1) and long battery life of up to 8 hrs. These
138 characteristics allow it to be easily integrated on the UAV.

139 2.2.1.2. Instrumentation for CO₂ concentration measurements

140 A TSI (TSI, Shoreview, Minnesota, United States) IAQ-calc 7545 model was chosen to measure CO₂ concentrations. Its
141 sensor is based on a dual-wavelength NDIR (non-dispersive infrared) with a sensitivity range between 0 to 5,000 ppm and an
142 accuracy of $\pm 3.0\%$ of reading or ± 50 ppm (whichever is greater). The measurement resolution is 1 ppm with a maximum
143 time resolution of 1s. Similar to the DISCmini, the advantages of using the IAQ-calc are: its small dimensions (178 x 84 x 44
144 mm); low weight (270 g, with batteries, significantly lower than the DISCmini), and a battery life of 10 hours.
145 The readings of the IAQ-clac for CO₂ were compared with those measured by the on-board PICARRO G2401 analyser.
146 Both the DISCmini and the IAQ-calc were tested and calibrated in the laboratory prior to the commencement of the
147 measurements (Figure S1 in the Supplementary Material). All data was logged with a 1 s time interval.



148

149 **Figure 1. The UAV system with the on-board instrumentation: the DISCmini and the IAQ-calc.**

150



151 **2.3. Meteorological data**

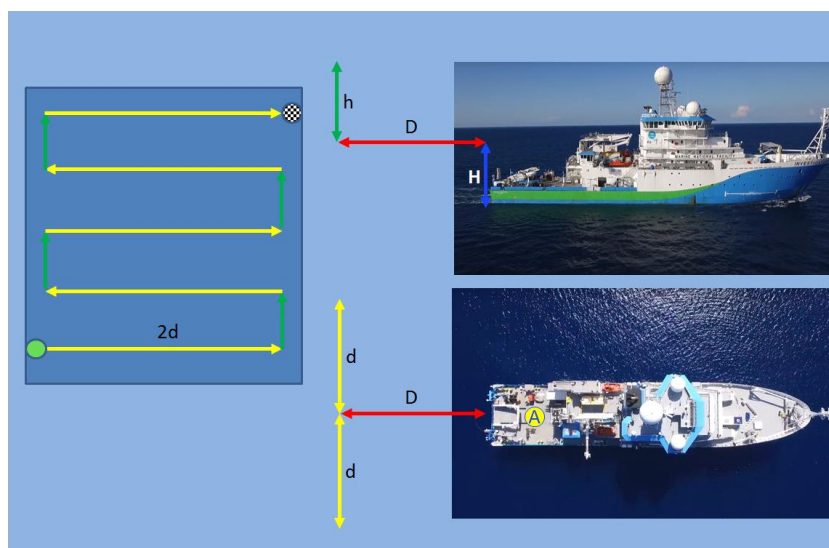
152 Meteorological data (including air temperature, relative humidity, atmospheric pressure, wind speed and direction) were
153 recorded by the RV Investigators on-board instrumentation during the entire voyage with a 60 s time interval, 24/h a day.

154 **2.4. Study design**

155 During the two measurement days of this study, the vessel was heading into the wind whilst idling the UAV missions at sea.
156 This positioning caused the exhaust plume to extend downwind, directly behind the ship. The UAV system was launched off
157 the back deck, autonomously sampling at varying altitudes and distances into the downwind plume. Flight speed of the UAV
158 was 1.5 m/s, the minimum for the S800.

159 Day 1 was used to optimise the study design, focusing on finding the flight path most suitable to capture the ship plume.
160 Figure 2 shows the programmed flight path, which consisted of a continuous flight beginning at a distance (D) and from an
161 altitude (H) above the surface. Point A, located on the back deck of the RV Investigator, represents the 'home point'. In
162 UAV terminology this refers to the position where the UAV system takes off and lands. The UAV system was programmed
163 to move horizontally by a distance ($2d$), perpendicular to the ship, then climb vertically for 10 m (h) before flying in the
164 opposite horizontal direction for the same distance ($2d$). The UAV was then programmed to climb another 10 m (h) before
165 repeating this pattern until the UAV reached an altitude of 65 m above the ocean. During day 1, the UAV system followed
166 three different flight paths, each one with both a different distance D behind the ship (20, 50 and 100 m), and a different
167 horizontal distance $2d$ (50, 100 and 150 m).

168 The optimised flight path for day 2 started 20 m behind the ship and 25 m above the surface, with no altitude variation. The
169 UAV path was limited to a continuous horizontal flight of 50 m ($2d$) at steady speed of 2 m s^{-1} . This path and flying speed
170 allowed up to 4 horizontal transects to capture the ship plume.



171

172 **Figure 2.** Flight path used to capture the plume: H - height from the ocean, D - distance behind the ship to the flight beginning
173 point, h - rising altitude after the horizontal transect, $2d$ - full length of the horizontal transect

174 **2.5. Experimental procedure**

175 The UAV can fly either manually or autonomously. As a safety precaution, every take-off and landing was performed using
176 the manual flight mode. Once in the air, the UAV was switched to autonomous flight mode, allowing the platform to follow
177 the pre-programmed flight path discussed in the previous section. The flight path consisted of waypoints, which are three-



178 dimensional GPS points that dictate the position of the UAV along the flight path. The waypoints and flight plans for each
179 flight were programmed using the aforementioned DJI Wookong ground station software. The DISCmini and the IAQ-calc
180 were fitted on the underside of the UAV at the beginning of each measuring day. Five flights were performed across the two
181 measurement days, providing a total of 27 horizontal transects perpendicular to the ship's exhaust plume.

182 2.6. Emission factors

183 The calculation of an emission factor for particle number concentration (EF_{PN}) from the collected ship plume measurements
184 was performed using Eq. (1). This method has previously been used for ship (Westerlund et al. 2015), road vehicle (Hak et
185 al. 2009) and aircraft (Mazaheri et al. 2009) emissions. The measured values of PN concentration were related to the amount
186 of fuel consumed by the engine in question through the use of the simultaneous measurements of CO₂ concentration taken by
187 the UAV. This was achieved by using a published value for a ship emission factor of CO₂ (EF_{gas}) of 3.2 Kg CO₂ (Kg fuel)⁻¹
188 (Hallquist et al. 2013b; Hobbs et al. 2000).

189 Eq.(1).

$$190 \quad EF_{PN} = \frac{\Delta PN}{\Delta gas} \times EF_{gas} \quad (1)$$

191 The ΔPN and Δgas in Eq. (1) represent changes in the measured particle number and CO₂ concentrations, respectively.
192 Background concentrations of PN and CO₂ were subtracted and EF_{PN} was calculated by integrating the peak plume
193 concentration measured by the DISCmini and IAQ-clac mounted on the UAV system; which is defined as the average
194 concentration measured by the DISCmini and IAQ-calc outside the ship plume.

195

196 3. Results and Discussion

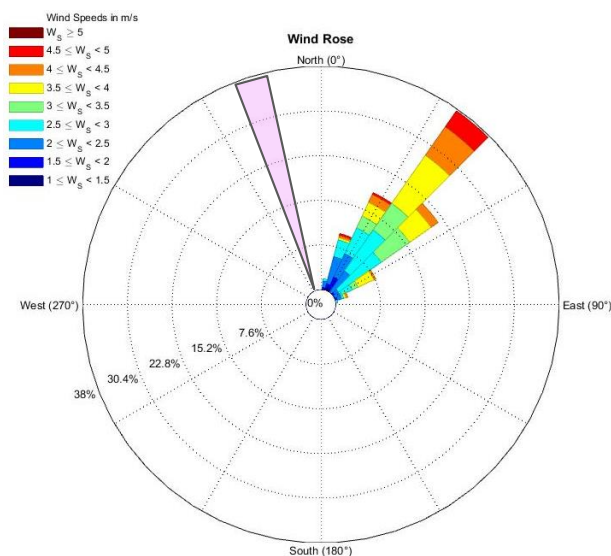
197 3.1. Meteorological and Investigator data

198 Wind conditions were very stable during both day 1 and day 2, following one main pattern for the entire flight time. The
199 wind speed ranged from 3 - 13 m s⁻¹. The wind direction was predominantly from the NE during day 1 and ESE during day
200 2.

201 The wind rose graphs in Figure 3a and 3b illustrate the wind data recorded with the on-board weather instrumentation during
202 all horizontal transects flown during day 1 and 2 respectively. The prevalent wind direction was ESE, which corresponded to
203 the heading of the RV Investigator (indicated by the rose triangle).

204 The wind direction changed occasionally to E during the flight, causing the UAV to fail to capture the RV Investigator
205 plume during some transects. As a result, 2 of the 8 horizontal transects collected on day 2 were excluded from the analysis.

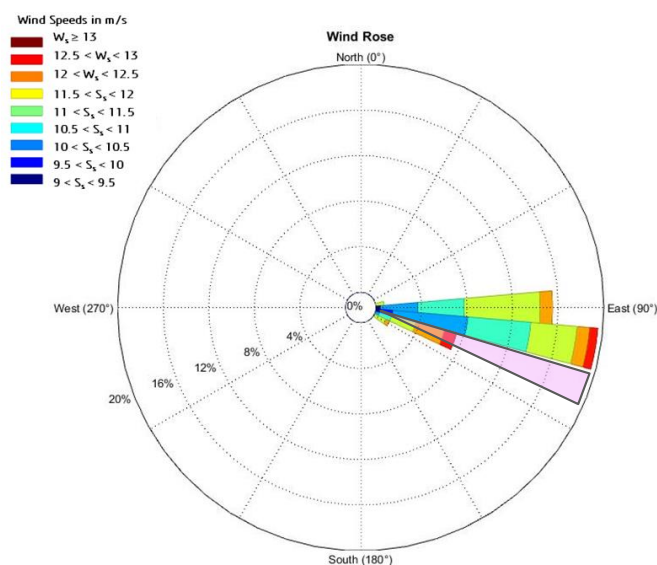
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207

208 Figure 3a – Wind rose showing wind speed and direction during day 1. Rose triangle shows RV Investigator direction during the
 209 measurements.

210



211

212 Figure 3b – Wind rose showing wind speed and direction during day 2 optimized flight. Rose triangle shows RV Investigator
 213 direction during the measurements.

214 3.2. UAV system horizontal transects inside and outside the plume

215 The UAV system acquired data for a total of 27 horizontal transects for day 1 and day 2. Data was collected at altitudes
 216 between 25 m and 65 m above the water surface. During day 1 the plume was captured once when the UAV was at 25 m



217 altitude and 20 m downwind of the ship; and again at both 25 and 35 m altitude 100 m downwind of the ship. These
218 observations lead to the optimized flight used on day 2, which started downwind at 25 m above the surface and 20 m behind
219 the ship. On day 2 the UAV system successfully captured the plume during 6 of the 8 transects performed. Across the two
220 days this lead to a total of 9 transects that captured the plume and which have been considered for discussion, shown in
221 Table 1.

222

Measuring day	Altitude	Distance behind the Investigator	Number of transects
Day 1	25 m	20 m	1
*Day 1	25 m	100 m	1
Day 1	35 m	100 m	1
Day 2	25 m	20 m	6

223

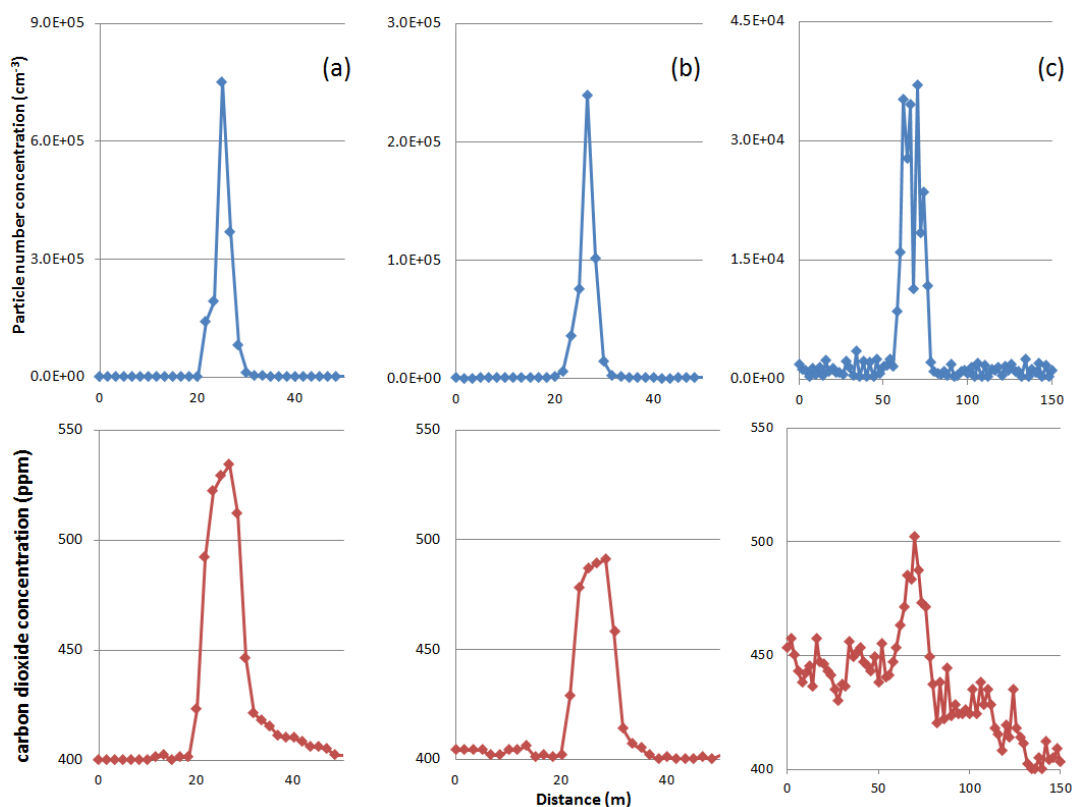
224 **Table 1** – Specifications of the transects considered for the data analysis. The (*) indicates the transect of Day 1 of which PN
225 concentration and CO₂ profiles are presented in Figure 4.

226

227 Figure 4 shows the PN concentration and CO₂ profiles, collected during two (a; b) transects on day 2, and (c) during one
228 transect of day 1 (Spec. in Table 1, Day1*).

229 The PN concentration profiles for the (a) and (b) transects in Figure 4 show that the concentration varied by five orders of
230 magnitude between the outside and inside the plume, while the CO₂ profiles show an increase up to 140 ppm above the
231 background.

232 The profiles in (c) show that the PN concentration was four orders of magnitude greater inside the plume at 100 m behind the
233 ship and that the CO₂ concentration was up to 100 ppm higher inside the plume.



234

235 **Figure 4 – (a) and (b) show the PN concentration and CO₂ profiles collected at 20 m behind the ship 25 m above the surface during**
236 **one of the flight in day 2. (c) shows the PN concentration and CO₂ profiles collected during flight 3 of day 1 at 100 m behind the**
237 **ship, 25 m above the surface.**

238

239 Figure 4 (a) and (b) both show transects at 25 m altitude and 20 m behind the ship. Both the PN concentration and CO₂
240 measurements show clear, single peaks as the UAV crosses the plume. However, the maximum PN concentrations measured
241 in (a) ($7.5 \times 10^5 \text{ \#.cm}^{-3}$) are approximately three times greater than those in (b) ($2.4 \times 10^5 \text{ \#.cm}^{-3}$). Furthermore, the CO₂
242 measurements between (a) and (b) have a difference of (43ppm). As the ship engine remained under steady load throughout
243 these measurements, the variations between (a) and (b) can be attributed to several factors which reduce the effectiveness of
244 the UAV transect for capturing the plume. Slight changes in ambient conditions such as temperature, wind direction and
245 intensity will alter the path of the plume as it moves away from the ship. The UAVs automated flight path cannot account for
246 these variations. Therefore, the degree to which the UAV enters the plume, and thus the concentrations it measures, will be
247 different on each transect. Both CO₂ and PN concentration measurements will be similarly affected by this variance.
248 However; it is expected that this will contribute to the calculated error margin of the final result.

249 In comparison to Figure 4 (a) and (b), the graphs in (c) show substantially less defined, wider peaks with lower pollutant
250 concentrations. This is attributed to a difference in flight paths, with Figure (c) representing data from a transect 100 m
251 behind the ship; whilst (a) and (b) were performed 20 m behind the ship. As the plume travels away from the ship it will
252 begin to turbulently mix with the surrounding air mass; causing concentrations to decrease and the plume to broaden as the
253 pollutants spread into the atmosphere.



254 A potential benefit of the 100 m transect is that it provides more data points inside the plume when compared to the 20 m
 255 transect. However, there are clear variations in the measurements across the plume, indicating that the plume was not
 256 homogenous at this distance. This could be due to localized perturbations in the wind causing inconsistent mixing with the
 257 surrounding air mass. Furthermore, the CO_2 measurements do not follow the PN concentration measurements; with the peak
 258 being significantly broader and not returning to its expected background value of around 400 ppm. These issues indicate that
 259 more distant measurements, whilst providing more data points, potentially provide less accurate data for the calculation of
 260 emission factors. More accurate transect measurements could be achieved by slowing the UAV flight speed for transects
 261 closer to the emission source. However, this was not possible in this study as the S800 hexacopter UAV was flown at its
 262 minimum speed of 1.5 m/s during all transects.

263 3.3. PN Emission Factors

264 EF_{PN} values were calculated relative to the fuel consumption using the fuel combustion derived plume CO_2 , (Eq. 1) and the
 265 data from the nine transects listed in Table 1.

266 ΔPN was calculated by integrating the peak plume concentration (average of five data points) measured by the DISCmini,
 267 after subtraction of the background concentration. Background concentration is defined as the concentration (average of five
 268 data points) measured outside the plume. The same calculation was made to obtain the ΔCO_2 .

269 Table 2 shows, for each of the 9 transects, where the plume was captured, the measured concentration values of ΔPN
 270 and ΔCO_2 , in Kg per cubic meter, and the calculated EF_{PN} .

Day	Plume captured (distance and altitude)	$\Delta PN \text{ m}^3$	$\Delta \text{CO}_2 \text{ (Kg)}$	EF_{PN}
1	20 m; 25 m	1.94×10^{11}	1.21×10^{-4}	5.11×10^{15}
	100 m; 25 m	2.83×10^{10}	9.86×10^{-5}	9.19×10^{14}
	100 m; 35 m	4.72×10^{10}	8.88×10^{-5}	1.70×10^{15}
2	20 m; 25 m	3.07×10^{11}	2.30×10^{-4}	4.27×10^{15}
	20 m; 25 m	9.18×10^{10}	1.41×10^{-4}	2.08×10^{15}
	20 m; 25 m	4.81×10^{10}	9.55×10^{-5}	1.61×10^{15}
	20 m; 25 m	1.78×10^{11}	1.94×10^{-4}	2.94×10^{15}
	20 m; 25 m	8.05×10^{10}	8.29×10^{-5}	3.11×10^{15}
	20 m; 25 m	7.46×10^{10}	1.21×10^{-4}	1.98×10^{15}

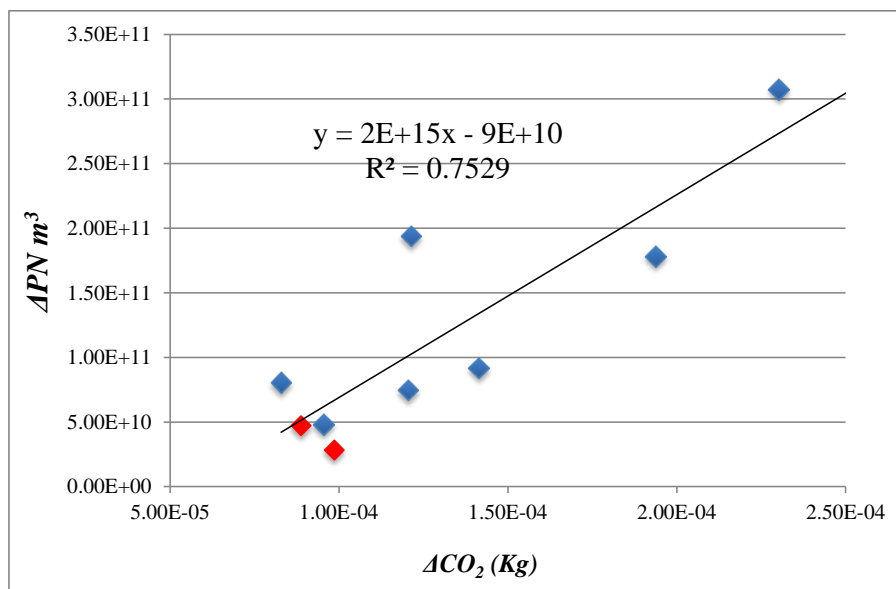
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272 **Table 2 – ΔPN and ΔCO_2 concentration emission/rate of the RV Investigator and calculated Emission Factors for PN.**

273

274 The ΔPN and ΔCO_2 values were plotted and correlated against each other as shown in Figure 5. ΔPN and ΔCO_2 were found

275 to have a good linear relationship with an R^2 value of 0.7529.



276

277 **Figure 5** $-\Delta PN$ and ΔCO_2 for the nine transects considered for the data analysis. Red markers indicate the measurements taken at
 278 **100 m behind the ship.**

279

280 The calculated EF_{PN} values for the RV Investigator ranged from 9.19×10^{14} to $5.11 \times 10^{15} \# \cdot (\text{Kg fuel})^{-1}$. The two 100 meter
 281 transects provided the lowest two emission factors measured ($9.19 \times 10^{14} \# \cdot (\text{Kg fuel})^{-1}$ and $1.70 \times 10^{15} \# \cdot (\text{Kg fuel})^{-1}$). This is
 282 likely a consequence of the noted differences between the plume measurements of the 20 and 100 m transects. The clear
 283 distinction between the background and the plume measurements of the 20 m transect indicate that the EF_{PN} calculated using
 284 them will be more representative of the RV Investigator emissions at 30% engine load. Therefore, the 100 m transects were
 285 discounted from the calculation of the mean EF_{PN} and the corresponding standard error. These values were calculated as 3.0
 286 $\times 10^{15} \pm 0.5 \times 10^{15} \# \cdot (\text{Kg fuel})^{-1}$. As presented in Table 3, this value is comparable with those reported in the literature for
 287 cruise and cargo ship plumes; which range from 0.2×10^{16} to $6.2 \times 10^{16} \# \cdot (\text{Kg fuel})^{-1}$ (Alföldy et al. 2013; Beecken et al.
 288 2014b; Jonsson et al. 2011; Juwono et al. 2013; Lack et al. 2011; Pirjola et al. 2014b; Sinha et al. 2003; Westerlund et al.
 289 2015).

290 The calculated EF_{PN} for the Investigator were lower compared to those reported by Beecken et al. (Beecken et al. 2014a) for
 291 passenger ships while accelerating ($0.91 \pm 0.18 \times 10^{16} \# \cdot (\text{Kg fuel})^{-1}$). However, the RV Investigator measurements were
 292 undertaken whilst its engine was under 30% load. Accelerating ships will typically be under higher engine loads and hence
 293 have a correspondingly higher EF_{PN} (Westerlund et al. 2015), which explains part of this discrepancy. Furthermore, the RV
 294 Investigator is a sophisticated modern vessel built for use in regions such as Antarctica. As such, it is design to have high
 295 efficiency engines, a diesel-electric energy generation system, and uses refined, ultra-low sulphur diesel fuel. These factors
 296 lead to the RV Investigator being more efficient and less polluting than most other ships at sea. This explains why the results
 297 of this study are comparable to the lower end of those found in the literature.

298 The RV investigator also uses low sulphur content diesel fuel which is similar in quality to the fuels used in the ground
 299 transport industry. In fact, the results presented here were comparable to those for in-land transportation, ranging from $4.8 \times$
 300 10^{14} (25% engine load) to 7.2 (100% engine load) $\times 10^{15} \# \cdot (\text{Kg fuel})^{-1}$ (Jayaratne et al. 2009). The calculated values for the



301 RV Investigators EF_{PN} are also close to data for commercial aircrafts during landing and taxing, which range from 4.16 to
 302 $7.74 \pm 1.46 \times 10^{15} \# (\text{Kg fuel})^{-1}$ (Mazaheri et al. 2009).

Reference	Measuring Platform	EF (PN)	Number of ships	Location
This Study	Unmanned Aerial Vehicle	0.3×10^{16}	1	Open water
Westerlund et al. (2015)	Land based	$2.35 \pm 0.20 \times 10^{16}$	154	Harbor, Ship Channel
Beecken et al. (2014)	Airborne	$1.8 \pm 1.3 \times 10^{16}$	174	Open water
Pirjola et al. (2014)	Land based	0.32×10^{16}	11	Harbor, Ship Channel
Alföldy et al. (2013)	Land based	0.8×10^{16}	497	Harbor
Juwono et al. (2012)	On board	0.22×10^{16}	2	Harbor, Ship Channel
Jonsson et al. (2011)	Land based	$2.55 \pm 0.11 \times 10^{16}$	734	Harbor
Lack et al. (2011)	Airborne	$1.0 \pm 0.2 \times 10^{16}$	1	Open water
Sinha et al. (2003)	Airborne	$6.2 \pm 0.6 \times 10^{16}$	2	Open water

303

304 **Table 3 – Comparison of the Emission Factor for the RV Investigator found in this study with other relevant values found in**
 305 **literature.**

306 4. Summary and conclusion

307 The UAV system used in this study successfully measured PN and CO₂ concentrations from the exhaust plume of the RV
 308 Investigator whilst operating at sea. Several different flight paths were tested and an optimal transect flying perpendicular to
 309 the plume at a distance of 20 meters from the ship was adopted. The EF_{PN} calculated for the RV investigator ranged from
 310 9.19×10^{14} to $5.11 \times 10^{15} \# \cdot (\text{Kg fuel})^{-1}$ relative to both consumed fuel and engine load. This EF_{PN} was within the lower end
 311 of values reported in literature, thus validating the novel UAV system for this application.

312 In comparison with other methods, the UAV system presented provides a cost effective and accessible solution for the rapid
 313 measurement and quantification of ship emissions. Its ability for deployment both in harbour and at sea, coupled with the
 314 possibility of altering its flight path to account for variances in wind conditions; gives this UAV system a distinct advantage
 315 over ground based and manned aerial vehicles. Furthermore, the UAV can sample considerably closer to the plume emission
 316 source than other methodologies, providing more accurate measurements for the calculation of EF_{PN} .

317 These attributes indicate that this UAV system provides a basis for wide-scale quantification of ultrafine particle emission
 318 factors from commercial shipping. This is critical to improve our understanding of shipping's impact on climate and health.
 319 Furthermore, it will both inform regulatory bodies, and provide them with the tools to monitor emissions in harbours and at
 320 sea.

321 4.1. Recommendations

322 The possibilities of this UAV system extend far beyond what is described here. This study is intended as both: a proof of
 323 concept; and to provide useful information both for the future of this project, as well as any other UAV sampling systems



324 being developed. The instruments on-board this system were used for the measurement of PN and CO₂ concentrations in
325 order to calculate EF_{PN} . However, this methodology could also be expanded to measure other important ship emission
326 factors, including NO_x and volatile organic compounds (VOCs).
327 Further possibilities and potential improvements can also be made to the plume transect sampling method used here. The
328 sampling error could be reduced by collecting more data points inside of the plume. One method to achieve this would be to
329 find an optimal transect distance which provides the broadest plume cross-section, whilst also providing a clear
330 differentiation between plume and the surrounding air mass. An alternative approach would be the use of a different UAV
331 with a lower minimum operational speed to increase the time of the plume transect. Other study possibilities include:
332 comparisons between EF_{PN} for different loads both in the harbour and at sea, and investigations into the use of a single flight
333 to transect multiple ship plumes.
334 The transect-based sampling approach provides researchers with a relatively simple method of capturing data inside the
335 plume. The principal flaws with this method are that there is no guarantee that the plume will be captured during a transect,
336 and the degree to which the UAV enters the plume can vary between transects. A potential answer to these issues is a non-
337 transect based approach in which the UAV system is made to hover inside the plume for a given period of time, ensuring
338 data is collected. This also allows for the collection of many more data points inside the plume, ensuring accurate and
339 repeatable data. Despite these advantages this method has proven to be challenging as it is difficult to verify whether the
340 UAV is within the plume, when it is not visible to the naked eye especially in variable wind conditions. A potential solution
341 is the implementation of sensors and instrumentation which transmit data to the ground station in real time. Using this data
342 as a feedback mechanism, it would be possible to orient the UAV position so it hovers within the plume, ensuring that more
343 accurate and repeatable data is collected on every flight.

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