

1 **Characterization of the Particle Emission from a Ship Operating at** 2 **Sea Using an Unmanned Aerial Vehicle**

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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 a
12 significant source of climatically relevant aerosol particles” on-board the RV Investigator around the Australian Great Barrier
13 Reef. Measurements of the RV Investigator exhaust plume were carried out while the ship was operating at sea, at a steady
14 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 performed,
17 providing a total of 27 horizontal transects perpendicular to the ship exhaust plume. Results show that the most appropriate
18 altitude and distance to effectively capture the plume was 25 m above sea level and 20 m downwind.

19 Particle number emission factors (EF_{PN}) 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 The calculated EF_{PN} was $7.6 \pm 1.4 \times 10^{15} \text{ \#} \cdot \text{Kg}_{\text{fuel}}^{-1}$, which is in line with those reported in the literature for ship emissions
22 ranging from $0.2 \times 10^{16} \text{ \#} \cdot \text{Kg}_{\text{fuel}}^{-1}$ to $6.2 \times 10^{16} \text{ \#} \cdot \text{Kg}_{\text{fuel}}^{-1}$.

23 This UAV system successfully assessed ship emissions to derive EF_{PN} under real world conditions. This is significant as it
24 provides a novel, relatively inexpensive and accessible way to assess ship EF_{PN} at sea.

25 **1. Introduction**

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

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

40 these gaseous pollutants through the Annex VI of the International Convention for the Prevention of Pollution from Ships –
41 the Marine Pollution Convention (MARPOL). The IMO expected that these regulations would lead to an indirect decrease in
42 particle number (PN) concentration due to the reduction of NO_x emissions and the use of fuel with lower sulphur content
43 (Chen et al., 2005). However, it has been found that the use of some low sulphur fuels lead to increased PN concentrations at
44 lower engine loads (Anderson et al., 2015), which stresses the importance for regulation specifically addressing particulate
45 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 (Corbett and Farrell,
50 2002;Corbett et al., 2007;Isakson et al., 2001;Williams et al., 2009;Reda et al., 2015;Mueller et al., 2015;Anderson et al.,
51 2015;Ristovski et al., 2012;Blasco et al., 2014). To achieve this, wide-scale evaluation of ship emission factors (EFs) is
52 necessary. EFs are commonly expressed as the amount of pollutant (x) emitted per unit mass of fuel consumed g(x). (Kg fuel)
53 ¹. Different methods have been used to investigate ship EFs, including laboratory test-bench studies, on-board measurements,
54 and measurement of ship emission plumes.

55 Test-bench studies (Anderson et al., 2015;Mueller et al., 2015;Reda et al., 2015;Petzold et al., 2010;Petzold et al., 2008;Kasper
56 et al., 2007) 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;Murphy et al., 2009;Blasco et al., 2014). To date, only a few studies have been undertaken on-board ships to
60 calculate real emission factors (Juwono et al., 2013;Hallquist 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 (Westerlund et al., 2015;Pirjola et al., 2014;Cappa et al., 2014;Beecken et al., 2014;Balzani Lööv et al.,
63 2014;Berg et al., 2012;Lack et al., 2009;Lack et al., 2008;Sinha et al., 2003) offer an alternative method of in-situ
64 measurements without requiring on-board monitoring stations. In the past the deployment cost of these systems, and the risks
65 associated with manned aircrafts have limited their feasibility. However, this has recently changed with the rapid advances
66 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;Malaver Rojas et al., 2015;Gonzalez et al., 2011). 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 measurements
74 of the RV investigators exhaust plume were taken over several flights at various altitudes and distances from the ship. Data
75 collected was used to optimize the sampling flight path and successfully quantify the RV investigators EF for PN concentration.

76 2. Methodology and Measurement system

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

81 **2.1. The RV Investigator and the voyage**

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

88 A suite of instrumentation for atmospheric research is available on the RV Investigator. This includes a radar system capable
89 of collecting weather information within a 150 km radius of the vessel, and instruments measuring: sunlight parameters; aerosol
90 composition, particle concentration and size distributions; cloud condensation nuclei; gas concentrations; and various other
91 components of the atmosphere. These instruments are housed inside two dedicated on-board laboratories for aerosol and for
92 atmospheric chemistry research. An atmospheric aerosol sample is continuously drawn into the laboratories for analysis
93 through a specialized inlet fitted to the foremast of the ship. Of particular interest to this study, the ship contains a PICARRO
94 (PICARRO Inc., Santa Clara, California, USA) G2401 analyser (CASA, 2014) that continuously measures CO₂, CO, H₂O and
95 CH₄. It has an operation range between 0-1000 ppm and a parts-per-billion sensitivity (ppb) for CO₂.

96 The two day UAV measurement study was possible as part of the RV Investigator voyage “The Great Barrier Reef as a
97 significant source of climatically relevant aerosol particles”, which started in Brisbane on the 28th of September 2016. The ship
98 was used as both: a floating platform to allow launch and recovery of the UAV system; and as the source of an exhaust plume
99 measured by the UAV system for EF calculation. During a several day stationary period on the Great Barrier Reef off the coast
100 of Australia, it was possible to measure the ship plume under stable real world conditions over two consecutive days. One of
101 the three ship engines was maintained at a steady engine load of 25 – 30 % of the maximum engine power during all
102 measurements.

103 **2.2. UAV system**

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

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

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

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

123 with a maximum altitude of approximately 120 m and within a radius of 3 nmi of an airport. UAVs in this category are not
124 permitted for research unless the research institution has been granted a permit exception. These exceptions can be granted if
125 the institution in question has or collaborates with an UAV operation team who must have: an experienced UAV pilot who is
126 also radio controller specialist; a license for commercial UAV operation; and appropriate liability insurance (CASA, 2014).
127 Queensland University of Technology (QUT) has an unmanned operator certificate and four pilots who have UAV controller
128 licenses.

129 2.2.1. Instrumentation

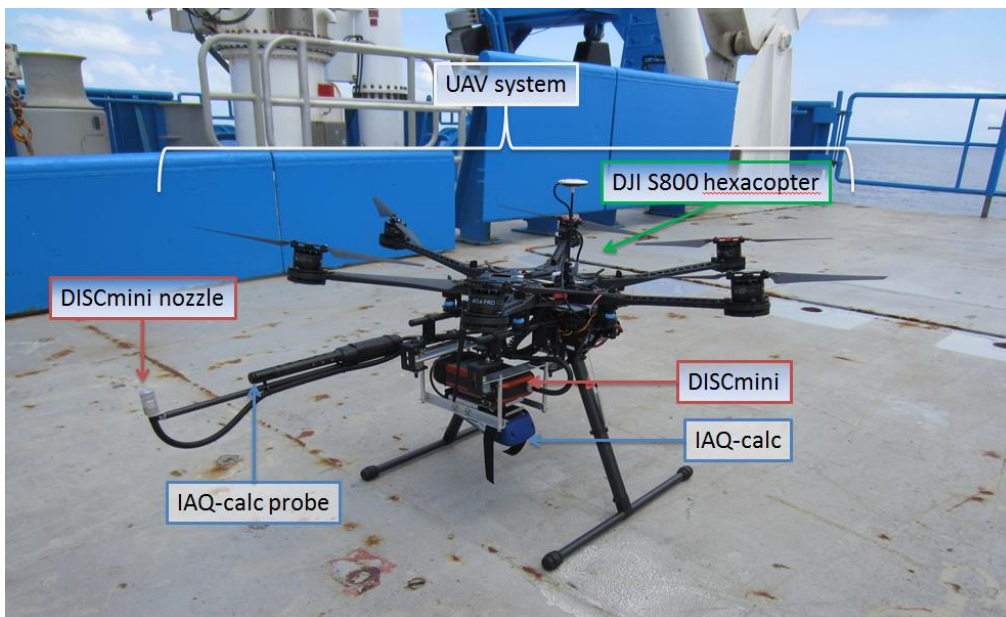
130 2.2.1.1. Instrumentation for PN concentration

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

138 2.2.1.2. Instrumentation for CO₂ concentration measurements

139 A TSI (TSI, Shoreview, Minnesota, United States) IAQ-calc 7545 model was chosen to measure CO₂ concentrations. Its sensor
140 is based on a dual-wavelength NDIR (non-dispersive infrared) with a sensitivity range between 0 to 5,000 ppm and an accuracy
141 of $\pm 3.0\%$ of reading or ± 50 ppm (whichever is greater). The measurement resolution is 1 ppm with a maximum time resolution
142 of 1 s. Similar to the DISCmini, the advantages of using the IAQ-calc are: its small dimensions (178 x 84 x 44 mm); low weight
143 (270 g, with batteries, significantly lower than the DISCmini), and a battery life of 10 hours.

144 The readings of the IAQ-calc for CO₂ were compared with those measured by the on-board PICARRO G2401 analyser.
145 Both the DISCmini and the IAQ-calc were tested and calibrated in the on-board laboratory using ambient aerosol
146 measurements at sea prior to the commencement of the measurements (Figure S2 in the Supplementary Material). All data
147 were 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**

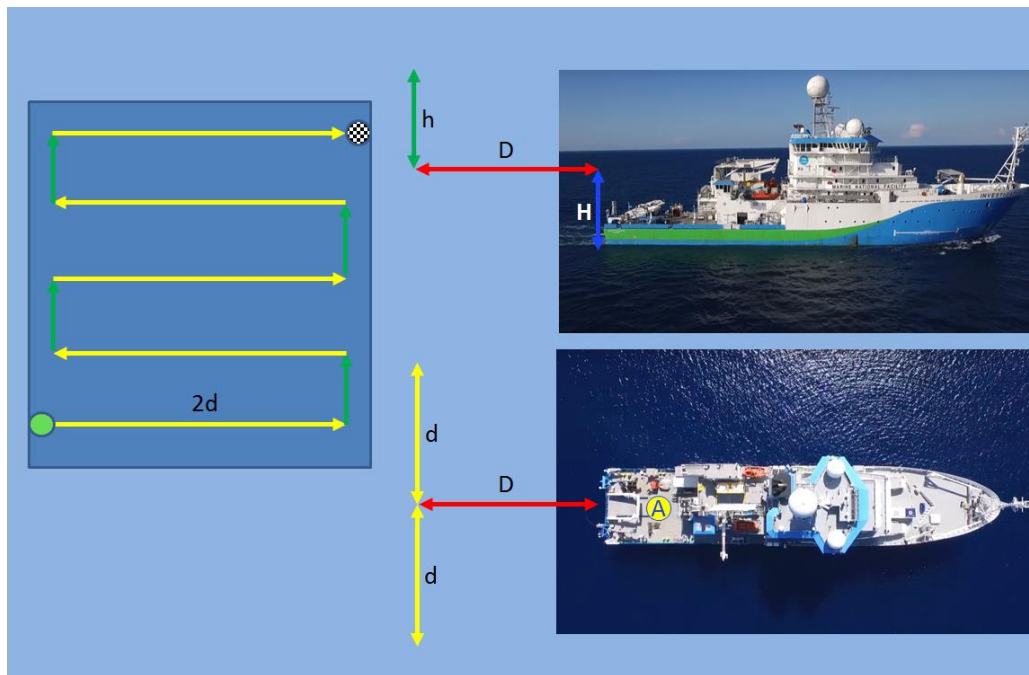
152 Meteorological data (including air temperature, relative humidity, atmospheric pressure, wind speed and direction) were
153 recorded by the RV Investigators on-board instrumentation during the entire voyage with a 60 s time interval, 24/h a day.

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. Figure
160 2 shows the programmed flight path, which consisted of a continuous flight beginning at a distance (D) and from an altitude
161 (H) above the surface. Point A, located on the back deck of the RV Investigator, represents the ‘home point’. In UAV
162 terminology this refers to the position where the UAV system takes off and lands. The UAV system was programmed to move
163 horizontally by a distance ($2d$), perpendicular to the ship, then climb vertically for 10 m (h) before flying in the opposite
164 horizontal direction for the same distance ($2d$). The UAV was then programmed to climb another 10 m (h) before repeating
165 this pattern until the UAV reached an altitude of 65 m above the ocean. During day 1, the UAV system followed three different
166 flight paths, each one with both a different distance D behind the ship (20, 50 and 100 m), and a different horizontal distance
167 $2d$ (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 point,**
173 **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., 2013; Hobbs et al., 2000) .

189 Eq.(1).

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

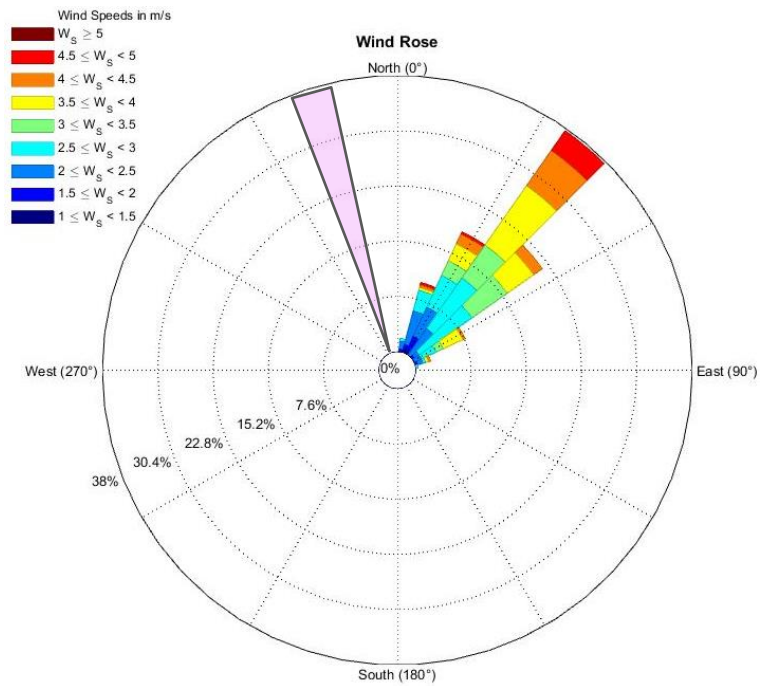
191 The ΔPN and Δgas in Eq. (1) represent the maximum particle concentration change above background in the measured
192 particle number and CO₂ concentrations, respectively. The DISCmini measurements were corrected against a reference
193 condensation particle counter (CPC). For each transect data series of PNC and CO₂, the averaged background concentration
194 were subtracted from the peak data corresponding to measurements inside the plume. The corrected peak data series were then
195 fit with a Gaussian curve using the inbuilt Matlab curve fitting application. The least absolute residuals (LAR) condition was
196 used as this most closely fits the curve to the highest magnitude data points in the series. The maximum peak height of the
197 fitted Gaussian curves were used as ΔPNC and ΔCO_2 in the calculation of emission factors for each transect.

198 **3. Results and Discussion**

199 **3.1. Meteorological and Investigator data**

200 Wind conditions were very stable during both day 1 and day 2, following one main pattern for the entire flight time. The wind
201 speed ranged from 3 - 13 m s⁻¹. The wind direction was predominantly from the NE during day 1 and ESE during day 2. The
202 wind rose graphs in Figure 3a and 3b illustrate the wind data recorded with the on-board weather instrumentation during all
203 horizontal transects flown during day 1 and 2 respectively. The prevalent wind direction was ESE, which corresponded to the
204 heading of the RV Investigator (indicated by the rose triangle). The wind direction changed occasionally to E during the flight,
205 causing the UAV to fail to capture the RV Investigator plume during some transects. As a result, 2 of the 8 horizontal transects
206 collected on day 2 were excluded from the analysis.

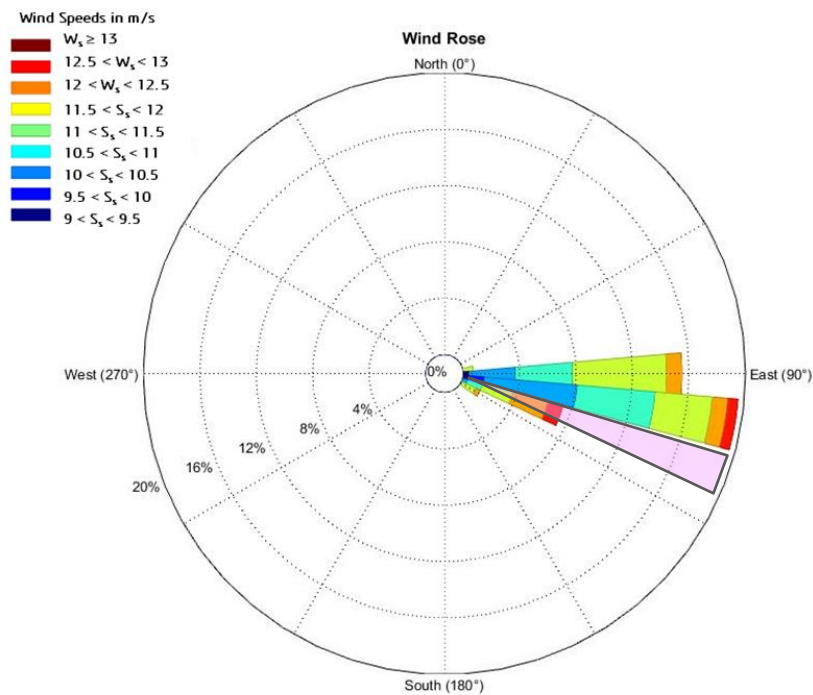
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208

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

211



212

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

215 **3.2. UAV system horizontal transects inside and outside the plume**

216 The UAV system acquired data for a total of 27 horizontal transects for day 1 and day 2. Data were collected at altitudes
 217 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

218 altitude and 20 m downwind of the ship; and again at both 25 and 35 m altitude 100 m downwind of the ship. These
 219 observations lead to the optimized flight used on day 2, which started downwind at 25 m above the surface and 20 m behind
 220 the ship. On day 2 the UAV system successfully captured the plume during 6 of the 8 transects performed. Across the two
 221 days this lead to a total of 9 transects that captured the plume and which have been considered for discussion, shown in Table
 222 1.

223

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

224

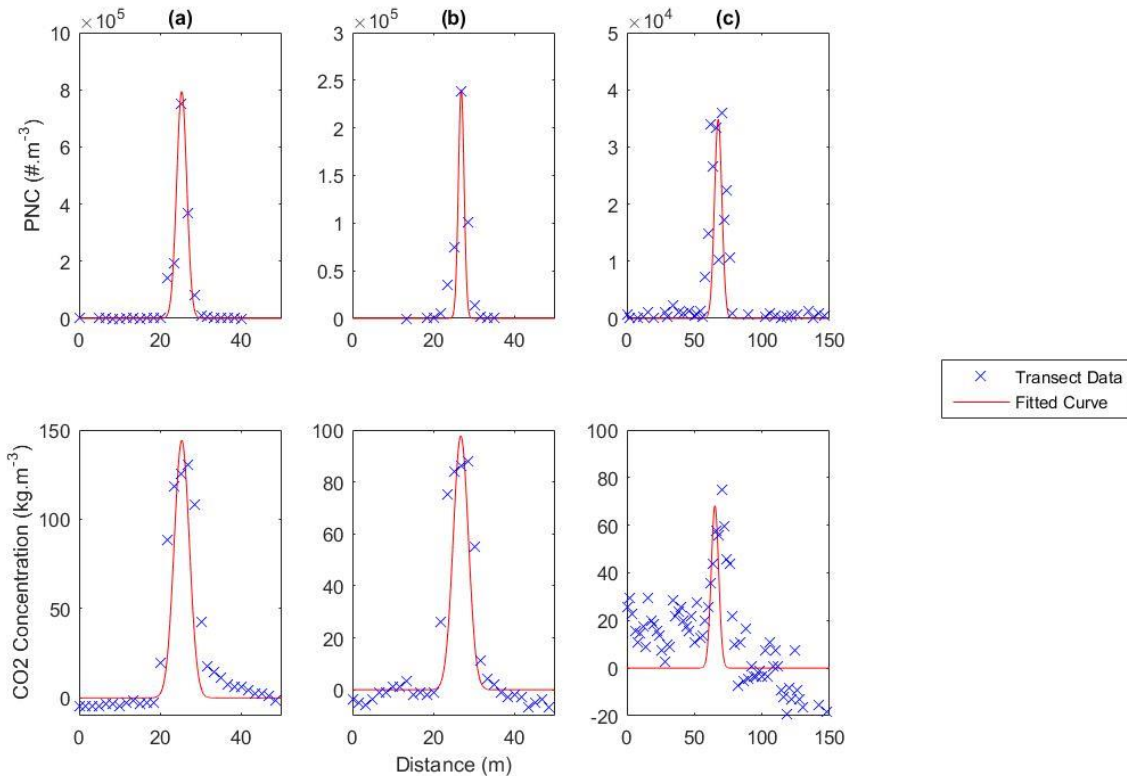
225 **Table 1 – Specifications of the transects considered for the data analysis. The (*) indicates the transect of Day 1 of which PN**
 226 **concentration and CO₂ profiles are presented in Figure 4.**

227

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

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

233 The profiles in (c) show that the PN concentration was four orders of magnitude greater inside the plume at 100 m behind the
 234 ship and that the CO₂ concentration was up to 70 ppm higher inside the plume.



235

236 Figure 4 – (a) and (b) show the measured PN and CO₂ concentration profiles and fitted Gaussian curves for two different transects
 237 20 m behind the ship 25 m above the surface during day 2. (c) shows the PN and CO₂ concentration profiles and fitted Gaussian
 238 curves collected during flight 3 of day 1 at 100 m behind the ship, 25 m above the surface.

239

240 Figure 4 (a) and (b) both show transects at 25 m altitude and 20 m behind the ship. Both the PN concentration and CO₂
 241 measurements show clear, single peaks as the UAV crosses the plume. As a consequence, these transects show a good fit with
 242 the corresponding Gaussian distribution curves with R² values of above 0.9 for both PNC and CO₂. In contrast Figure 4 (c)
 243 shows substantially less defined, wider peaks with lower pollutant concentrations. This is attributed to a difference in flight
 244 paths, with Figure 4 (c) representing data from a transect 100 m behind the ship. The additional time between emission and
 245 sampling has allowed the plume to broaden, become less homogenous, and take on a skewed cross-section. This results in a
 246 significantly lower R² value for the fitted Gaussian curves, with a value of 0.4998 for the CO₂ data in this transect. Therefore,
 247 whilst the 100 m transect does provide more data points inside the plume, the randomized variations inside the plume lead to
 248 less accurate calculations of emission factors.

249 Of further note in Figure 4, the maximum PN concentrations measured in (a) ($7.5 \times 10^5 \text{ \#} \cdot \text{cm}^{-3}$) is approximately three times
 250 greater than those in (b) ($2.4 \times 10^5 \text{ \#} \cdot \text{cm}^{-3}$) and the CO₂ concentrations in (a) are 43 ppm greater than (b). The transect flight plan
 251 and ship engine load remained constant throughout these measurements. The variations between (a) and (b) are attributed to
 252 several factors which reduce the effectiveness of the UAV transect for capturing the plume. Slight changes in ambient
 253 conditions such as temperature, wind direction and intensity will alter the path of the plume as it moves away from the ship.
 254 The UAVs automated flight path cannot account for these variations. Therefore, the degree to which the UAV enters the plume,
 255 and thus the concentrations it measures, will be different on each transect. Both CO₂ and PN concentration measurements will
 256 be similarly affected by this variance. However, differences in instrument response rates in conjunction with these variances
 257 will be one of the major contributors to variations in calculated emission factors.

258 3.3. PN Emission Factors

259 Table 2 shows the distance and altitude of each transect, the R² values of the fitted Gaussian curves for PNC and CO₂ data, the
 260 calculated values of ΔPNC and ΔCO_2 , and the calculated EF_{PN}.

Day	Dist/Alt (m)	R ² _{PNC}	R ² _{CO₂}	ΔPNC (#·m ⁻³)	ΔCO_2 (kg·m ⁻³)	EF _{PN} (#·kg _{fuel} ⁻¹)
1	100/25	0.9586	0.4998	5.05E+11	9.35E-05	1.73E+16
	100/35	0.4767	0.8967	4.8E+10	1.34E-04	1.15E+15
	20/25	0.9856	0.8915	1.09E+11	7.74E-05	4.52E+15
2	20/25	0.9842	0.9518	1.06E+12	2.83E-04	1.20E+16
	20/25	0.9852	0.8838	3.3E+11	1.92E-04	5.51E+15
	20/25	0.9489	0.9246	1.78E+11	1.11E-04	5.16E+15
	20/25	0.9721	0.8965	3.6E+11	2.23E-04	5.18E+15
	20/25	0.9508	0.8473	1.47E+11	1.31E-04	3.59E+15
	20/25	0.8517	0.6743	1.01E+11	9.68E-05	3.32E+15

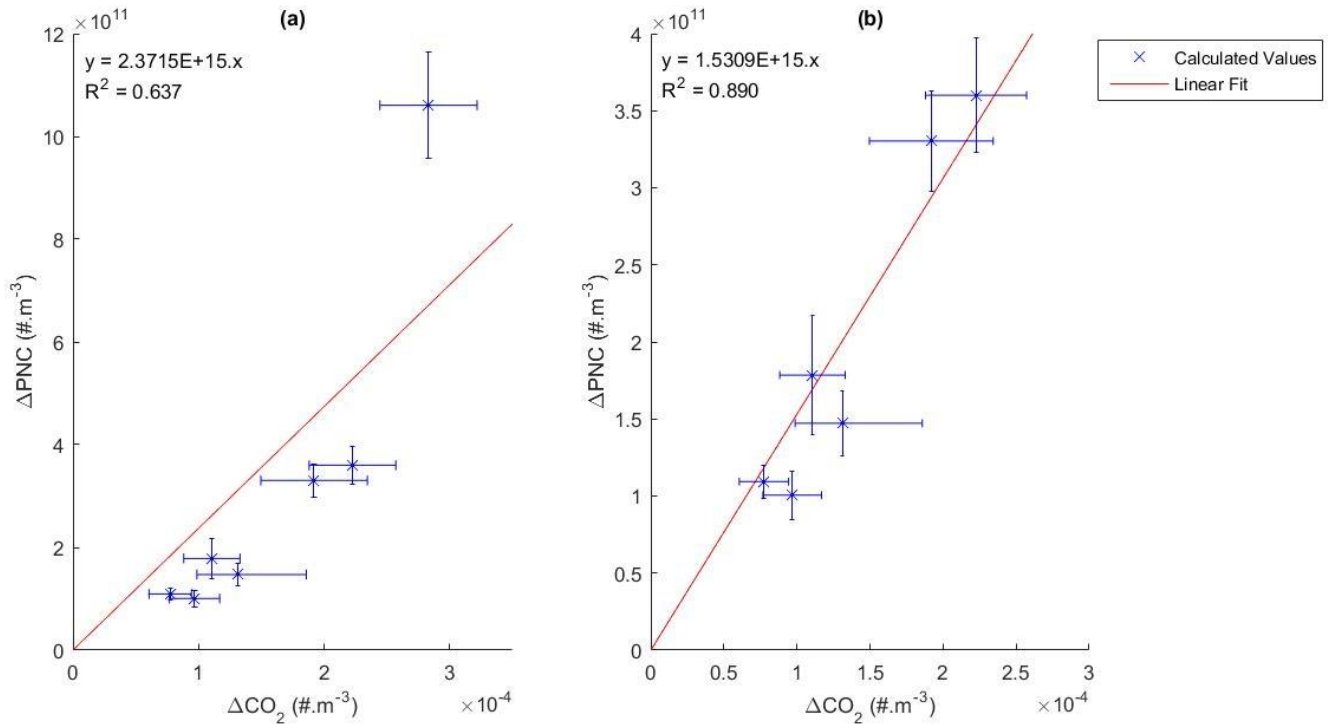
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262 Table 2 – Transect flight days and details, R² values for the Gaussian curve fits to both PNC and CO₂ data, ΔPNC and ΔCO_2
 263 concentration emission/rate of the RV Investigator, and calculated Emission Factors for PN.

264

265 The calculated EF_{PN} values for the RV Investigator ranged from 1.15×10^{15} to $1.73 \times 10^{16} \text{ \#} \cdot \text{kg}_{\text{fuel}}^{-1}$. The two 100 m transects
 266 provided the worst Gaussian fits as well as the highest and lowest calculated emission factors. This indicates that it is important
 267 to filter out transects with data which does not fit the expected Gaussian distribution suitably as they can generate significant

268 error. To this end, the 100 m transects were excluded from further analysis. Δ PNC and Δ CO₂ values for remaining transects
 269 were plotted against each other as shown in Figure 5.



270

271 **Figure 5 –(a) Δ PNC against Δ CO₂ with 95% confidence interval for the six transects considered for the data analysis. (b) Δ PNC against**
 272 **Δ CO₂ with 95% confidence interval with the removal of the outlier transect from the first flight of day 2**

273

274 Figure 5 (a) and (b) show the plots of the remaining transects Δ PNC against Δ CO₂ with and without the values of the first
 275 flight of day 2. This transect represents a clear outlier in the linear trend, with the R^2 value of the linear fit increasing from
 276 0.637 to 0.890 with its exclusion. Furthermore, whilst the linear fit falls within the confidence interval of only one point in (a),
 277 it falls within all data points confidence intervals in (b). This occurs despite both R^2 values for the fitted Gaussians of this
 278 transect being very high ($R^2_{\text{PNC}} = 0.9842$, $R^2_{\text{CO}_2} = 0.9518$). This highlights a limitation with this methodology which can be
 279 best observed in the difference between Figure 4 (a) and (b). The combination of UAV velocity, sampling rate and response
 280 time of the DISCmini results in the PNC transect data having only one data point defining the peak height of the transect.
 281 Relying on a single sample point leads to the potential for random instrumentation effects heavily biasing results in a way
 282 which does not strongly impact the R^2 values of Gaussian fits used to identify successful transects. Therefore, it is unclear
 283 whether this is a variation in the ship emissions or an instrumentation error.

284 The slope and standard error of the linear fit for Figure 4 (a) was input unto Equation 1 to calculate an overall emission factor
 285 of $7.6 \pm 1.4 \times 10^{15} \text{ \#} \cdot \text{kg}_{\text{fuel}}^{-1}$. As presented in Table 3, this value is comparable with those reported in the literature for cruise
 286 and cargo ship plumes; which range from 0.2×10^{16} to $6.2 \times 10^{16} \text{ \#} \cdot \text{Kg}_{\text{fuel}}^{-1}$ (Alföldy et al., 2013;Beecken et al., 2014;Jonsson
 287 et al., 2011;Juwono et al., 2013;Lack et al., 2011;Lack et al., 2009;Pirjola et al., 2014;Sinha et al., 2003;Westerlund et al.,
 288 2015)

289

Reference	Platform	EFPN (#.kg _{fuel} ⁻¹)	Number of ships	Location
This Study	UAV	$7.6 \pm 1.4 \times 10^{15}$	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. (2009)	Ship	$0.71 \pm 0.55 \times 10^{16}$ (>13nm)* $1.27 \pm 0.95 \times 10^{16}$ (>5nm)**	172 165	Open Water, Shipping Channel
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

290

291 **Table 3 – Comparison of the Emission Factor for the RV Investigator found in this study with other relevant values found in**
 292 **literature. * P_{NEF} for particles above 13nm. ** P_{NEF} for particles above 5nm.**

293 The calculated EF_{PN} for the Investigator was lower compared to those reported by Beecken et al. (Beecken et al., 2014) for
 294 passenger ships while accelerating ($0.91 \pm 0.18 \times 10^{16}$ #.Kg_{fuel}⁻¹). However, the RV Investigator measurements were undertaken
 295 whilst its engine was under 30% load. Accelerating ships will typically be under higher engine loads and hence have a
 296 correspondingly higher EF_{PN} (Westerlund et al., 2015), which explains part of this discrepancy. Furthermore, the RV
 297 Investigator has high efficiency engines and utilizes ultra-low sulphur diesel fuel. Studies have shown that similar diesel
 298 engines burning fuel of this type have lower EF_{PN} than the same engine with higher sulphur content diesel (Chu-Van et al.,
 299 2017). Similar quality fuels used in the ground transport industry have yielded similar values of EF_{PN} , ranging from 4.8×10^{14}
 300 (25% engine load) to 7.2 (100% engine load) $\times 10^{15}$ #.Kg_{fuel}⁻¹ (Jayaratne et al., 2009).

301 3.4. Instrumentation Limitations

302 Lightweight UAVs have the potential to achieve aerial measurements at significantly less upfront and operational costs than
 303 fixed wing and manned aerial vehicles. Lightweight UAVs can be deployed faster with limited or no required launch and
 304 landing area compared to their manned and fixed wing counterparts. Yet, their primary disadvantage, particularly in this
 305 application, is a severely limited payload weight. To overcome this limitation, this project used the lightweight and portable
 306 DICSmini and IAQ-calc sensors. However, these instruments have lower sensitivities and greater uncertainties when compared
 307 to a high accuracy CPC and CO₂ monitor for measurements, which can influence results.

308 The DISCmini has a manufacturer listed measurement cut-off size of 10 nm. A previous study listed in Table 3 (Lack et al.,
 309 2009) shows that the cut-off size of instruments used to measure PNC is directly linked to the value of EF_{PN} , with the measured
 310 EF_{PN} doubling when the cut-off size is changed from 13 nm to 5 nm due to the large number of particles in this size range.
 311 This may have been another contributing factor to the EF_{PN} measured in this study being in the lower end of measured values
 312 in literature.

313 The two 100m transects were not accounted for in the final calculation of EF_{PN} due to their poor Gaussian curve fits. Whilst
 314 this has been attributed to the skewing of the plume at this distance, the limitations of the instrumentation could also have
 315 contributed. The lower concentrations of CO₂ at this distance result in the difference above background inside the plume being
 316 the same order of magnitude as the manufacturer specified error margin. Hence, the variability in the plume either side of the
 317 central peak as shown in figure 4 (c) could be due in part to instrumentation error.

318 Calibrations of sensors in this study were performed by comparison with reference instruments for ambient measurements at
319 sea. Ideally, calibration should be performed with in-plume measurements to have the same environmental conditions and
320 range as the real measurements. However, it was not possible to access the plume with reference instrumentation on board the
321 ship. Whilst this study provides a successful proof of concept with consistent results over two days and several flights, a
322 validation study is needed. This should include independent measurements of EF_{PN} using other established methodologies to
323 ascertain more precise correction factors and uncertainties.

324 **4. Summary and conclusion**

325 The UAV system used in this study successfully measured PN and CO₂ concentrations from the exhaust plume of the RV
326 Investigator whilst operating at sea. Several different flight paths were tested and an optimal transect flying perpendicular to
327 the plume at a distance of 20 meters from the ship was adopted. The EF_{PN} calculated for the RV investigator was $7.6 \pm 1.4 \times$
328 $10^{15} \text{ \#.kg}_{\text{fuel}}^{-1}$ at a constant 30 % engine load. This EF_{PN} was in agreement with values reported in literature, indicating this
329 UAV based system has potential for EF_{PN} quantification pending further evaluation.

330 In comparison with other methods, UAV systems have the potential to provide a cost effective and accessible solutions for the
331 rapid measurement and quantification of ship EF_{PNS} . Its ability for deployment both in harbour and at sea, coupled with the
332 possibility of altering its flight path to account for variances in wind conditions; gives this UAV system a high degree of
333 flexibility. Furthermore, the UAV can sample considerably closer to the plume emission source than other methodologies,
334 providing higher concentration measurements for the calculation of EF_{PN} .

335 Whilst further validation is necessary, results present here indicate that this UAV system has the potential to be used a low
336 cost tool for quantification of ultrafine particle emission factors from commercial shipping. This is critical to improve our
337 understanding of shipping's impact on climate and health.

338 **4.1. Recommendations**

339 The potential of this UAV system extend far beyond what is described here. This study is intended as both: a proof of concept;
340 and to provide useful information both for the future of this project, as well as any other UAV sampling systems being
341 developed. The most significant improvement to the method described would be the use a UAV with a lower minimum
342 airspeed. This would allow for more data points per transect and would minimize the impact potential outliers in
343 instrumentation data. Other related improvements to this include: the use of different sensors with higher response rates; and
344 additional flightpath investigations to find an optimal transect distance which provides the broadest plume cross-section,
345 without the plume becoming distorted and impacting accuracy.

346 Further optimization of the transect approach is necessary in order to conduct UAV measurements of shipping emissions on a
347 larger scale. After location of the plume the system could be set to make several repeat passes across the plume in rapid
348 succession to increase the sample size. Another alternative would involve the UAV hovering inside the plume over a period
349 of time collecting a continuous series of measurements from the centre of the plume. These methods would both require real
350 time sensor feedback to the UAV pilot and potentially adaptive autonomous controls to achieve a suitable result. Further
351 challenges include: operation under less favourable weather conditions; measurements in which the UAV is not launched from
352 the ship itself; and measurements taken for ships moving at full speed. This methodology could also be expanded to measure
353 other important ship emission factors, including NOX and volatile organic compounds (VOCs).

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