

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

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10 **Abstract.** This research demonstrates the use of an unmanned aerial vehicle (UAV) to characterize the gaseous (CO<sub>2</sub>) 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<sub>PN</sub>) 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 CO<sub>2</sub> 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<sub>PN</sub> under real world conditions. This is significant as it  
24 provides a novel, relatively inexpensive and accessible way to assess ship EF<sub>PN</sub> 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 (Fuglestad 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<sub>x</sub> and SO<sub>2</sub>. 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<sub>x</sub> 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 <sup>1</sup>. 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 CO<sub>2</sub> 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 CO<sub>2</sub> concentration emitted by the RV Investigator were taken using a PN and CO<sub>2</sub>  
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,  
83 ~~geoscience~~ geoscience and oceanographic research. The vessel is 94 m long, has a gross weight of 6,082 tons, a fuel capacity  
84 of 700 tons of ultra-low sulphur diesel fuel. It is powered by three 9 cylinder 3000 kW MaK diesel engines, each coupled to a  
85 690V AC Generator. Ship propulsion is achieved using two 2600 kW L3 AC reversible propulsion motors powered by these  
86 generators. The RV Investigator can host up to 30 crew members and 35 researchers for a maximum voyage period of 60 days  
87 with a maximum 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 CO<sub>2</sub>, CO, H<sub>2</sub>O and  
95 CH<sub>4</sub>. It has an operation range between 0-1000 ppm and a parts-per-billion sensitivity (ppb) for CO<sub>2</sub>.

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 28<sup>th</sup> 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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,

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

## 157 2.2.1. Instrumentation

### 158 2.2.1.1. Instrumentation for PN concentration

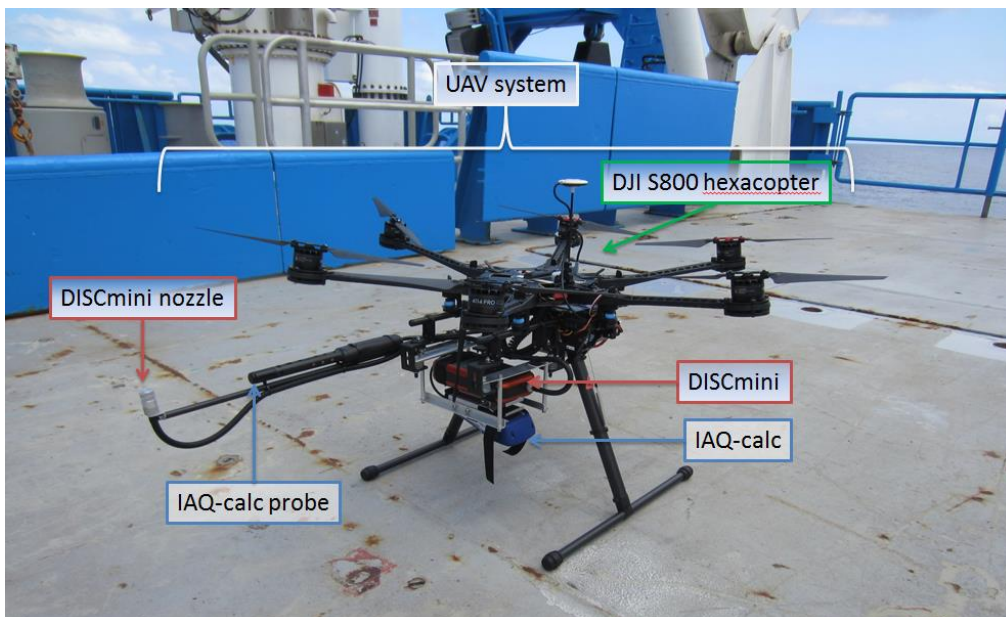
159 This study measured PN concentration using a Mini Diffusion Size Classifier (DISCmini), developed by the University of  
160 Applied Sciences, Windisch, Switzerland (Fierz et al., 2008). The DISCmini is a portable monitor used to measure  
161 concentration of particles in the 10-500 nm diameter size range, with a time resolution of up to 1 s (1 Hz). It can measure PN  
162 concentrations between  $10^3$  and  $10^6$  N/cm<sup>3</sup>. Measurement accuracy is dependent upon the particle shape, size distribution, and  
163 number concentration. The advantages of using the DISCmini are its relatively small dimensions (180 x 90 x 40 mm), low  
164 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  
165 be easily integrated on the UAV.

### 166 2.2.1.2. Instrumentation for CO<sub>2</sub> concentration measurements

167 A TSI (TSI, Shoreview, Minnesota, United States) IAQ-calc 7545 model was chosen to measure CO<sub>2</sub> concentrations. Its sensor  
168 is based on a dual-wavelength NDIR (non-dispersive infrared) with a sensitivity range between 0 to 5,000 ppm and an accuracy  
169 of  $\pm 3.0\%$  of reading or  $\pm 50$  ppm (whichever is greater). The measurement resolution is 1 ppm with a maximum time resolution  
170 of 1s. Similar to the DISCmini, the advantages of using the IAQ-calc are: its small dimensions (178 x 84 x 44 mm); low weight  
171 (270 g, with batteries, significantly lower than the DISCmini), and a battery life of 10 hours.

172 The readings of the IAQ-calc for CO<sub>2</sub> were compared with those measured by the on-board PICARRO G2401 analyser.

173 Both the DISCmini and the IAQ-calc were tested and calibrated in the on-board laboratory using ambient aerosol  
174 measurements at sea prior to the commencement of the measurements (Figure S2 in the Supplementary Material). All data  
175 were logged with a 1 s time interval.



176  
177 **Figure 1. The UAV system with the on-board instrumentation: the DISCmini and the IAQ-calc.**  
178

179 **2.3. Meteorological data**

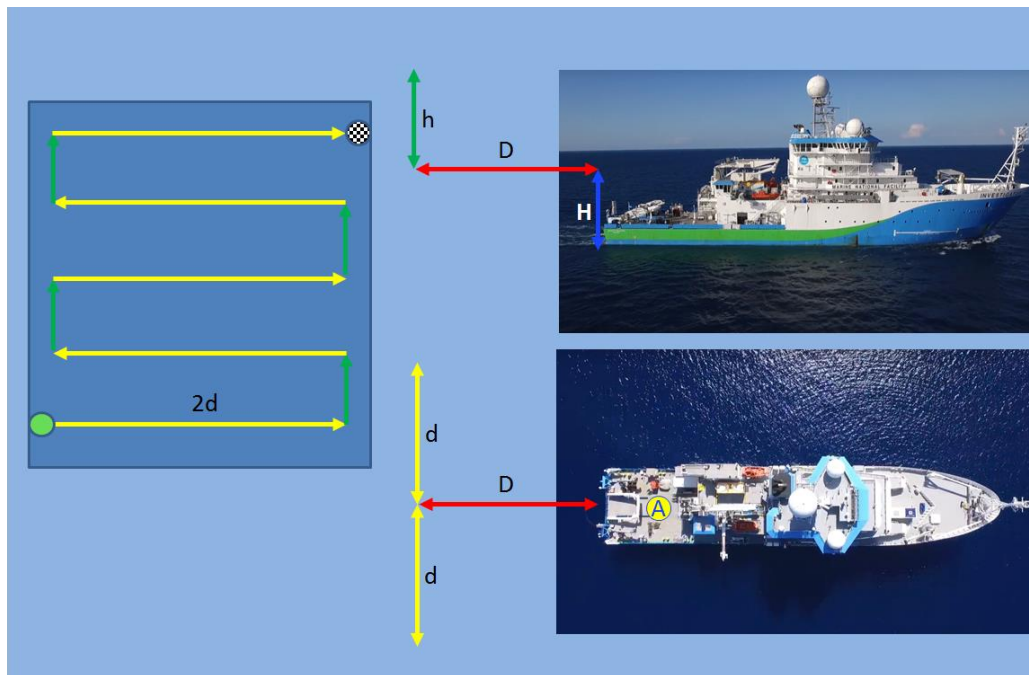
180 Meteorological data (including air temperature, relative humidity, atmospheric pressure, wind speed and direction) were  
181 recorded by the RV Investigators on-board instrumentation during the entire voyage with a 60 s time interval, 24/h a day.

182 **2.4. Study design**

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

187 Day 1 was used to optimise the study design, focusing on finding the flight path most suitable to capture the ship plume. Figure  
188 2 shows the programmed flight path, which consisted of a continuous flight beginning at a distance ( $D$ ) and from an altitude  
189 ( $H$ ) above the surface. Point A, located on the back deck of the RV Investigator, represents the 'home point'. In UAV  
190 terminology this refers to the position where the UAV system takes off and lands. The UAV system was programmed to move  
191 horizontally by a distance ( $2d$ ), perpendicular to the ship, then climb vertically for 10 m ( $h$ ) before flying in the opposite  
192 horizontal direction for the same distance ( $2d$ ). The UAV was then programmed to climb another 10 m ( $h$ ) before repeating  
193 this pattern until the UAV reached an altitude of 65 m above the ocean. During day 1, the UAV system followed three different  
194 flight paths, each one with both a different distance  $D$  behind the ship (20, 50 and 100 m), and a different horizontal distance  
195  $2d$  (50, 100 and 150 m).

196 The optimised flight path for day 2 started 20 m behind the ship and 25 m above the surface, with no altitude variation. The  
197 UAV path was limited to a continuous horizontal flight of 50 m ( $2d$ ) at steady speed of 2 m s<sup>-1</sup>. This path and flying speed  
198 allowed up to 4 horizontal transects to capture the ship plume.



199  
200 **Figure 2. Flight path used to capture the plume:  $H$  - height from the ocean,  $D$  – distance behind the ship to the flight beginning point,**  
201  **$h$  – rising altitude after the horizontal transect,  $2d$  – full length of the horizontal transect**

202 **2.5. Experimental procedure**

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

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

## 210 2.6. Emission factors

211 The calculation of an emission factor for particle number concentration ( $EF_{PN}$ ) from the collected ship plume measurements  
212 was performed using Eq. (1). This method has previously been used for ship (Westerlund et al., 2015), road vehicle (Hak et  
213 al., 2009) and aircraft (Mazaheri et al., 2009) emissions. The measured values of PN concentration were related to the amount  
214 of fuel consumed by the engine in question through the use of the simultaneous measurements of CO<sub>2</sub> concentration taken by  
215 the UAV. This was achieved by using a published value for a ship emission factor of CO<sub>2</sub> ( $EF_{gas}$ ) of 3.2 Kg CO<sub>2</sub> (Kg fuel)<sup>-1</sup>  
216 (Hallquist et al., 2013; Hobbs et al., 2000) .

217 Eq.(1).

$$218 \quad EF_{PN} = \frac{\Delta PN}{\Delta gas} \times EF_{gas} \quad (1)$$

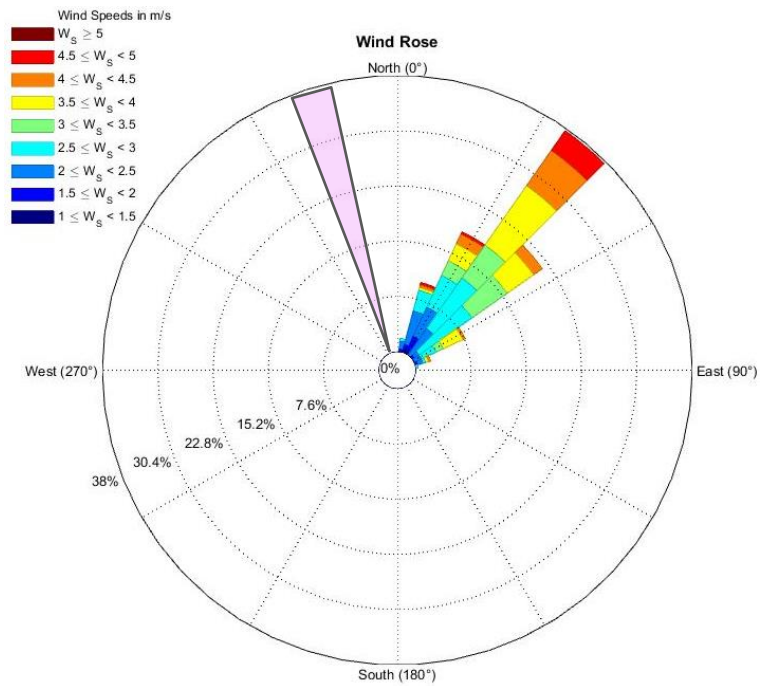
219 The  $\Delta PN$  and  $\Delta gas$  in Eq. (1) represent the maximum particle concentration change above background in the measured  
220 particle number and CO<sub>2</sub> concentrations, respectively. The DISCmini measurements were corrected against a reference  
221 [condensation particle counter \(CPC\)](#). For each transect data series of PNC and CO<sub>2</sub>, the averaged background concentration  
222 were subtracted from the peak data corresponding to measurements inside the plume. The corrected peak data series were then  
223 fit with a Gaussian curve using the inbuilt Matlab curve fitting application. The least absolute residuals (LAR) condition was  
224 used as this most closely fits the curve to the highest magnitude data points in the series. The maximum peak height of the  
225 fitted Gaussian curves were used as  $\Delta PNC$  and  $\Delta CO_2$  in the calculation of emission factors for each transect.

## 226 3. Results and Discussion

### 227 3.1. Meteorological and Investigator data

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

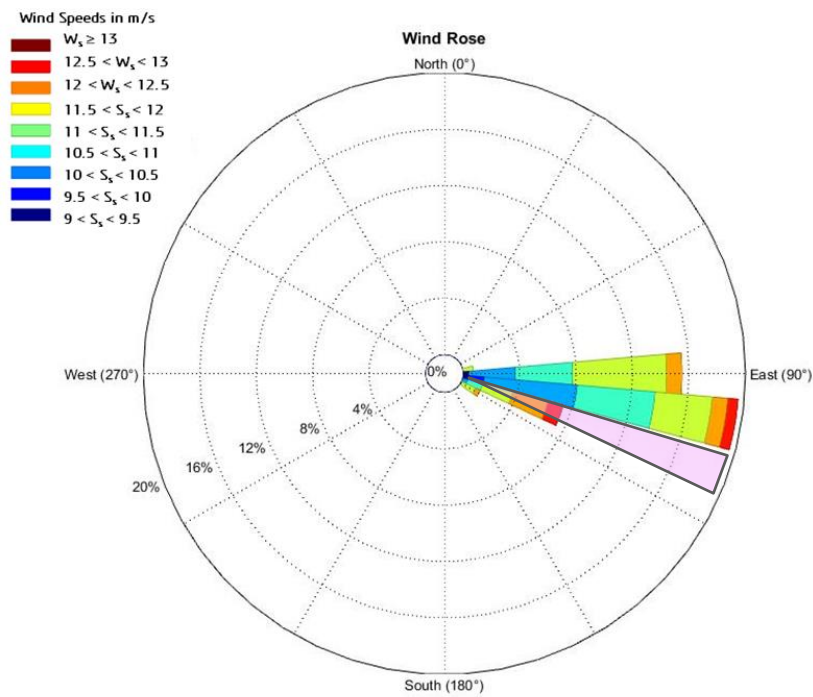
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236

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

239



240

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

243 **3.2. UAV system horizontal transects inside and outside the plume**

244 The UAV system acquired data for a total of 27 horizontal transects for day 1 and day 2. Data were collected at altitudes  
 245 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

246 altitude and 20 m downwind of the ship; and again at both 25 and 35 m altitude 100 m downwind of the ship. These  
 247 observations lead to the optimized flight used on day 2, which started downwind at 25 m above the surface and 20 m behind  
 248 the ship. On day 2 the UAV system successfully captured the plume during 6 of the 8 transects performed. Across the two  
 249 days this lead to a total of 9 transects that captured the plume and which have been considered for discussion, shown in Table  
 250 1.

251

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

252

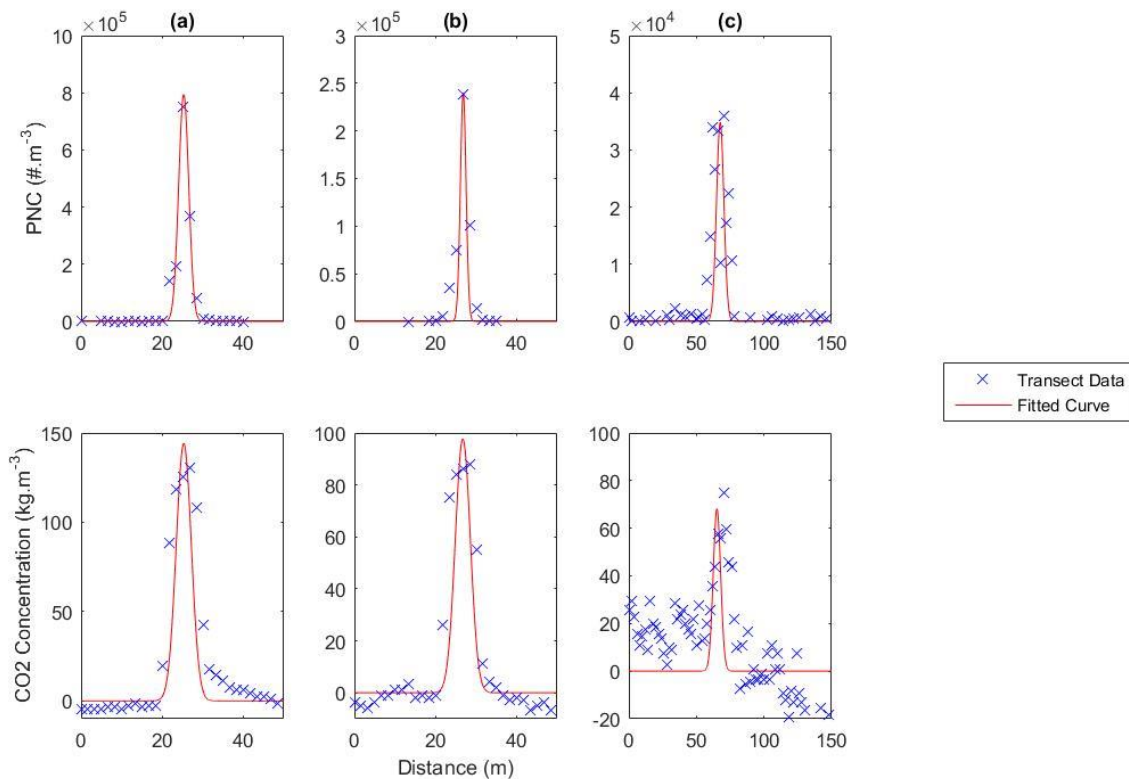
253 **Table 1 – Specifications of the transects considered for the data analysis. The (\*) indicates the transect of Day 1 of which PN**  
 254 **concentration and CO<sub>2</sub> profiles are presented in Figure 4.**

255

256 Figure 4 shows the PN concentration and CO<sub>2</sub> profiles, collected during two (a; b) transects on day 2, and (c) during one  
 257 transect of day 1 (Spec. in Table 1, Day1\*).

258 The PN concentration profiles for the (a) and (b) transects in Figure 4 show that the concentration varied by five orders of  
 259 magnitude between the outside and inside the plume, while the CO<sub>2</sub> profiles show an increase up to 140 ppm above the  
 260 background.

261 The profiles in (c) show that the PN concentration was four orders of magnitude greater inside the plume at 100 m behind the  
 262 ship and that the CO<sub>2</sub> concentration was up to 70 ppm higher inside the plume.



263



264 **Figure 4 – (a) and (b) show the measured PN and CO<sub>2</sub> concentration profiles and fitted Gaussian curves for two different transects**  
 265 **20 m behind the ship 25 m above the surface during day 2. (c) shows the PN and CO<sub>2</sub> concentration profiles and fitted Gaussian**  
 266 **curves collected during flight 3 of day 1 at 100 m behind the ship, 25 m above the surface.**

267

268 Figure 4 (a) and (b) both show transects at 25 m altitude and 20 m behind the ship. Both the PN concentration and CO<sub>2</sub>  
 269 measurements show clear, single peaks as the UAV crosses the plume. As a consequence, these transects show a good fit with  
 270 the corresponding Gaussian distribution curves with R<sup>2</sup> values of above 0.9 for both PNC and CO<sub>2</sub>. In contrast Figure 4 (c)  
 271 shows substantially less defined, wider peaks with lower pollutant concentrations. This is attributed to a difference in flight  
 272 paths, with Figure 4 (c) representing data from a transect 100 m behind the ship. The additional time between emission and  
 273 sampling has allowed the plume to broaden, become less homogenous, and take on a skewed cross-section. This results in a  
 274 significantly lower R<sup>2</sup> value for the fitted Gaussian curves, with a value of 0.4998 for the CO<sub>2</sub> data in this transect. Therefore,  
 275 whilst the 100 m transect does provide more data points inside the plume, the randomized variations inside the plume lead to  
 276 less accurate calculations of emission factors.

277 Of further note in Figure 4, the maximum PN concentrations measured in (a) ( $7.5 \times 10^5 \# \cdot \text{cm}^{-3}$ ) is approximately three times  
 278 greater than those in (b) ( $2.4 \times 10^5 \# \cdot \text{cm}^{-3}$ ) and the CO<sub>2</sub> concentrations in (a) are 43 ppm greater than (b). The transect flight plan  
 279 and ship engine load remained constant throughout these measurements. The variations between (a) and (b) are attributed to  
 280 several factors which reduce the effectiveness of the UAV transect for capturing the plume. Slight changes in ambient  
 281 conditions such as temperature, wind direction and intensity will alter the path of the plume as it moves away from the ship.  
 282 The UAVs automated flight path cannot account for these variations. Therefore, the degree to which the UAV enters the plume,  
 283 and thus the concentrations it measures, will be different on each transect. Both CO<sub>2</sub> and PN concentration measurements will  
 284 be similarly affected by this variance. However, differences in instrument response rates in conjunction with these variances  
 285 will be one of the major contributors to variations in calculated emission factors.

### 286 3.3. PN Emission Factors

287 Table 2 shows the distance and altitude of each transect, the R<sup>2</sup> values of the fitted Gaussian curves for PNC and CO<sub>2</sub> data, the  
 288 calculated values of  $\Delta\text{PNC}$  and  $\Delta\text{CO}_2$ , and the calculated EF<sub>PN</sub>.

Day	Dist/Alt (m)	R <sup>2</sup> <sub>PNC</sub>	R <sup>2</sup> <sub>CO<sub>2</sub></sub>	$\Delta\text{PNC}$ (#·m <sup>-3</sup> )	$\Delta\text{CO}_2$ (kg·m <sup>-3</sup> )	EF <sub>PN</sub> (#·kg <sub>fuel</sub> <sup>-1</sup> )
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

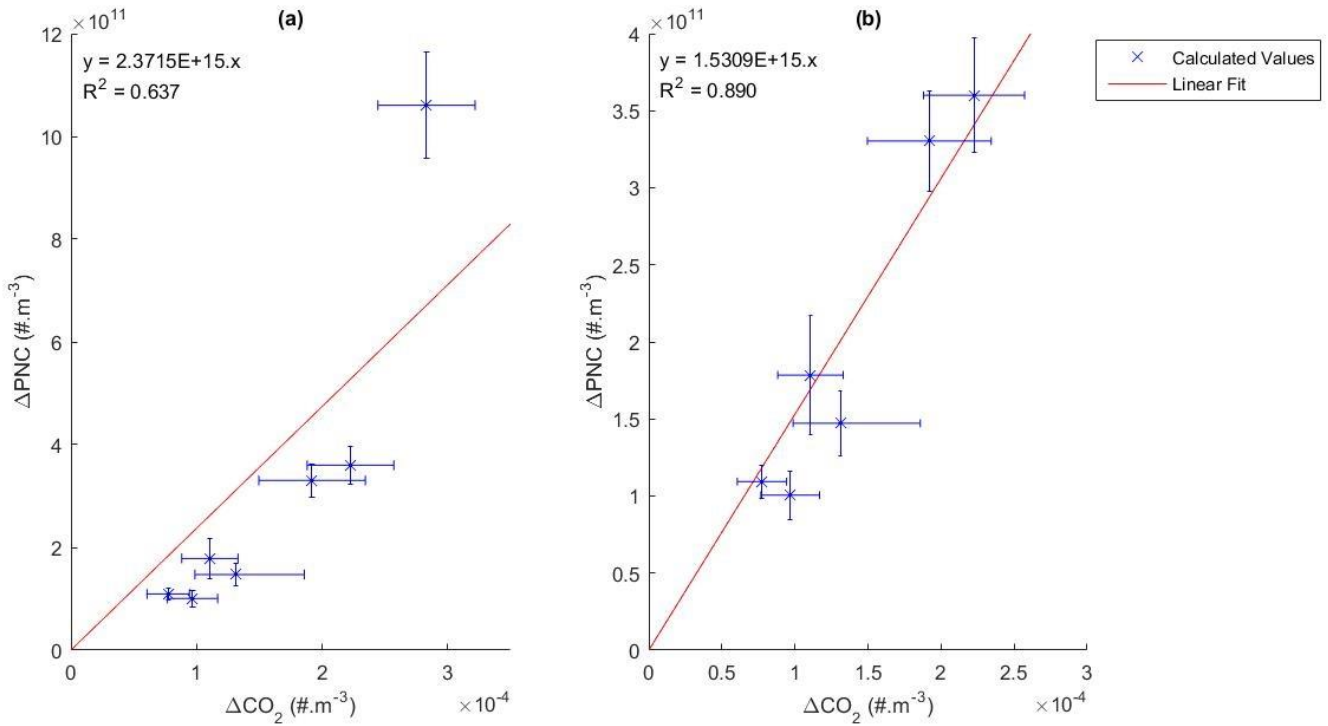
289

290 **Table 2 – Transect flight days and details, R<sup>2</sup> values for the Gaussian curve fits to both PNC and CO<sub>2</sub> data,  $\Delta\text{PNC}$  and  $\Delta\text{CO}_2$**   
 291 **concentration emission/rate of the RV Investigator, and calculated Emission Factors for PN.**

292

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

296 error. To this end, the 100 m transects were excluded from further analysis.  $\Delta$ PNC and  $\Delta$ CO<sub>2</sub> values for remaining transects  
 297 were plotted against each other as shown in Figure 5.



298

299 **Figure 5 –(a)  $\Delta$ PNC against  $\Delta$ CO<sub>2</sub> with 95% confidence interval for the six transects considered for the data analysis. (b)  $\Delta$ PNC against**  
 300  **$\Delta$ CO<sub>2</sub> with 95% confidence interval with the removal of the outlier transect from the first flight of day 2**

301

302 Figure 5 (a) and (b) show the plots of the remaining transects  $\Delta$ PNC against  $\Delta$ CO<sub>2</sub> with and without the values of the first  
 303 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  
 304 0.637 to 0.890 with its exclusion. Furthermore, whilst the linear fit falls within the confidence interval of only one point in (a),  
 305 it falls within all data points confidence intervals in (b). This occurs despite both  $R^2$  values for the fitted Gaussians of this  
 306 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  
 307 best observed in the difference between Figure 4 (a) and (b). The combination of UAV velocity, sampling rate and response  
 308 time of the DISCmini results in the PNC transect data having only one data point defining the peak height of the transect.  
 309 Relying on a single sample point leads to the potential for random instrumentation effects heavily biasing results in a way  
 310 which does not strongly impact the  $R^2$  values of Gaussian fits used to identify successful transects. Therefore, it is unclear  
 311 whether this is a variation in the ship emissions or an instrumentation error.

312 The slope and standard error of the linear fit for Figure 4 (a) was input unto Equation 1 to calculate an overall emission factor  
 313 of  $7.6 \pm 1.4 \times 10^{15} \# \cdot \text{kg}_{\text{fuel}}^{-1}$ . As presented in Table 3, this value is comparable with those reported in the literature for cruise  
 314 and cargo ship plumes; which range from  $0.2 \times 10^{16}$  to  $6.2 \times 10^{16} \# \cdot \text{kg}_{\text{fuel}}^{-1}$  (Alföldy et al., 2013;Beecken et al., 2014;Jonsson  
 315 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.,  
 316 2015)

317

Reference	Platform	EFPN (#.kg <sub>fuel</sub> <sup>-1</sup> )	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	$0.32 \times 10^{16}$	11	Harbor, Ship Channel
Alföldy et al (2013)	Land Based	$0.8 \times 10^{16}$	497	Harbor
Juwono et al. (2012)	On Board	$0.22 \times 10^{16}$	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

318

319 **Table 3 – Comparison of the Emission Factor for the RV Investigator found in this study with other relevant values found in**  
320 **literature. \* P<sub>NEF</sub> for particles above 13nm. \*\* P<sub>NEF</sub> for particles above 5nm.**

321 The calculated  $EF_{PN}$  for the Investigator was lower compared to those reported by Beecken et al. (Beecken et al., 2014) for  
322 passenger ships while accelerating ( $0.91 \pm 0.18 \times 10^{16}$  #.Kg<sub>fuel</sub><sup>-1</sup>). However, the RV Investigator measurements were undertaken  
323 whilst its engine was under 30% load. Accelerating ships will typically be under higher engine loads and hence have a  
324 correspondingly higher  $EF_{PN}$  (Westerlund et al., 2015), which explains part of this discrepancy. Furthermore, the RV  
325 Investigator has high efficiency engines and utilizes ultra-low sulphur diesel fuel. Studies have shown that similar diesel  
326 engines burning fuel of this type have lower  $EF_{PN}$  than the same engine with higher sulphur content diesel (Chu-Van et al.,  
327 2017). Similar quality fuels used in the ground transport industry have yielded similar values of  $EF_{PN}$ , ranging from  $4.8 \times 10^{14}$   
328 (25% engine load) to  $7.2$  (100% engine load)  $\times 10^{15}$  #.Kg<sub>fuel</sub><sup>-1</sup> (Jayaratne et al., 2009).

### 329 3.4. Instrumentation Limitations

330 Lightweight UAVs ~~present an opportunity~~ have the potential to achieve aerial measurements at significantly less upfront and  
331 operational costs than fixed wing and manned aerial vehicles. Lightweight UAVs can be deployed faster with limited or no  
332 required launch and landing area compared to their manned and fixed wing counterparts. Yet, their primary disadvantage,  
333 particularly in this application, is a severely limited payload weight. To overcome this limitation, this project used the  
334 lightweight and portable DICSmini and IAQ-calc sensors. However, these instruments have lower sensitivities and greater  
335 uncertainties when compared to a high accuracy CPC and CO<sub>2</sub> monitor for measurements, which can influence results.

336 The DISCmini has a manufacturer listed measurement cut-off size of 10 nm. A previous study listed in Table 3 (Lack et al.,  
337 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  
338  $EF_{PN}$  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.  
339 This may have been another contributing factor to the  $EF_{PN}$  measured in this study being in the lower end of measured values  
340 in literature.

341 The two 100m transects were not accounted for in the final calculation of  $EF_{PN}$  due to their poor Gaussian curve fits. Whilst  
342 this has been attributed to the skewing of the plume at this distance, the limitations of the instrumentation could also have  
343 contributed. The lower concentrations of CO<sub>2</sub> at this distance result in the difference above background inside the plume being  
344 the same order of magnitude as the manufacturer specified error margin. Hence, the variability in the plume either side of the  
345 central peak as shown in figure 4 (c) could be due in part to instrumentation error.

346 Calibrations of sensors in this study were performed by comparison with reference instruments for ambient measurements at  
347 sea. Ideally, calibration should be performed with in-plume measurements to have the same environmental conditions and  
348 range as the real measurements. However, it was not possible to access the plume with reference instrumentation on board  
349 the ship. Whilst this study provides a successful proof of concept with consistent results over ~~multiple-two~~ days and several  
350 flights, a validation study is needed. This should include independent measurements of EF<sub>PN</sub> using other established  
351 methodologies to ascertain more precise correction factors and uncertainties.

#### 352 4. Summary and conclusion

353 The UAV system used in this study successfully measured PN and CO<sub>2</sub> concentrations from the exhaust plume of the RV  
354 Investigator whilst operating at sea. Several different flight paths were tested and an optimal transect flying perpendicular to  
355 the plume at a distance of 20 meters from the ship was adopted. The EF<sub>PN</sub> calculated for the RV investigator was  $7.6 \pm 1.4 \times$   
356  $10^{15} \text{ \#} \cdot \text{kg}_{\text{fuel}}^{-1}$  at a constant 30% engine load. This EF<sub>PN</sub> was in agreement with values reported in literature, indicating this  
357 ~~novel~~ UAV based system has potential for EF<sub>PN</sub> quantification pending further evaluation.

358 In comparison with other methods, ~~the UAV systems~~ have the potential to provide ~~presented provides~~ a cost effective and  
359 accessible solutions for the rapid measurement and quantification of ship EF<sub>PN</sub>s. Its ability for deployment both in harbour and  
360 at sea, coupled with the possibility of altering its flight path to account for variances in wind conditions; gives this UAV system  
361 a ~~distinct advantage over ground based and manned aerial vehicles~~ high degree of flexibility. Furthermore, the UAV can sample  
362 considerably closer to the plume emission source than other methodologies, providing higher concentration measurements for  
363 the calculation of EF<sub>PN</sub>.

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

#### 367 4.1. Recommendations

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

375 Further optimization of the transect approach is ~~also possible~~ necessary in order to conduct UAV measurements of shipping  
376 emissions on a larger scale. After location of the plume the system could be set to make several repeat passes across the plume  
377 in rapid succession to increase the sample size. Another alternative would involve the UAV hovering inside the plume over a  
378 period of time collecting a continuous series of measurements from the centre of the plume. These methods would both require  
379 real time sensor feedback to the UAV pilot and potentially adaptive autonomous controls to achieve a suitable result. Further  
380 ~~This methodology could also be expanded to measure other important ship emission factors, including NO<sub>x</sub> and volatile~~  
381 ~~organic compounds (VOCs).~~ challenges include: operation under less favourable weather conditions; measurements in which  
382 the UAV is not launched from the ship itself; and measurements taken for ships moving at full speed. This methodology could  
383 also be expanded to measure other important ship emission factors, including NO<sub>x</sub> and volatile organic compounds (VOCs).

## 384 Acknowledgements

385 The authors would like to acknowledge the ARCAA Operations Team (Dirk Lessner, Gavin Broadbent) who operated the  
386 Unmanned Aerial Vehicle (S800). This research was supported by the Australian Research Council Discovery Grant  
387 DP150101649 and the Marine National Facility. The authors would like to thank the Captain and the crew of the RV  
388 Investigator as well as the on board MNF support staff as without their support and effort this research would not have been  
389 possible.

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