General considerations

It looks like some major effort has been done to make this article acceptable for publication, but still some major issues remain. I did not have the impression that all changes were included in the last version (version 4), although they have been mentioned to be solved in the author responses. I would request a thorough revision before providing the manuscript again to the reviewers.

Analysis of the responses of the author

Comment 1 from reviewer 1: Line 22 : Typo in first emission factor is still not corrected

Answer 1: **Emission factor value has been corrected in Line 22**

Response to the answers from reviewer 3

Comment 1: The authors do not mention some highly relevant projects, studies and operations that have been executed, or are ongoing in Europe whether or not with UAV systems on the subject of airborne and remote ship emission monitoring. Although the study has some interesting and innovative aspects, the use of UAV systems for emission monitoring is not new and should not be resented as such.

Answer 1: The authors considered the Reviewer comment, yet the emphasis was intended to be on the fact that EFPN of ships has never been evaluated with UAVs. The updated manuscript has been modified in multiple lines to clarify this.

Response1: The manuscript is still not clear about the focus on EFPN in several places still "ship emissions" in the broader sense are mentioned without emphasising on EFPN (e.g. in the abstract line 23 and in the conclusion line 336)

Response Answer 1: Line 24 and line 333 have been clarified.

Comment 2: Line 23: The authors indicate that emissions were assessed during real world conditions. This is not assessed as such as all measurements were performed from and for one ship. Besides the measured RV is a relatively small vessel (94m) while average merchant vessels are in the order of 200-400m. The RV was also running on ultra low sulphur marine diesel fuel while in reality only a fraction of the international merchant vessels use this fuel type. Different factors may influence the successful assessment of ship emissions among others are: ship-type, ship-age, shipsize, ship-shape, shipactivity, fuel-type, funnel height, funnel shape, wind conditions, inversion layers, etc For a realistic assessment during real world conditions these factors should have been elaborated. Furthermore for this study the flight path was based on the ship position, in real life ship position is not known in detail, AIS only provides basic navigation info e.g. there is no information on the location and shape of the funnel on the ship. The limited autonomy, range and payload of the UAV make this UAV not suitable for realistic operational measurements at sea during real world conditions, the study can therefore hardly be used as a proof of concept. For actual (cost) effective operations offshore, much more robust fixed- or rotary-wing UAV systems should be used, these systems have other specifications (speed, manoeuvrability etc.) than the one used in this study.

Answer 2: The phrase “real world conditions” is intended to indicate that rather than in a lab or simulated conditions, the UAV was launched on a ship performing operations at sea and measured the exhaust plume. The focus of this paper is on a proof on concept of the methodology. It is not a proof of concept for widespread deployment of this methodology in the field for regulatory or commercial use. That is far beyond the scope of this manuscript. The authors disagree that ship type, class, fuel type, and other differing factors would prevent this methodology from being used. Provided there is an exhaust plume which can be intercepted by the UAV, this methodology can be used to assess emission factors of PNC. The wording of the paper has been changed in multiple places to highlight this as a proof of concept.

Response 2: An explanations on “the real world conditions” and “proof of concept” should be included in the text, as the text is not clear on this phrasing. The reviewer agrees that as a proof of concept this method is promising, but stating that this method “provides a reliable inexpensive and accessible way to asses and potentially regulate ship emissions” as mentioned in line 23 is a premature conclusion (see Comment 3)

Response Answer 2: The authors believe the meaning of “real world conditions” and “proof of concept” are self-evident given the context provided in the paper. The word ‘reliable’ has been removed from line 24. The authors maintain that the methodology described is inexpensive and accessible in comparison to other methodologies currently available.

Comment 3: Line 24: The authors indicate that for the first time ship emissions can be assessed and regulated on a reliable and inexpensive way. This is incorrect, as emissions from ships are already assessed and regulated from both airborne, land based and shipborne sensors in Belgium, The Netherlands, Denmark, Germany and Finland since 2015 at a large scale and on a reliable and cost efficient manner. The use of the UAV’s is not necessarily more cost-effective, especially if operated from a ship, and often more time-consuming with less operational output capacity per flight hour. Clearly more information is required to establish cost-effectiveness (platform cost, number of ship measurements per hour, personnel involved, robustness of platform in offshore conditions, ...) . Furthermore the use of UAV's for emission monitoring operations is not new, in 2016 EMSA ordered a feasibility study, granted to CLS, concerning the use of RPAS for emission monitoring (STEAM project), in addition the Danish company EXPLICIT performed some successful emission measurements with small drones. The only aspect which might be innovative in this study is the measurement of PM emissions from ships using drones, but as this is not yet regulated by international law, this has (currently) only academic use.

Answer 3: The novel aspect of this paper is the measurement of particle number (PN) emission factors using a relatively inexpensive UAV. This is primarily for academic purposes. However, PN has been identified as critical to both health and climate and thus developing the basis for tools which may suggest that potential regulatory of PN emissions is important.

Response 3: The abstract in version 4 of the manuscript was not changed on this matter, the reviewer request rephrasing the abstract with emphasis on the novel aspects of PM monitoring using UAV.

-UAV's have and are being used for ship emission measurements, so no claim can be made that this is done for the first time.

-Accessibility can hardly be claimed as all operations were performed from an RV to measure the RV itself (it would have been possible if test were conducted from shore to measure ships inbound of ports)

-Reliability has to be assessed on a much wider sample size, this was not assed, neither mentioned in this proof of concept, so no conclusion on reliability can be drawn on reliability.

-No cost efficiency analysis has been made to use this proof of concept for ship emission monitoring (others than the ship it is operating from), therefore it is premature to call this method inexpensive

-The author mentions "potentially regulate", as no regulation is currently in place for PM from ships, it is premature to mention this

Response Answer 3:

- This is the first time a UAV has been used for EF_{PN} measurements, this has been clarified as discussed previously.

- The point of departure and landing of the UAV bears no impact on the methodology described provided that the ship is in flight range. A land base or small boat could be easily used, hence accessibility. The authors feel that this has been made clear in the paper.
- The authors agree, reliability has been removed from line 24
- The upfront cost of this setup is orders of magnitude less than a fixed wing aircraft. Operation of the UAV requires no fuel and a pilot with relatively minimal training in comparison to a fixed wing aircraft. The authors feel that this warrants the use of the term "inexpensive" and that a cost efficiency analysis is irrelevant to the focus of this manuscript.
- The authors agree and have removed the points on regulation from the manuscript.

Comment 4: Line 64: The authors make the assumption that manned aircraft are not feasible for airborne measurements of ship emissions, although the EU funded CompMon project clearly showed the feasibility of manned aircraft for operational emission regulatory airborne surveillance (e.g. operations in Belgium with >2500 monitored ships in 3 years and operations in Denmark with >1000 monitored ships in 2 years).

Answer 4: The UAV-based methodology detailed in this manuscript offers an operational setup with orders of magnitude less upfront and operational costs than manned aircraft. The project listed is of a far larger scale and budget than typical research projects.

Response 4: Line 69 still mentions the limited feasibility of manned aircraft for ship emission monitoring. Note that in Belgium more than 1000 ships per year are monitored for FSC (via measurement of SO2 and CO2). So claiming that manned aircraft are not feasible is not correct. Deploying drones from vessels to inspect the same amount of ships in the same area (in shipping lanes at ca. 60 km from shore) would have been possible, but at significant higher cost. The Belgian aircraft is able to provide a measurement for less than 200€ per inspection, navigating a ship to deploy a drone will cost a multitude of this amount. Drones have a commentary value in for instance the monitoring of inland waters, ports, or when shipping lanes cross near shore, this nuance was not made, simply stating that aircraft are expensive compared to drones is not correct as this requires a description of the operational framework and the application. Furthermore, large scale drones with sufficient radius require similar budgets than manned aircraft and face similar limitations in matter of flying restrictions.

The authors mention risks as well as a reason why manned aircraft have limited feasibility, like for all flying operations risks are involved and have to be assessed in a risk analysis, therefore the risks have to be considered indeed, but on the other hand do not necessarily limit the feasibility.

Response Answer 4:

The aircraft discussed measures SO_2 and CO_2 which are gaseous pollutants not relevant to the EFPNs which are the focus of this methodology. It is likely this aircraft can only be operated viably at such a cost due to the high demand of measuring these pollutants for regulatory applications. Measurement of PN emissions for research applications are in much lower demand, hence why there are no dedicated manned aircraft available for these measurements at such a cost. This aside, the reviewer has indicated several scenarios in which manned vehicles are not viable, supporting the authors claim that they have limited feasibility.

Comment 5: Line 142: Sensitivity range for CO_2 is 50ppm, this is important as this is same order of magnitude as the delta CO_2 for measurements at 100m, this aspect should be discussed further in the article in an overall assessment of the margin of error, which is currently missing.

Answer 5: The updated manuscript addresses instrumentation sensitivities and error margins. In particular this comment has been discussed in lines 313-317.

Response 5: The discussion in the text from line 324 to 328 now sufficiently describes this issue

Comment 6: Line146: Significantly more detailed information should be provided on the calibration method (references samples, calibration-factors, offset, : : :). It is also not clear if a calibration was performed before (and after) every measuring day, this should have been done to ensure the validity of the data. Line 147 (Figure S1): More information is required for the comparison of the CPC with the DISC, it is not clear what kind of air samples were used for the comparison, it looks like this is just done based on continuous ambient air measurements on board of the RV, for a proper validation a comparison should be made with real emissions. A comparison of the IAQ with the PICARO is completely missing here. If only a comparison (validation) is possible in a lab, this comparison should at least be done during similar conditions as during the field measurement (exposure time, concentration, temperature,), this is clearly not the case as the particle concentrations is very low in this comparison. It looks like the intercept of the linear regression is not put at zero, why is this, was a zero calibration performed? Especially for CO_2 it is important to perform the calibration in the same range as the measurement range as the IR absorption is nonlinear, no comments were made on this aspect in the article. Furthermore it should be noted that a linear regression is not an ideal method to compare 2 sensors, the Bland Allman method is more appropriate (Statistical Methods for Assessing Agreement Between Two Methods of Clinical Measurement," by JM Bland and DG Altman, The Lancet, February 8, 1986, 307310).

Answer 6: Methodology has been expanded upon significantly in the updated manuscript and a C02 picaro comparison is provided in supplementary material.

Response 6: Figure S2 does not use a linear regression like for the comparison between the PM sensors. An explanation is missing why a different comparison method was used.

Line 151 in the CO2 section refers to Figure S1 instead of figure S2.

Figure S2 shows the extensive variation and measurement difference between the Picaro (393ppm) and the IAQ-Calc (486.5ppm), this issue should have been elaborated in the text (see Comment 6). Please explain why the authors did not choose to conduct a calibration with a set of reference span gasses (including zero gas) as this is the most common way to calibration gas analysing sensors?

The sensor comparison in Figure S1 uses a linear regression (that intercepts at 0) therefore a calibration can be done of the DISC data using the regression coefficient as the main calibration factor, for the IAQ only an offset is used (93) and no calibration factor, please explain why this different approach was used.

Response Answer 6

Line 149 has been corrected.

The calibrations of the instrumentation were performed using ambient aerosol measurements. This has been clarified in line 148. The PN concentrations show a linear response trend which allowed for a good fit. As is shown in S2 no such trend was observed for $C0_2$, instead an offset error were calculated through the mean value indicated as shown in the supplementary material.

Comment 7: Line 158: Flight speed is here expressed as 1.5m/s, it is not clear if this is the airspeed or ground speed. If this is the airspeed, the actual ground speed will depend on the wind conditions, therefore the flight speed through the plume is dependent on the wind conditions too. During the first day, the wind was cross on the ship heading. The plume would be expected at 180° if transect were flown with alternating heading 250° and 70° (perpendicular to the ship heading), the transect with heading 250° would have been flown with a significant different ground speed (ca. 6.5 m/s instead of 1.5 m/s), no mention is made of this in the article.

Answer 7: Flight speed listed is the airspeed. Whilst the wind conditions will influence the ground speed, the only influence on the measurements will be a variation in the amount of data points captured inside the plume during transect. The discussion of the amount of in plume data points in a transect and its importance is in the updated manuscript in lines 277-281.

Response 7: Wind conditions will have an impact in the usability of this method as they impact the number of data points but also the concentrations in the plume (dilution) and therefore depending on the wind condition the measurement could fall outside sensor sensitivity. Furthermore the transect and flight path have to be adjusted depending on the wind conditions, eg. in case of cross wind the measurements behind the ship would not make any sense.

Response Answer 7: The impact of wind direction and ambient condition changes are addressed in Paragraph 252-260. As stated in Line 158 the RV Investigator was heading into the wind during measurements, therefore the flight path was discussed in terms of position from rear of ship to ensure clear discussion. The authors believe it is self-explanatory that the flight path would need to be changed if the ship was not oriented into the wind.

Comment 8: Line 208: I would suggest adding an indication of the resulting plume location and flight pattern on the graphs. These graphs would also visualise the different airspeed between the transects (see comment line 158).

Answer 8: The emphasis in the graphs is on the clear detection of the plume by each instrument. The authors do not believe that plume locations would not provide any further information and would overcomplicate the graphs.

Response 8: Plotting the estimated location of the smoke plume could visualise the issue that was explained in response 7.

Response Answer 8:

The authors believe this issue to be sufficiently discussed and maintain that further information would overcomplicate the graphs.

Comment 9: Line220: Only 9 times the plume was sampled, very few statistical conclusions can be made based on this small sample size, especially the linear regression on line 277 is questionable.

Answer 9: The methodology has been updated in the updated manuscript.

Response 9: The reviewer argues that the limited number of successful measurements was not sufficiently considered in the manuscript (see comment 3).

Response Answer 9:

Despite the number of samples being relatively small, there are two aspects that support our assertion that the result is significant and reliable. First, we note that the best line in the graph passes through the error margins of every one of the viable data points collected, suggesting that there is a linear relationship. Secondly, the value of the emission factor calculated using the linear regression is well within the range of values found in a number of other published studies and listed in Table 3. The paper is transparent about the size of the data set used.

Comment 10: Line 229: The distance (25m) is missing in this sentence.

Answer 10: This has been clarified in the updated manuscript.

Response 10: This is sufficiently clarified by the authors

Comment 11: Line 232: It is mentioned that the CO_2 is up to 100 ppm higher in the plume, this is not clear on the graph (only 50-75 ppm), this will be the part for integration to amount to the delta CO_2. Furthermore it should be noted that the peaks for CO_2 at a distance of 100 m is of the same order of magnitude of the sensor accuracy.

Answer 11: The C02 is up to 144ppm counts above background inside the plume in graph 4(a). The graph has been replotted with background removed in the updated manuscript to clarify this. The short 100m transect data has also been discussed in more detail.

Response 11: This is sufficiently clarified by the authors

Comment 12: Line 262: Another flight transect could have been used where the UAV would be flown at the same speed and heading as the RV and hovered in the plume, this would require a transmission of measurement info to the control station to adjust flight altitude and pattern to successfully find the plume and measure the plume for longer periods.

Answer 12: The focus of this project was the measurement of EFPN through transects of the ship plume. Due to time constraints alternative methodologies could not be investigated, though this suggestion is one of the recommendations for further research listed in the manuscript.

Response 12: This is sufficiently clarified by the authors

Comment 13: Line280: Instead of a comparison between calculated emission factors and the emission factors from previous studies a comparison with the emission factors calculated based on a plume measurement with the other equipment on board of the RV (e.g. Picaro) would have made more sense.

Answer 13: There was no possibility of accessing the plume with the larger instrumentation such as the picaro or CPC. This is one of the primary advantages of UAV-based platforms. A future validation study would look into this. This is a recommendation in the updated manuscript.

Response 13: This is sufficiently clarified by the authors

Comment 14: Line 312: Generalization and misconception that the use of UAV systems would consist of a reduced cost. It is definitely not presented in this article that UAV systems could provide a real cost effective alternative to other surveillance methods as no cost benefit comparison was made between different surveillance methods (both fixed stations and airborne sensors; operational output capacity; personnel and supporting platform etc.) and all missions were carried out from a vessel, which has a higher operational cost per hour as an aircraft and a lower speed and therefore a much lower cost efficiency. Note that a higher cost efficiency could maybe be acquired with this setup where these operations would be combined with other task carried out by patrol vessels, pilot ships or research vessels assuming that these vessels would operate within 2 km of shipping lanes. This was not mentioned in the article.

Answer 14: The authors have addressed this concern in Answer 4, the setup and operational costs of this UAV system are orders of magnitude less than manned aircraft. The focus of this manuscript was on the development of the methodology. Whilst some suggestions for future applications are made, it is premature and beyond the scope of this paper to recommend wide-scale deployments of UAVs and cost benefit comparisons with other methodologies.

Response 14: The reviewer does not want to argue that this cost efficiency comparison should have been made, but claiming that UAV's are less expensive and more effective than other methods also requires a description off the operational framework. The reviewer agrees that for the concept of this research a UAV was most likely less expensive than an aircraft as the RV was monitored during another campaign. Concerning the proximity to the plume, note that the Danish company Explicit is conducting helicopter measurements and is also involved in the development of sensors for drones, in their operational procedures drones fly further away from the ships than the helicopter. The text from line 335 to 339 should be changed as this is based on unfounded assumptions that do not reflect the reality of airborne measurements.

Response Answer 14: The authors feel that their claims to the relative inexpensiveness of this system have been explained and are justified. In regards to the comparison between this methodology for the measurement of EF_{PN} for research applications and the commercial measurement of regulated pollutants by private companies, please consider the response given in Answer 4.

Comment 15: Line 326: SO2 is completely missing here, SO2 is the only emission regulation which is effectively monitored using airborne platforms at this moment and should therefore at least be included in the discussion.

Answer 15: The focus of this study was on PN emissions. SO2 would be an interesting alternate application. To the authors knowledge the main challenge for such a system would be that fast and accurate SO2 meters are significantly above the payload of any lightweight UAV, include fixed wings.

Response 15: The sensor-system from Explicit is able to measure SO2, CO2 and NO. This sensor system is in the order of magnitude of a few kg and is deployed on drones and helicopters (slow time response requires monitoring in smoke plumes for up to 30 sec). The sensor used by the Belgian coastguard measures SO2 and CO2, weights 40 kg and has conducted more than 3000 measurements, this sensor uses a Thermo 43I TLE that has been specially modified for faster time response (1-2 sec)

Response Answer 15: While there are a number of pollutants that could have been measured, this study was focussed on particle number concentration, as this is a parameter that has not been monitored in ships plumes using UAVs in the past. Aside from $S0_2$ sensors not being relevant to the calculations of EF_{PN}, again we argue that the main challenge for such a system would be that fast and accurate SO_2 meters would be significantly above the payload of any lightweight UAV.

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

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Abstract. This research demonstrates the use of an unmanned aerial vehicle (UAV) to characterize the gaseous (CO_2) and 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 significant source of climatically relevant aerosol particles" on-board the RV Investigator around the Australian Great Barrier Reef. Measurements of the RV Investigator exhaust plume were carried out while the ship was operating at sea, at a steady engine load of 30%.

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

Particle number emission factors (EF_{PN}) were calculated in terms of number of particles emitted (#) per weight of fuel consumed (Kg fuel). Fuel consumption was calculated using the simultaneous measurements of plume CO_2 concentration.

The calculated EF_{PN} was 7.6 ± 1.4 x 10^{15} #.Kg_{fuel}^{-1}, which is in line with those reported in the literature for ship emissions ranging from 0.2 x 10^{16} #.Kg_{fuel}^{-1} to 6.2 x 10^{16} #.Kg_{fuel}^{-1}.

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

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1. Introduction

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

Despite these findings, emissions from shipping have consistently been subject to less regulation than those of land-based transport with ship emissions in international waters remaining one of the least regulated parts of the global transportation system (Cooper, 2001, 2005;Corbett and Koehler, 2003;Corbett and Farrell, 2002;Eyring et al., 2005;Streets et al., 1997;USEPA-OTAC, 2012). Currently, no specific restrictions for ship-emitted particulate matter (PM) exist, with the only regulated pollutants being NOx and SO_2. The International Maritime Organization (IMO) recently revised the regulation of these gaseous pollutants through the Annex VI of the International Convention for the Prevention of Pollution from Ships – the Marine Pollution Convention (MARPOL). The IMO expected that these regulations would lead to an indirect decrease in particle number (PN) concentration due to the reduction of NOx emissions and the use of fuel with lower sulphur content [14]. However, it has been found that the use of some low sulphur fuels lead to increased PN concentrations at lower engine loads (Anderson et al., 2015), which stresses the importance for regulation specifically addressing particulate matter (PM).

The majority of emitted PM is in the ultrafine size range, < 0.1 µm, which have been demonstrated to have a particularly significant impact on health and the environment (WHO, 2013). However, due to the lack in regulation, ultrafine particles, in terms of PN concentration, emitted from ships have remained unassessed in real world conditions. Quantifying PN concentration is critical to improve our understanding of shipping's impact on health and climate (Chen et al., 2005;Cooper, 2001;Corbett and Farrell, 2002;Isakson et al., 2001;Williams et al., 2009;Reda et al., 2015;Mueller et al., 2015;Anderson et al., 2015;Blasco et al., 2014;Ristovski et al., 2012;Corbett et al., 2007b). To achieve this, wide-scale evaluation of ship emission factors (EFs) is necessary. EFs are commonly expressed as the amount of pollutant (x) emitted per unit mass of fuel consumed g(x). $(\text{Kg fuel})^{-1}$. Different methods have been used to investigate ship EFs, including laboratory test-bench studies, on-board measurements, and measurement of ship emission plumes.

Test-bench studies (Reda et al., 2015;Mueller et al., 2015;Anderson et al., 2015;Petzold et al., 2010;Petzold et al., 2008;Kasper et al., 2007) have been used to characterize emissions from different engines at various loads in laboratory conditions. However, engine performance and emissions have been shown to be different in real world operations when compared to laboratory studies. This calls for measurements of ship emissions in-situ to collect reliable data for EF calculations (Blasco et al., 2014;Murphy et al., 2009;Agrawal et al., 2008). To date, only a few studies have been undertaken on-board ships to calculate real emission factors (Juwono et al., 2013;Hallquist et al., 2013b). This is attributed to the prohibitive costs and time commitments of setting up and maintaining on-board measurement equipment on commercial ships. Airborne ship plume measurements (Westerlund et al., 2015;Schreier et al., 2015;Pirjola et al., 2014;Cappa et al., 2014;Beecken et al., 2014;Balzani Lööv et al., 2014;Berg et al., 2012;Lack et al., 2009;Lack et al., 2008;Sinha et al., 2003) offer an alternative method of in-situ measurements without requiring on-board monitoring stations. In the past the deployment cost of these systems, and the risks associated with manned aircrafts have limited their feasibility. However, this has recently changed with the rapid advances being made in commercially available Unmanned Aerial Vehicle (UAV) technology.

Hexacopter UAVs have seen a wide scale increase in industry and research applications due to their ease of use and comparatively low cost (Malaver Rojas et al., 2015;Gonzalez et al., 2011;Brady et al., 2016). Used in conjunction with air monitoring equipment, these systems provide, for the first time, the ability to perform relatively simplistic and cost-effective airborne measurements of ship emissions. However, to date no studies have reported the use of a UAV system capable of collecting data to calculate the EF of PN concentration for ships at sea.

This research utilized a customized hexacopter UAV carrying instruments for PN concentration and CO_2 measurements to derive EF_{PN}. The UAV system was deployed from the RV Investigator research vessel while at sea. Autonomous measurements of the RV investigators exhaust plume were taken over several flights at various altitudes and distances from the ship. Data collected was used to optimize the sampling flight path and successfully quantify the RV investigators EF for PN concentration.

2. Methodology and Measurement system

Measurements were conducted as part of the research voyage "The Great Barrier Reef as a significant source of climatically relevant aerosol particles" aboard the RV Investigator research vessel over a two day period of the 13 and 14 October 2016 (day 1 and day 2). Measurements of PN and CO_2 concentration emitted by the RV Investigator were taken using a PN and CO_2 monitor mounted on a customized DJI EVO S800 hexacopter UAV (DJI, 2014).

2.1. The RV Investigator and the voyage

The RV Investigator is an ocean research vessel configured to enable a wide range of atmospheric, biological, goescience and 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 sulphur diesel fuel. It is powered by three 9 cylinder 3000 kW MaK diesel engines, each coupled to a 690V AC Generator. Ship propulsion is achieved using two 2600 kW L3 AC reversible propulsion motors powered by these generators. The RV Investigator can host up to 30 crew members and 35 researchers for a maximum voyage period of 60 days with a maximum cruising speed of 12 knots.

A suite of instrumentation for atmospheric research is available on the RV Investigator. This includes a radar system capable of collecting weather information within a 150 km radius of the vessel, and instruments measuring: sunlight parameters; aerosol composition, particle concentration and size distributions; cloud condensation nuclei; gas concentrations; and various other components of the atmosphere. These instruments are housed inside two dedicated on-board laboratories for aerosol and for atmospheric chemistry research. An atmospheric aerosol sample is continuously drawn into the laboratories for analysis through a specialized inlet fitted to the foremast of the ship. Of particular interest to this study, the ship contains a PICARRO (PICARRO Inc., Santa Clara, California, USA) G2401 analyser (Inc., 2017) that continuously measures CO_2, CO, H_2O and CH_4. It has an operation range between 0-1000 ppm and a parts-per-billion sensitivity (ppb) for CO_2.

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

2.2. UAV system

Measurements of PN and CO_2 concentrations in the ship plume were performed using two commercial sensors mounted on-board a hexacopter UAV. The UAV used (Figure 1) is a composite material S800 EVO manufactured by DJI (DJI, 2014). The UAV is 800 mm wide and 320 mm in height, with an unloaded weight of 3.7 kg. Minimum and maximum take-off weights are 6.7 kg and 8 kg, respectively. The UAV contains a 16000 mAh LiPo 6 cell battery, which provides a hover time of approximately 20 min when operating at minimum take-off weight. The telemetry range of the UAV is 2 km, which was adequate to cover the desired sampling area (See Figure 2).

The payload consisted of a PN concentration and a CO_2 monitor mounted on-board underneath the UAV. Careful placement of the payload was required to prevent flight issues caused by an altered centre of gravity. Also included was a carbon fibre rod, which extended outward horizontally from the UAV. The sampling lines for the monitors were attached to the end of this rod to ensure that measurements were not affected by the downwash of the UAV rotors. The total weight of the payload 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).

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

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. This reduces operational costs and avoid subjection to the tighter regulations of larger platforms. Small UAV cannot be operated above any person, or closer than 30 m of populated areas, houses and people. Furthermore, current Civil Aviation Safety Australia (CASA) regulations restrict the use of small UAV (2 and 20 kg) to visual line-of-sight daylight operation, with a maximum altitude of approximately 120 m and within a radius of 3 nmi of an airport. UAVs in this category are not permitted for research unless the research institution has been granted a permit exception. These exceptions can be granted if the institution in question has or collaborates with an UAV operation team who must have: an experienced UAV pilot who is also radio controller specialist; a license for commercial UAV operation; and appropriate liability insurance (NPRM 1309OS - Remotely Piloted Aircraft Systems). Queensland University of Technology (QUT) has an unmanned operator certificate and four pilots who have UAV controller licenses.

2.2.1. Instrumentation

2.2.1.1. Instrumentation for PN concentration

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

2.2.1.2. Instrumentation for CO2 concentration measurements

A TSI (TSI, Shoreview, Minnesota, United States) IAQ-calc 7545 model was chosen to measure CO_2 concentrations. Its sensor is based on a dual-wavelength NDIR (non-dispersive infrared) with a sensitivity range between 0 to 5,000 ppm and an accuracy of ± 3.0% of reading or ± 50 ppm (whichever is greater). The measurement resolution is 1 ppm with a maximum time resolution 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 (270 g, with batteries, significantly lower than the DISCmini), and a battery life of 10 hours.

The readings of the IAQ-clac for CO_2 were compared with those measured by the on-board PICARRO G2401 analyser.

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

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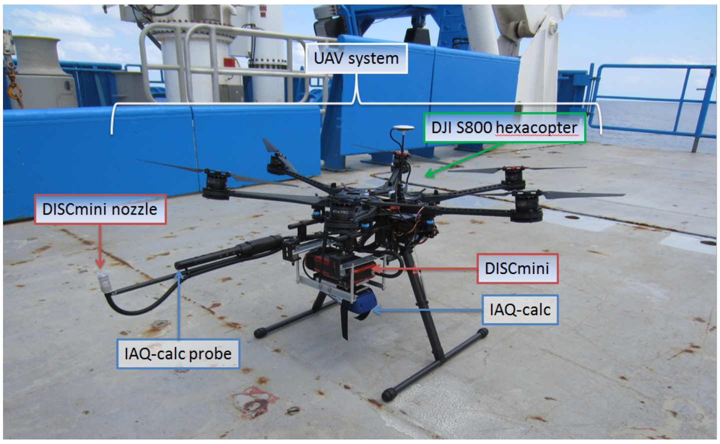

Figure 1. The UAV system with the on-board instrumentation: the DISCmini and the IAQ-calc.

2.3. Meteorological data

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

2.4. Study design

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

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

The optimised flight path for day 2 started 20 m behind the ship and 25 m above the surface, with no altitude variation. The 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 allowed up to 4 horizontal transects to capture the ship plume.

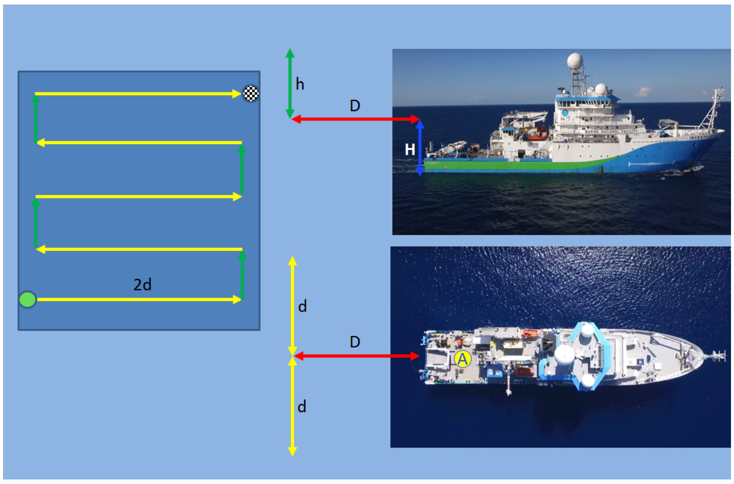

Figure 2. Flight path used to capture the plume: H - height from the ocean, D – distance behind the ship to the flight beginning point, h – rising altitude after the horizontal transect, 2d – full length of the horizontal transect

2.5. Experimental procedure

The UAV can fly either manually or autonomously. As a safety precaution, every take-off and landing was performed using the manual flight mode. Once in the air, the UAV was switched to autonomous flight mode, allowing the platform to follow the pre-programmed flight path discussed in the previous section. The flight path consisted of waypoints, which are threedimensional GPS points that dictate the position of the UAV along the fight path. The waypoints and flight plans for each flight were programmed using the aforementioned DJI Wookong ground station software. The DISCmini and the IAQ-calc were fitted on the underside of the UAV at the beginning of each measuring day. Five flights were performed across the two measurement days, providing a total of 27 horizontal transects perpendicular to the ship's exhaust plume.

2.6. Emission factors

The calculation of an emission factor for particle number concentration (EF_{PN}) from the collected ship plume measurements was performed using Eq. (1). This method has previously been used for ship (Westerlund et al., 2015), road vehicle (Hak et al., 2009) and aircraft (Mazaheri et al., 2009) emissions. The measured values of PN concentration were related to the amount of fuel consumed by the engine in question through the use of the simultaneous measurements of CO_2 concentration taken by the UAV. This was achieved by using a published value for a ship emission factor of CO_2 (EF_{gas}) of 3.2 Kg CO_2 (Kg fuel)$^{-1}$ (Hallquist et al., 2013b;Hobbs et al., 2000) .

Eq.(1).

$$EF_{PN} = \frac{\Delta PN}{\Delta gas} \text{ x } EF_{gas} \quad (1)$$

The ΔPN and Δgas in Eq. (1) represent the maximum particle concentration change above background in the measured particle number and CO_2 concentrations, respectively. The DISCmini measurements were corrected against a reference CPC. For each transect data series of PNC and CO_2, the averaged background concentration were subtracted from the peak data corresponding to measurements inside the plume. The corrected peak data series were then fit with a Gaussian curve using the inbuilt Matlab curve fitting application. The least absolute residuals (LAR) condition was used as this most closely fits the curve to the highest magnitude data points in the series. The maximum peak height of the fitted Gaussian curves were used as ΔPNC and ΔCO_2 in the calculation of emission factors for each transect.

3. Results and Discussion

3.1. Meteorological and Investigator data

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

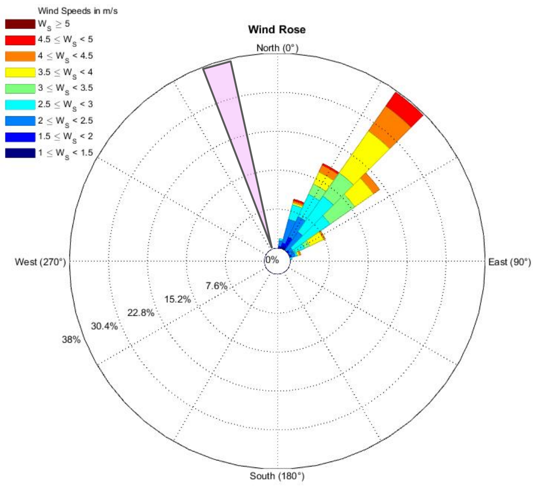

Figure 3a – Wind rose showing wind speed and direction during day 1. Rose triangle shows RV Investigator direction during the measurements.

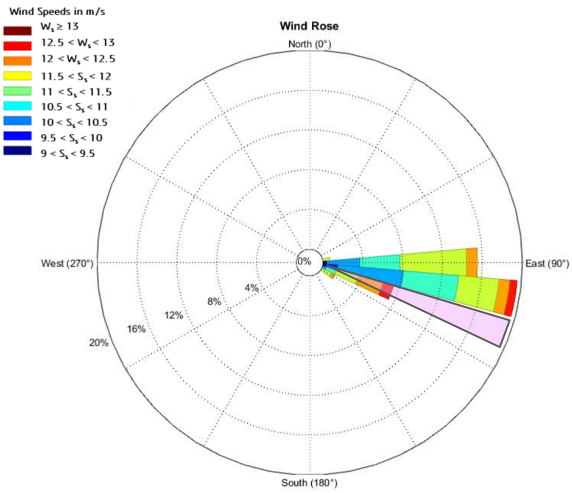

Figure 3b – Wind rose showing wind speed and direction during day 2 optimized flight. Rose triangle shows RV Investigator direction during the measurements.

3.2. UAV system horizontal transects inside and outside the plume

The UAV system acquired data for a total of 27 horizontal transects for day 1 and day 2. Data were collected at altitudes 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 altitude and 20 m downwind of the ship; and again at both 25 and 35 m altitude 100 m downwind of the ship. These observations lead to the optimized flight used on day 2, which started downwind at 25 m above the surface and 20 m behind the ship. On day 2 the UAV system successfully captured the plume during 6 of the 8 transects performed. Across the two days this lead to a total of 9 transects that captured the plume and which have been considered for discussion, shown in Table 1.

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**

Table 1 – Specifications of the transects considered for the data analysis. The (*) indicates the transect of Day 1 of which PN concentration and CO_2 profiles are presented in Figure 4.

Figure 4 shows the PN concentration and CO_2 profiles, collected during two (a; b) transects on day 2, and (c) during one transect of day 1 (Spec. in Table 1, Day1*).

The PN concentration profiles for the (a) and (b) transects in Figure 4 show that the concentration varied by five orders of magnitude between the outside and inside the plume, while the CO_2 profiles show an increase up to 140 ppm above the background.

The profiles in (c) show that the PN concentration was four orders of magnitude greater inside the plume at 100 m behind the ship and that the CO_2 concentration was up to 70 ppm higher inside the plume.

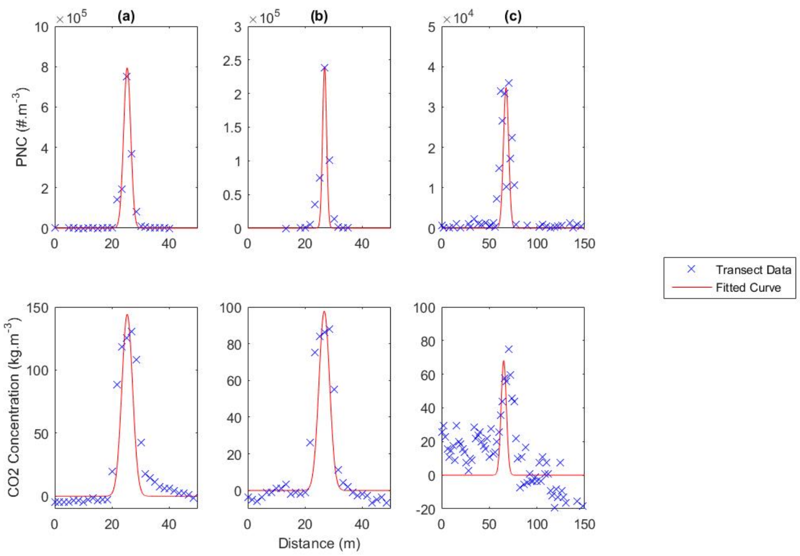

Figure 4 – (a) and (b) show the measured PN and CO_2 concentration profiles and fitted Gaussian curves for two different transects 20 m behind the ship 25 m above the surface during day 2. (c) shows the PN and CO_2 concentration profiles and fitted Gaussian curves collected during flight 3 of day 1 at 100 m behind the ship, 25 m above the surface.

Figure 4 (a) and (b) both show transects at 25 m altitude and 20 m behind the ship. Both the PN concentration and CO_2 measurements show clear, single peaks as the UAV crosses the plume. As a consequence, these transects show a good fit with the corresponding Gaussian distribution curves with R^2 values of above 0.9 for both PNC and CO_2. In contrast Figure 4 (c) shows substantially less defined, wider peaks with lower pollutant concentrations. This is attributed to a difference in flight paths, with Figure 4 (c) representing data from a transect 100 m behind the ship. The additional time between emission and sampling has allowed the plume to broaden, become less homogenous, and take on a skewed cross-section. This results in a significantly lower R^2 value for the fitted Gaussian curves, with a value of 0.4998 for the CO_2 data in this transect. Therefore, whilst the 100 m transect does provide more data points inside the plume, the randomized variations inside the plume lead to less accurate calculations of emission factors.

Of further note in Figure 4, the maximum PN concentrations measured in (a) (7.5×10^5 #.cm^{-3}) is approximately three times greater than those in (b) (2.4×10^5 #.cm^{-3}) and the CO_2 concentrations in (a) are 43 ppm greater than (b). The transect flight plan and ship engine load remained constant throughout these measurements. The variations between (a) and (b) are attributed to several factors which reduce the effectiveness of the UAV transect for capturing the plume. Slight changes in ambient conditions such as temperature, wind direction and intensity will alter the path of the plume as it moves away from the ship. The UAVs automated flight path cannot account for these variations. Therefore, the degree to which the UAV enters the plume, and thus the concentrations it measures, will be different on each transect. Both CO_2 and PN concentration measurements will be similarly affected by this variance. However, differences in instrument response rates in conjunction with these variances will be one of the major contributors to variations in calculated emission factors.

3.3. PN Emission Factors

Table 2 shows the distance and altitude of each transect, the R^2 values of the fitted Gaussian curves for PNC and CO_2 data, the 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

Table 2 – Transect flight days and details, R^2 values for the Gaussian curve fits to both PNC and CO_2 data, ΔPNC and ΔCO_2 concentration emission/rate of the RV Investigator, and calculated Emission Factors for PN.

The calculated EF_{PN} values for the RV Investigator ranged from 1.15×10^{15} to 1.73×10^{16} #.Kg$_{fuel}^{-1}$. The two 100 m transects provided the worst Gaussian fits as well as the highest and lowest calculated emission factors. This indicates that it is important to filter out transects with data which does not fit the expected Gaussian distribution suitably as they can generate significant error. To this end, the 100 m transects were excluded from further analysis. ΔPNC and ΔCO_2 values for remaining transects were plotted against each other as shown in Figure 5.

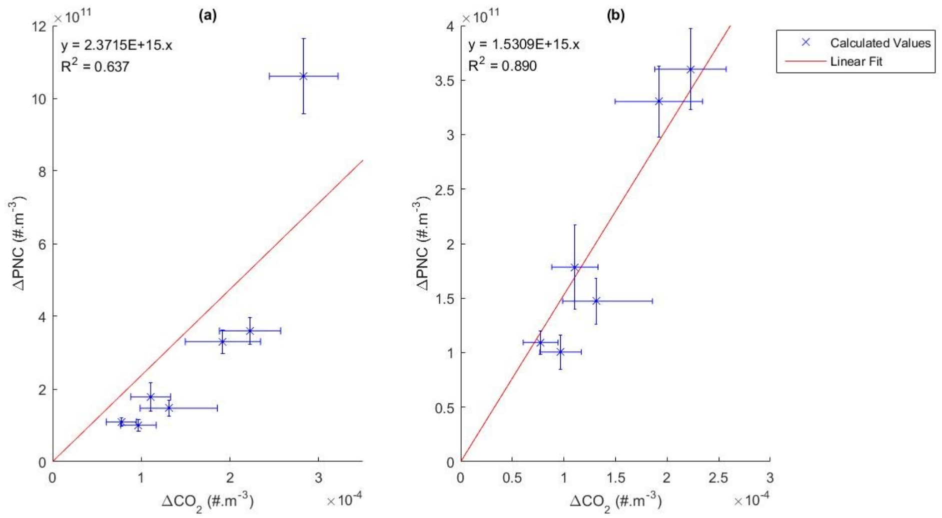

Figure 5 –(a) ΔPNC against ΔCO_2 with 95% confidence interval for the six transects considered for the data analysis. (b) ΔPNC against ΔCO_2 with 95% confidence interval with the removal of the outlier transect from the first flight of day 2

Figure 5 (a) and (b) show the plots of the remaining transects ΔPNC against ΔCO_2 with and without the values of the first 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 0.637 to 0.890 with its exclusion. Furthermore, whilst the linear fit falls within the confidence interval of only one point in (a), it falls within all data points confidence intervals in (b). This occurs despite both R^2 values for the fitted Gaussians of this transect being very high ($R^2_{PNC} = 0.9842$, $R^2_{CO2} = 0.9518$). This highlights a limitation with this methodology which can be best observed in the difference between Figure 4 (a) and (b). The combination of UAV velocity, sampling rate and response time of the DISCmini results in the PNC transect data having only one data point defining the peak height of the transect. Relying on a single sample point leads to the potential for random instrumentation effects heavily biasing results in a way which does not strongly impact the R^2 values of Gaussian fits used to identify successful transects. Therefore, it is unclear whether this is a variation in the ship emissions or an instrumentation error.

The slope and standard error of the linear fit for Figure 4 (a) was input unto Equation 1 to calculate an overall emission factor of $7.6 \pm 1.4 \times 10^{15}$ $\#.kg_{fuel}^{-1}$ As presented in Table 3, this value is comparable with those reported in the literature for cruise and cargo ship plumes; which range from 0.2×10^{16} to 6.2×10^{16} $\#.Kg_{fuel}^{-1}$ {, #45;Alföldy, 2013 #52;Beecken, 2014 #34;Jonsson, 2011 #53;Juwono, 2013 #29;Lack, 2011 #7;Lack, 2009 #37;Pirjola, 2014 #32;Sinha, 2003 #39;Westerlund, 2015 #30}

Reference	Platform	EFPN (#.kgfuel^{-1})	Number of ships	Location
This Study	UAV	7.6 ± 1.4 x 10^{15}	1	Open Water
Westerlund et al. (2015)	Land Based	2.35 ± 0.20 x 10^{16}	154	Harbor, Ship Channel
Beecken et al. (2014)	Airborne	1.8 ± 1.3 x 10^{16}	174	Open Water
Pirjola et al. (2014)	Land Based	0.32 x 10^{16}	11	Harbor, Ship Channel
Alföldy et al (2013)	Land Based	0.8 x 10^{16}	497	Harbor
Juwono et al. (2012)	On Board	0.22 x 10^{16}	2	Harbor, Ship Channel
Jonsson et al. (2011)	Land Based	2.55 ± 0.11 x 10^{16}	734	Harbor
Lack et al. (2009)	Ship	0.71 ± 0.55 x 10^{16} (>13nm)* 1.27 ± 0.95 x 10^{16} (>5nm)**	172 165	Open Water, Shipping Channel
Lack et al. (2011)	Airborne	1.0 ± 0.2 x 10^{16}	1	Open Water
Sinha et al. (2003)	Airborne	6.2 ± 0.6 x10^{16}	2	Open Water

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

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

3.4. Instrumentation Limitations

Lightweight UAVs present an opportunity to achieve aerial measurements at significantly less upfront and operational costs than fixed wing and manned aerial vehicles. Lightweight UAVs can be deployed faster with limited or no required launch and landing area compared to their manned and fixed wing counterparts. Yet, their primary disadvantage, particularly in this application, is a severely limited payload weight. To overcome this limitation, this project used the lightweight and portable DICSmini and IAQ-calc sensors. However, these instruments have lower sensitivities and greater uncertainties when compared to a high accuracy CPC and CO_2 monitor for measurements, which can influence results.

The DISCmini has a manufacturer listed measurement cut-off size of 10 nm. A previous study listed in Table 3 (Lack, Corbett et al. 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 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. This may have been another contributing factor to the EFPN measured in this study being in the lower end of measured values in literature.

The two 100m transects were not accounted for in the final calculation of EF_{PN} due to their poor Gaussian curve fits. Whilst this has been attributed to the skewing of the plume at this distance, the limitations of the instrumentation could also have contributed. The lower concentrations of CO_2 at this distance result in the difference above background inside the plume being the same order of magnitude as the manufacturer specified error margin. Hence, the variability in the plume either side of the central peak as shown in figure 4 (c) could be due in part to instrumentation error.

Calibrations of sensors in this study were performed by comparison with reference instruments for ambient measurements at sea. Ideally, calibration should be performed with in-plume measurements, however it was not possible to access the plume with reference instrumentation on board the ship. Whilst this study provides a successful proof of concept with consistent results over multiple days and flights, a validation study is needed. This should include independent measurements of EF_{PN} using other established methodologies to ascertain more precise correction factors and uncertainties.

4. Summary and conclusion

The UAV system used in this study successfully measured PN and CO_2 concentrations from the exhaust plume of the RV Investigator whilst operating at sea. Several different flight paths were tested and an optimal transect flying perpendicular to the plume at a distance of 20 meters from the ship was adopted. The EF_{PN} calculated for the RV investigator 7.6 ± 1.4 x 10^{15} $\#.kg_{fuel}^{-1}$ at a constant 30% engine load. This EF_{PN} was in agreement with values reported in literature, indicating this novel UAV system has potential for EF_{PN} quantification pending further evaluation.

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

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

Deleted: Furthermore, with PN emissions becomi of ever increasing interest, it will both inform regulatory bodies, and provide them with the tools to monitor emissions in harbours and at sea.

4.1. Recommendations

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

Further optimization of the transect approach is also possible. After location of the plume the system could be set to make several repeat passes across the plume in rapid succession to increase the sample size. Another alternative would involve the UAV hovering inside the plume over a period of time collecting a continuous series of measurements from the centre of the plume. These methods would both require real time sensor feedback to the UAV pilot and potentially adaptive autonomous controls to achieve a suitable result. This methodology could also be expanded to measure other important ship emission factors, including NO_X and volatile organic compounds (VOCs).

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