Response to Reviewers Comments:

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

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The authors thank the Reviewer for the\ comments, and they have modified the manuscript to address them.

Reviewer #1:

Comment 1: Line 22: There is a typo with the first emission factor given.

Answer 1: The typo in the text of the manuscript has been corrected.

Line 22

Comment 2: L24: The authors indicate that they have demonstrated a "reliable, inexpensive and accessible" way of measuring ship emissions. The measurements here required the UAV being deployed from on board the ship. This seems potentially quite limiting. It would be useful if the authors were to rethink the concept of "accessible." I do not dispute that the method here has potential. But, it has not been demonstrated that it is an "accessible" method, especially given the need to optimize the flight path before performing measurements.

Answer 2: The authors have taken into consideration the comment of the Reviewers. The use of the UAV system has been defined accessible because it can be deployed from land and specific flight paths can be designed to assess emissions from ships approaching the port area. Such paths can take into consideration different parameters and conditions such as the morphology of the territory, physical barriers and flying speed. This study was a proof of concept and it was decided to deploy form on board the ship to be able to fly away from other ships and without have to obtain permissions from port authorities and civil aviation authorities. In fact this are still the main limiting factors for a large deployment of UAVs.

Comment 3: L83: I suggest that both “sophisticated” and “world class” be removed. There is no need for these superlatives, nor are they justified by the description.

Answer 3: The authors thank the Reviewer for the comment. The claim has been addressed in the manuscript and the adjectives “sophisticated” and “world class” have been removed from the manuscript.

Line 83

Comment 4: Substantailly more information regarding calibration and testing of the DISCmini and the IAQ-calc are needed. The supplemental has no information on the CO_2 comparison. This should be added. For the particle comparison, the authors should indicate the measurement conditions. As they note, the calibration depends on the assumed particle size distribution. What was chosen for calibration? Was this just ambient air? Particles produced from an atomizer? Are the calibration particles relevant to the particles in the plume in terms of the size distribution? Were the DISCmini concentrations corrected to account for the difference in slopes in Fig. S1? Is that what is meant by calibration, or are the instruments just being compared? How is uncertainty estimated?

Answer 4: The authors thank the Reviewer for the comment. The following paragraph has been added to the Supplementary material document:

“The DISCmini was run in parallel to a CPC 3772 (TSI INCORPORATED 500 Cardigan Road Shoreview, MN, USA) which has a low cut off point of 10 nm. The two instruments were used to sample ambient air from the front mast of the ship. The uncertainty was estimated from the fitting procedure as shown in Figure 5. The IAQ-Calc was placed on the front mast of the ship and the readings were compared to those acquired by the PICARRO spectrophotometer G2301.”

Comment 5 :Eqn. 1: The authors use the integrated peak concentrations to calculate the ratio between delta values and the EF. From Fig. 4 it is evident that the CO_2 plume is broader than the PN plume. The authors should consider discussing this issue in the context of how it impacts their EF estimates.

Answer 5: The issues with the low amount of data points inside the peak has been addressed in the updated discussion.

Page 11; Lines 279-288

“Figure 5 (a) and (b) show the plots of the remaining transects ΔPNC against ΔCO2 with and without the values of the first flight of day 2. This transect represents a clear outlier in the linear trend, with the R2 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 R2 values for the fitted Gaussians of this transect being very high (R2PNC = 0.9842, R2CO2 = 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 R2 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.”

Comment 6: L240: Here, the authors focus on differences in absolute values. Such differences can result for a variety of reasons. What really matters, though, is how different the derived EF values are. I suggest that the authors bring the EFs for these plumes into the discussion.

Answer 6: The authors have considered the Reviewer’s comment and believe that the claim has been addressed in the discussion paragraph from line 290-297.

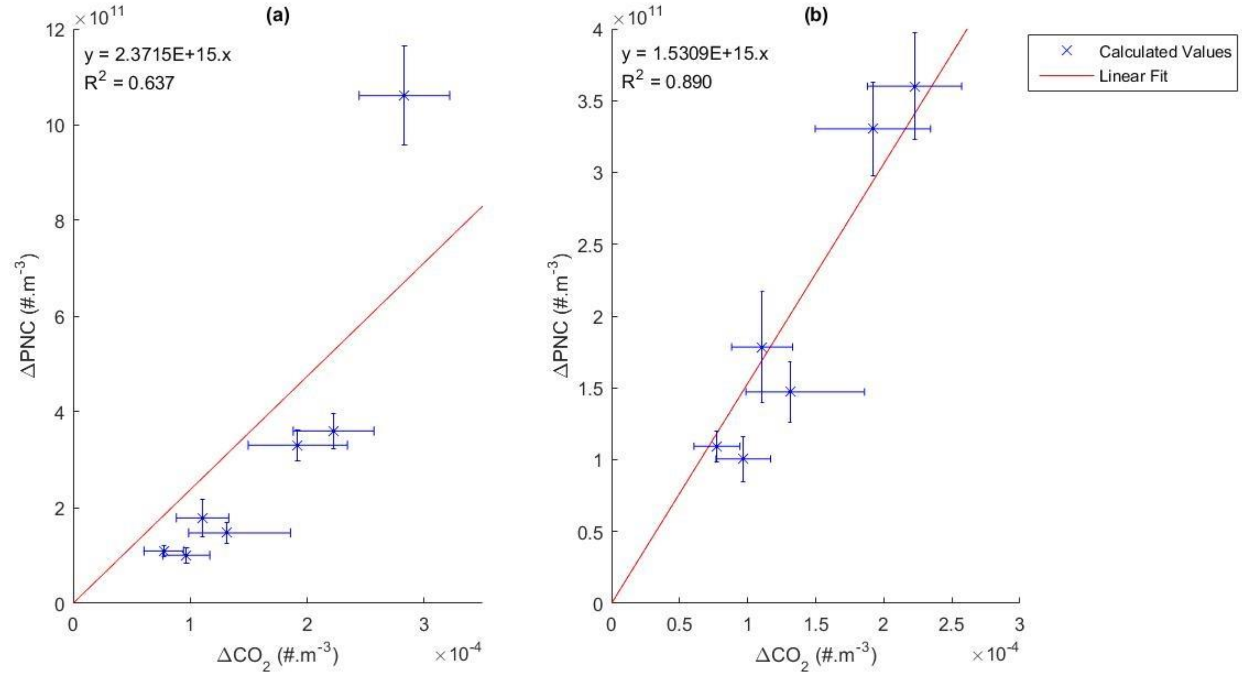
Comment 7: Table 2 and Fig. 5: The units given for CO_2 are not correct. This must be kg/mˆ3. It is not possible to simply have kg as the units, since the volume is not known. Also, if the units are not kg/mˆ3, then the units on the derived EFs will not make sense.

Answer 7: The authors thank the Reviewer for the comment. The claim has been addressed in the manuscript and both Table 2 and Fig. 5 have been corrected.

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

Comment 8: Table 2 vs. Fig. 5: There seems to be an inconsistency. The slope from Fig. 5 can be converted to an EF by multiplying by 3.2 kg CO2/kg-fuel. This yields 6.4e15 particles/kg-fuel. But, the average from Table 2 is only 2.6e15 particles/kg-fuel. These should be closer. The difference may be because the authors have not fixed their intercept to zero. This should be looked at by the authors. Also, in Fig. 5 the x-axis should start from zero.

Answer 8: The authors have considered the Reviewer's comment. In the new methodology the final EF_{PN} is calculated using the slope of the line with the intercept fixed at zero as recommended here. The axis has also been updated to start from origin.



Comment 9: L283: The authors assert that the 20 m intercepts will give more reliable results than the 100 m intercepts. However, at 100 m the plumes are wider, which offsets somewhat the benefit of greater amplitude of the 20 m intercepts. The authors do not provide an uncertainty analysis currently. The statement here should be justified by demonstrating that the EFs from the 20 m intercepts truly do have lower uncertainties than the 100 m intercepts. The contrast with the background is part of the story, but not the only factor that impacts the uncertainty. For a methods paper, I expect to see more rigorous consideration of measurement uncertainty than is currently provided.

Answer 9: The Reviewer's comment has been addressed with the methodology being updated. The updated methodology is based on the fitting on Gaussian curves to the transect data in order to find more significant Δ values. As discussed in the updated manuscript, the broadening of the plume results in significantly poorer Gaussian fits for several reasons. To this end the were excluded from the final calculation of EF_{PN}. This discussion can be found in lines 262-267.

Comment 10: L286: While yes, the observations are "comparable" with other measurements, the authors should certainly note that their measurements are very much on the low end of the literature range.

Answer 10:The updated calculations result in an EF_{PN} more within the range of published values, however can still be considered to be in the lower end. Discussion has been added which address potential reasons for this in lines 290-297 and 308-312.

Comment 11: Table 3: The authors need to include Lack et al. (2009, JGR) in their comparison table and in discussion in the text. Lack et al. (2009) report measurements from a variety of different ship types based on plume intercepts. Their work also clearly shows that the exact EF that one obtains for particles depends on the lower size threshold of the measurement. Here, the authors indicate that it is 10 nm. But, at the same time, the calibration is dependent on the particle size distribution. These issues should certainly be discussed in the context of discussing the measurement accuracy. Perhaps the measured EFs here are on the low side because they really are. But, it may be that some aspect of this is a result of the particular calibration method and the measurement uncertainty. Uncertainties must be discussed more fully, in general.

Answer 11:

Lack et al. (2009, JGR) has been added into Table 3. Discussion into the limitations around PNC measurements with the current methodology, included size, has been expanded upon in lines 308-312.

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

Comment 12: L299: it is unclear what “in-land transportation” means. Only in looking at the reference is it clear that this means buses operating on “compressed natural gas and ultralow sulfur diesel.” It seems that the authors are arguing here that their low PN EF values are a result of the fuel sulfur difference from some of the literature studies. However, I do not find this argument compelling for the simple reason that bus engines are not comparable to marine engines. If the authors want to make this argument, they should compare more directly with ship measurements. For example, the Lack et al. (2011) paper compares PN EF values from before and after a ship in operation switches to low sulfur fuel. They see a negligible difference on the particle number, although the particle mass concentration decreases. This conflicts with the argument that the authors seem to be advancing here through their comparison with a bus study. The same goes for the comparison to the aircraft study. While it is perhaps interesting to compare between engine types, this does not provide any indication that the fuel is what drove this difference.

Answer 12:The authors have discussed the Reviewer’s comment. In Lack et al. (2011) paper referenced the comparison is between very high sulfur fuel and high sulfur fuel, where reduction in PM mass is observed. The ultra-low sulfur diesel used by the investigator has significantly lower sulfur content than this. In a paper by Ristovski et al. (2006) it was shown the reduction to comparable levels of sulfur content does lead to a reduction in PM number concentration. This has been added into the discussion section. This is elaborated in lines 290-297.

Comment 13: L311: The authors talk about their method being "validated" because they fall in the range previously observed for ships. To me, this is marginal. A true validation would have used a separate method to measure the EF for this particular ship. This was not done. No discussion of measurement uncertainty has been provided. Thus, we have no way of knowing whether the fact that the measurements here are on the low end of the literature range is because the ship simply had a lower EF or was a result of the measurement itself. For a methods paper, this lacks sufficient details regarding measurement calibration and testing. This is certainly an interesting proof of concept. But, I have substantial concerns regarding the use of terms such as "validation" given the lack of uncertainty analysis or full discussion of specific issues associated with PN measurement using the DISCmini. I think that this paper will only be publishable with a substantially more robust discussion of uncertainties.

Answer 13:The updated manuscript attempts to deal with the uncertainties involved in this study with a more robust data analysis and consideration of experimental errors. The word "validated" has been removed as it is agreed that it will be necessary to compare this alongside other developed methodologies before it can be truly validated. Instead, we are treating the study as a proof of concept, and have attempted to highlight the benefits and drawbacks to inform future method development. This has involved many changes across the results and discussion section.

Comment 14: Grammar note: The authors consistently say that the "Data was." It should usually be "data are."

Answer 14: The authors thank the Reviewer for the grammar note, this has been fixed in the updated manuscript.

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.

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, ship-size, 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 is a proof on concept.

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 PM emission factors using a relatively inexpensive UAV. This is primarily is for academic purposes. However, PM has been identified as critical to both health and climate and thus developing the basis for tools which may have potential regulatory of PM emissions is important.

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.

Comment 5: Line 142: Sensitivity range for CO2 is 50ppm, this is important as this is same order of magnitude as the delta CO2 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.

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 CO2 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, 307-310).

Answer 6: Methodology has been expanded upon significantly in the updated manuscript and a $C0_2$ picaro comparison is provided in 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 ransect 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.

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.

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.

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

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

Comment 11: Line 232: It is mentioned that the CO2 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 CO2. Furthermore it should be noted that the peaks for CO2 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.

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 EF_{PN} 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.

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.

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.

Comment 15: Line 326: SO_2 is completely missing here, SO_2 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. SO_2 would be an interesting alternate application. To the authors knowledge the main challenge for such a system would be that fast and accurate SO_2 meters are significantly above the payload of any lightweight UAV, include fixed wings.