Answer to Referee #1

We would like to thank Referee #1 for his/her positive and constructive comments and suggestions. We have studied comments carefully and made corrections, which we hope meet with approval. Comments and responses are listed as follows. In order to facilitate the reference to the questions and proposed changes, we use the following color coding:

Color coding:

Referee comment

Our answer

Proposed change in manuscript

The title is sloppy: "High-precision monitoring of compliance with fuel sulfur content through UAV measurements of ship emissions" Should be rather: "High-precision monitoring of compliance with fuel sulfur content regulations/limitation/standards through UAV measurements of ship emissions"

Considering the content of the manuscript, we guess the word "regulations" is more appropriate. At the same time, Referee #2 suggest leaving out the word "High-precision" because "in ECA areas, with a limit of 0.1%, an uncertainty of 0.03% is not very good". In addition, with the consideration that the precision will improve further, with the progress of technology and method. "High-precision" seems not appropriate, we changed the title as:

"Monitoring of compliance with fuel sulfur content regulations through UAV measurements of ship emissions".

1. Introduction

The literature review is poor.

The suggestion is very important. We have rewritten this part carefully. The major changes are listed below.

Page1, line20: Year of citation is 2005. Add current data here since anthropocentric SO_2 emission might change significantly during 14 years.

This paragraph has been rewritten.

Estimations show that ships contribute 4-9% of global SO_2 emissions and 15% of NO_x (Eyring et al., 2010). According to the United Nations Conference on Trade And Development (UNCTAD, 2017), the volume of the world's seaborne trade grew by 66% between 2000 and 2015. As global commerce expands, ocean-going ships consume more fuels, generally low-quality residual fuels containing high concentrations of sulfur and heavy metals (Lack et al., 2011). From the viewpoint of spatial distribution, the highest emissions of SO_2 per unit area occur in the eastern and southern China seas, sea areas in south-eastern and southern Asia, Red Sea, Mediterranean Sea, North Atlantic near the European coast,

Gulf of Mexico and Caribbean Sea, and along the western coast of North America. (Johansson et al., 2017). Ship-emitted pollutants influence air quality, human health, and climate. They not only affect the air quality in coastal areas but even influence the inland areas hundreds of kilometers away from the emission sources (Liu et al., 2016).

Reference:

[1] Eyring, V., Isaksen, I. S., Berntsen, T., Collins, W. J., Corbett, J. J., Endresen, O., Grainger, R. G., Moldanova, J., Schlager, H., and Stevenson, D. S.: Transport impacts on atmosphere and climate: Shipping, Atmos. Environ., 44, 4735–4771, https://doi.org/10.1016/j.atmosenv.2009.04.059, 2010.

[2] UNCTAD: World seaborne trade by types of cargo and by group of economies, annual, United Nations Conference on Trade and Development, available at: https://unctadstat.unctad.org/wds/TableViewer/tableView.aspx?ReportId=32363, last access: 5 March 2017.

[3] Lack, D. A., Cappa, C. D., Langridge, J., Bahreini, R., Buffaloe, G., Brock, C., Cerully, K., Coffman, D., Hayden, K., Holloway, J., Lerner, B., Massoli, P., Li, S.-M., McLaren, R., Middle-brook, A. M., Moore, R., Nenes, A., Nuaaman, I., Onasch, T. B., Peischl, J., Perring, A., Quinn, P. K., Ryerson, T., Schwartz, J. P., Spackman, R., Wofsy, S. C., Worsnop, D., Xiang, B., and Williams, E.: Impact of Fuel Quality Regulation and Speed Reductions on Shipping Emissions: Implications for Climate and Air Quality, Environ. Sci. Technol., 45, 9052-9060, https://doi.org/10.1021/es2013424, 2011.

[4] Johansson, L., Jalkanen, J. P., and Kukkonen, J.: Global assessment of shipping emissions in 2015 on a high spatial and temporal resolution, Atmos. Environ., 167, 403-415, https://doi.org/10.1016/j.atmosenv.2017.08.042, 2017.

[5] Liu, H., Fu, M., Jin, X., Shang, Y., Shindell, D., Faluvegi, G., Shindell, C., and He, K.: Health and climate impacts of ocean-going vessels in East Asia, Nat. Clim. Change., 6, 1037-1041, 10.1038/nclimate3083, 2016.

Pg1, ln25: "some regulation went into effect" - needed to be rephased.

These sentences have been rewritten.

In 2005, some regulations went into effect after being received by appropriate laws of the signatory states (at the European level it was received with the directives 1999/32/EC, 1999, and 2005/33/EC, 2005), and introduces limits to marine fuel sulfur content and engine performance to reduce SO_x and NO_x emissions. Further amendments to Annex VI were adopted in 2008 and entered into force in 2010.

Reference:

[1] Directive 1999/32/EC: Official Journal of the European Union, L 121, p. 13, 26 April 1999.

[2] Directive 2005/33/EC: Official Journal of the European Union, L 191, p. 59, 22 July 2005.

Pg2, ln4: "To cope with..." ???

This sentence has been rewritten.

In order to reduce the air pollution caused by ship emissions, the Atmospheric Pollution Prevention and Control Law of the People's Republic of China was promulgated in 2015 (Standing Committee of the National People's Congress, 2015).

Reference:

[1] Standing Committee of the National People's Congress, Atmospheric Pollution Prevention and Control Law of the People's Republic of China, 2015.

The overview of techniques (Pg2, ln15-ln26) is incorrect. The optical methods (LIDAR, UV cam, DOAS) can measure only the SO_2 emission rate. For emission factor calculation simultaneous CO_2 emission rate measurement is needed preferable on the same parcel of the plume. It can be implemented by open path FTIR technique but it is quite challenging. Another solution could be to model the CO_2 emission rate based on the ship's technical properties and sailing characteristics. Then the SO_2 or S emission factor (EF) as well as the fuel sulphur content (FSC) can be calculated.

In the manuscript, we only discuss that the optical methods can be used to measure ship emissions (do not indicate that they could measure the CO_2). We think this part was not clear enough, so we have rewritten it combined with the suggestion.

Optical methods analyze variations in light properties after interactions with the exhaust plume, and the local wind field before determining the SO_2 emission rate is observed. The simultaneous measurement of CO_2 and SO_2 emissions on a routine basis is unrealistic at present. Thus, the amount of fuel burned at the time of measurement is unknown and has to be estimated via modeling for calculating the FSC. For instance, the model STEAM (ship traffic emission assessment model), developed by the Finnish Meteorological Institute (Jalkanen et al., 2009) was used in the research for estimating FSC by Balzani Lööv et al. (2014). In addition, using the ratio of SO_2 and NO_2 measured via DOAS in the ship plume can be used as an indicator of FSC (Johan, R et al. 2017, Cheng, Y et al, 2019).

Reference:

[1] Balzani Lööv, J. M., Alfoldy, B., Gast, L. F. L., Hjorth, J., Lagler, F., Mellqvist, J., Beecken, J., Berg, N., Duyzer, J., Westrate, H., Swart, D. P. J., Berkhout, A. J. C., Jalkanen, J.-P., Prata, A. J., vander Hoff, G. R., and Borowiak, A.: Field test of available methods to measure remotely SO_x and NO_x emissions from ships, Atmos. Meas. Tech., 7, 2597–2613, doi:10.5194/amt-7-2597-2014, 2014.

[2] Cheng, Y., Wang, S., Zhu, J., Guo, Y., Zhang, R., Liu, Y., Zhang, Y., Yu, Q., Ma, W., and Zhou, B.: Surveillance of SO_2 and NO_2 from ship emissions by MAX-DOAS measurements and implication to compliance of fuel sulfur content, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-369, in review, 2019.

[3] Johan, R., Conde, V., Beecken, Jörg and Ekholm, J.: Certification of an aircraft and airborne surveillance of fuel sulfur content in ships at the SECA border, CompMon (https://compmon.eu/), 2017.

[4] Jalkanen, J.-P., Brink, A., Kalli, J., Pettersson, H., Kukkonen, J.,and Stipa, T.: A modelling system for the exhaust emissions of marine traffic and its application in the Baltic Sea area, Atmos. Chem.

Phys., 9, 9209–9223, doi:10.5194/acp-9-9209-2009, 2009.

Pg2, ln25: What is the effect of NO_x sensor on FSC measurement?

The SO_2 analyzer (fluorescence) response has cross sensitivity to NO. The supplementary explanation is given in the manuscript.

The "sniffing" method is based on simultaneous measurement of elevated SO_2 and CO_2 concentrations in the exhaust plume from the target ship and comparing them with the background. The measurement of CO_2 allows for relating the measurement of SO_2 to the amount of fuel burned at a given time, thus enabling the calculation of FSC directly. The concentration of SO_2 in plumes was generally measured using UV fluorescence sensors, and CO_2 was measured using a non-dispersive infrared analyzer (NDIR) or cavity ring down spectrometer (CRDS). The advantage of the "sniffing" method is that it offers more accuracy estimation for FSC. However, the instrument must be placed in the plume exhausted by the target ship. In some studies (Van Roy and Scheldeman, 2016a, 2016b), the "sniffing" method offers a measurement accuracy between 0.1–0.2% (m/m) FSC, which can be further increased up to 0.05–0.1% (m/m) FSC if combined with an additional NO_x sensor. This is because the response of SO_2 analyzers (fluorescence) has cross sensitivity to NO. Deviations are not the same at different FSC levels, with an estimated relative uncertainty of 20% (m/m) for ships with 1% (m/m) FSC and a relative uncertainty of 50–100% at 0.1% (m/m) FSC. Balzani Lööv et al. (2014) obtained the following FSC measurements based on the "sniffer" principle: 0.86±0.23% (m/m) from land, 1.2±0.15% (m/m) from an on-board stack, and 1.13±0.18% (m/m) from a mobile platform. There was a 6% relative uncertainty for an FSC of 1% (m/m) but a 60% relative uncertainty for an FSC of 0.1% (m/m).

Reference:

[1] Balzani Lööv, J. M., Alfoldy, B., Gast, L. F. L., Hjorth, J., Lagler, F., Mellqvist, J., Beecken, J., Berg, N., Duyzer, J., Westrate, H., Swart, D. P. J., Berkhout, A. J. C., Jalkanen, J.-P., Prata, A. J., vander Hoff, G. R., and Borowiak, A.: Field test of available methods to measure remotely SOx and NOx emissions from ships, Atmos. Meas. Tech., 7, 2597–2613, doi:10.5194/amt-7-2597-2014, 2014.

[2] Van Roy, W. and Scheldeman, K.: Results MARPOL Annex VI Monitoring Report Belgian Sniffer Campaign 2016, CompMon (https://compmon.eu/), 2016a.

[3] Van Roy, W. and Scheldeman, K.: Best Practices Airborne MARPOL Annex VI Monitoring, CompMon (https://compmon.eu/), 2016b.

In addition, several UAV applications have been done before that must be mentioned here.

After reviewing relevant literatures, we found that there are some UAVs used to measure greenhouse gases and volcanic eruptions. Only one paper has been found on the measurement of ship emissions. We also have carried on the supplementary discussion in the manuscript.

Ship emission measurements can be divided into land-based (Kattner et al., 2015, Yang et al., 2016), marine-based (Cappa et al., 2014), airborne-based (Beecken et al., 2014, Aliabadi et al., 2016), satellite-based (Ding et al., 2018) and Unmanned Aerial Vehicle (UAV)-based (Villa et al., 2019) according to different platforms.

…

UAV-based measurements have gradually increased in the research regarding the atmosphere (Malaver Rojas et al., 2015, Mori et al., 2016). However, to date, there are relatively few applications of these measurements in ship emissions.

Reference:

[1] Aliabadi, A. A., Thomas, J. L., Herber, A. B., Staebler, R. M., Leaitch, W. R., Schulz, H., Law, K. S., Marelle, L., Burkart, J., Willis, M. D., Bozem, H., Hoor, P. M., Köllner, F., Schneider, J., Levasseur, M., and Abbatt, J. P. D.: Ship emissions measurement in the Arctic by plume intercepts of the Canadian Coast Guard icebreaker Amundsen from the Polar 6 aircraft platform, Atmos. Chem. Phys., 16, 7899–7916, doi:10.5194/acp-16-7899-2016, 2016.

[2] Beecken, J., Mellqvist, J., Salo, K., Ekholm, J., and Jalkanen, J.P.: Airborne emission measurements of SO_2, NO_x and particles from individual ships using a sniffer technique, Atmos. Meas. Tech., 7, 1957–1968, doi:10.5194/amt-7-1957-2014, 2014.

[3] Cappa, C. D., Williams, E. J., Lack, D. A., Buffaloe, G. M., Coffman, D., Hayden, K. L., Herndon, S. C., Lerner, B. M., Li, S.M., Massoli, P., McLaren, R., Nuaaman, I., Onasch, T. B., and Quinn, P. K.: A case study into the measurement of ship emissions from plume intercepts of the NOAA ship Miller Freeman, Atmos. Chem. Phys., 14, 1337–1352, doi:10.5194/acp-14-1337-2014, 2014.

[4] Ding, J., van der A, R. J., Mijling, B., Jalkanen,J.-P.,Johansson,L.,and Levelt,P.F.: Maritime NOx emissions over Chinese seas derived from satellite observations. Geophysical Research Letters, 45, 2031–2037, doi:10.1002/2017GL076788, 2018.

[5] Kattner, L., Mathieu-Üffing, B., Burrows, J. P., Richter, A., Schmolke, S., Seyler, A., and Wittrock, F.: Monitoring compliance with sulfur content regulations of shipping fuel by in situ measurements of ship emissions, Atmos. Chem. Phys., 15, 10087–10092, doi:10.5194/acp-15-10087-2015, 2015.

[6] Villa, T. F., Brown, R. A., Jayaratne, E. R., Gonzalez, L. F., Morawska, L., and Ristovski, Z. D.: Characterization of the particle emission from a ship operating at sea using an unmanned aerial vehicle, Atmos. Meas. Tech., 12, 691-702, https://doi.org/10.5194/amt-12-691-2019, 2019.

[7] Yang, M., Bell, T. G., Hopkins, F. E., and Smyth, T. J.: Attribution of atmospheric sulfur dioxide over the English Channel to dimethyl sulfide and changing ship emissions, Atmos. Chem. Phys., 16,4771–4783, https://doi.org/10.5194/acp-16-4771-2016, 2016.

As a summary; a new and more precise Introduction is needed.

We have tried our best to rewrite the introduction.

2. Measurement:

Avoid mini-sniffer term. Sniffing technique supposes an airflow through the analyzer. In this context the CO_2 analyzer can sniff but the SO_2 sensor definitely cant. Better to use sensing or

electrochemical sensing terms.

The term of “mini-sniffer” in the manuscripts has been revised.

This section is also incomplete. What sensors were used? Manufacturers, types, characteristics? The description of the sniffing technique (Pg5) is poor and incomplete. For example, the proper handling of the water vapor interference with CO_2 measurements is crucial. How did the authors handle that?

The sensors are commercially available. This information has been supplemented.

In the measurement process, the ship exhaust is pumped into the pod by the gas pump. After the filter removes the water vapor, the sensors react and the communication module sends the measurement results to the receiving end. The sensors included instrumentation for both SO_2 and CO_2 measurements. These sensors were purchased from HH Feuerungstechnik GmbH, Germany.

In addition, we supplement the description in figure 1.

Figure 1. Image of the modified UAV platform. The black box installed under the UAV is a pod which was designed and customized by us. It carries a gas pump (to collect the ship's exhaust), gas circuit, a filter (to remove water vapor), sensors for SO_2 and CO_2, a small motor (to provide energy for pumping), a camera, and communication modules.

In the experiment, we used the MATRICE 600 UAV (SZ DJI Technology Co., Ltd.), and modified it. We designed and customized a special pod, which was installed underneath the UAV, to carry sensors, communication circuit boards, gas circuit systems, and other modules, as shown in Fig.1.

Ln13: "calibrated 3 month or 180 working hours apart" ??? - why is this big difference?

This sentence has been rewritten.

Sensor calibration is required when the equipment is used daily. The time interval for sensor calibration is three months or when the accumulated working time of the sensor exceeds 180 h. If either of these conditions is met, calibration will be carried out.

I miss evidences of laboratory experiments where the sensors were calibrated and tested, effects of environmental factors (temperature, humidity) were investigated as well as interference of other components (water vapor) was checked.

In the laboratory experiment, we mainly test the stability and safety of the whole UAV system as well as communication modules. At the same time, it also allows the UAV operator to practice how to operate the UAV for sampling close to the smoke stack. There is a risk of getting too close, the operator needs to practice.
We used commercial sensors. We chose the type of sensors according to the need of experiment. We have added some details (sources of sensors, technical parameters, calibration, filters, etc) about the whole pod as mentioned above. We hope this will make the readers more familiar with our work.

In the experiment, we used the MATRICE 600 UAV (SZ DJI Technology Co., Ltd.), and modified it. We designed and customized a special pod, which was installed underneath the UAV, to carry sensors, communication circuit boards, gas circuit systems, and other modules, as shown in Fig.1. After the successful assembly of the UAV platform, we first carried out preliminary experiments in the automatic engine room laboratory of Shanghai Maritime University. Through the preliminary test, we verified the stability and security of the whole UAV system. At the same time, it also allowed the UAV operator to practice how to operate the UAV for sampling close to the smoke stack.

4. Results

The authors claimed that they measured 20 ships. Why only 6 plumes were presented here? It is not clear how the authors accepted or discarded results. What were the main steps of the consideration?

At the time of initial submission, we listed 12 plumes. However, similar plumes do not seem to need to be listed multiple times. Therefore, we chose the typical six plumes and discussed in detail the process of selecting the peak and background values. The data of these six plumes are only suitable for our discussion method, and have no other particularities. In addition, we supplemented the results of all 23 monitoring experiments in the manuscript.

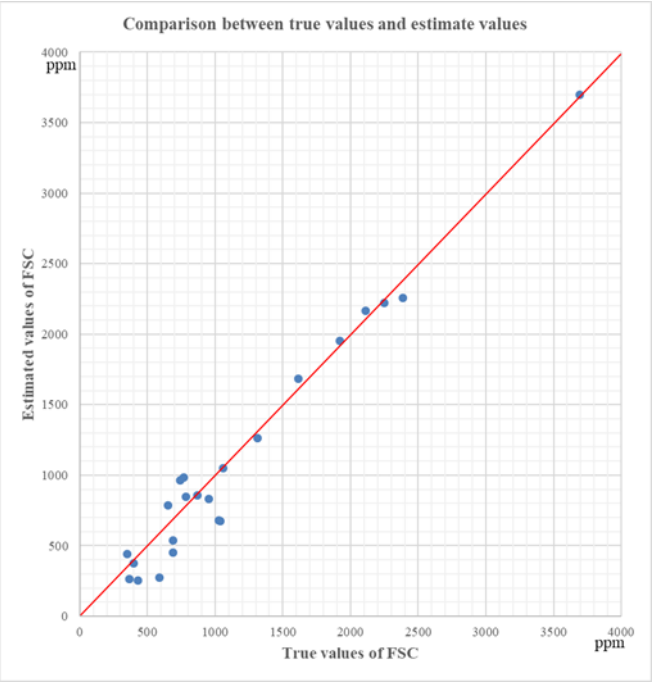

Figure 6. Comparison between the true values of FSC (x-axis) against the estimated values of FSC (y-axis) of 23 times measurement.

As shown in Fig 6, the FSC in our experiments was mainly at a level of 0.035% (m/m) to 0.24% (m/m) (only one measurement of 0.37% (m/m), not enough for reference). The deviation of the estimated FSC value calculated using the proposed method was within 0.03% (m/m), although there was some uncertainty. Considering the uncertainties listed in section 3.3, the proposed method provides accurate results. Overall, the estimated FSC is smaller than the true value in many cases. This is because 1–19% of the sulfur in the fuel is emitted in other forms, possibly SO_3 or SO_4.

On the other hands, the main strength of the work that the authors compared their plume measurements with the chemical analysis of the fuel. The authors did not mention the biggest challenge of the technique, namely how can we synchronize the time variations of two different measurements (SO_2 and CO_2) in order to calculate their ratio. In case of broader plume parcels (and thus longer measurement time) during conventional sniffing technique the uncertainty of the integrals are negligible, so more or less exact ratio can be calculated. On the other hands, the sensing technique provides narrow peaks of both components where the uncertainty of the integral is significant. How can the authors describe the differences in the time variation of the two components (see the figures on Pg10)?

Yes, this is indeed a key technical problem we encountered. We have added the following explanation:

The response time of both sensors is less than 1s. Even if the sampling rates of the two sensors are set to be consistent, the two sensors cannot be completely synchronized. This makes it difficult to calculate

the ratio of SO_2 and CO_2. Our approach is that the sensor sends the average measurement value of the last 10 s to the receiver at an interval of 10 s. Therefore, the interval of integration in Eq. (1) is 10 s. We determined that taking the mean of measurements directly or at shorter intervals leads to too many narrow peaks in one measurement process. This makes it difficult to select the peak value, and the calculation results are unstable. At the same time, the interval should not be set too long, which will make the crest very inconspicuous or too flat. Therefore, we selected 10 s as the empirical parameter value after several experiments.

Also, in the description of result:

After the measurement of plume 5, the communication module was fault when we wanted to adjust sampling rate. We consequently replaced the communication protocol "HTTP protocol" with the "TCP/IP protocol". The main changes involved adjusting the data sampling rate from 10 to 2 s to make it easier to find the peak value (the sensor sends the average measurement value of the last 10 s to the receiver at an interval of 2 s), and the sensors were consequently recalibrated by standard mixture gas.

In Plume 5 and Plume 6 the CO_2 sensor was saturated at different concentrations (1900 vs 5000 ppm). What was the reason?

The CO_2 sensor has a range of 5000ppm. There appears saturation in plume 6 but not in plume 5. It looks like saturated, but it's not saturated. We checked the data as fellow:

time	SO_2	CO_2
11:18:14	774	1122.8
11:18:25	993	1616
11:18:35	938	1895.2
11:18:46	938	1895.2
11:18:57	1049	1896.8
11:19:08	1406	1893.6
11:19:18	1348	1894.8
11:19:29	1078	1896
11:19:40	765	1886
11:19:51	635	1516
11:20:01	603	1066.8
11:20:12	576	811.6

This kind of situation is rare. It is difficult to draw conclusions at this time. We guess that this may be due to sensor uncertainty. In any case, the data in this period were not used as peak values of the plumes as present in the manuscripts. Because there was no significant spike in the data over this time period.

In the end, we thank the Referee #1 for his/her positive and constructive comments.

Answer to Referee #2

We would like to thank Referee #2 for his/her positive and constructive comments and suggestions. We have studied comments carefully and made corrections, which we hope meet with approval. Comments and responses are listed as follows. In order to facilitate the reference to the questions and proposed changes, we use the following color coding:

Color coding:

Referee comment

Our answer

Proposed change in manuscript

Title: Without trying to be negative I would suggest leaving out the words "High-precision".

Our initial use of the term was based on the fact that UAV measurements can be made more closely to the funnel of ship, to obtain high-precision results. But on reflection, we think the precision will improve further, with the progress of technology and method. "High-precision" is not appropriate; we changed the title as:

"Monitoring of compliance with fuel sulfur content regulations through UAV measurements of ship emissions"

Abstract: As in the title I would suggest leaving out " high precision" in the last sentence. I would also mention the range of sulfur contents that were encountered in the study i.e. how many non-conformities were encountered. And I would like to mention more explicitly that the deviation of the estimated value for +FSC is less than 0.03% (m/m) at a level of 0.04 % to 0.24 % FSC. Note that in ECA areas, with a limit of 0.1%, an uncertainty of 0.03% is not very good. I would also suggest mentioning that in all cases the estimated FSC was always lower than the actual FSC derived from samples taken on board. This is an important aspect with a strong impact on the usefulness of the method in SECA areas with a 0.1% limit value.

"High-precision" has been leaving in whole manuscript. The range of sulfur contents is very important for this research, which should be mentioned in the abstract, result and conclusion. These parts have been rewritten. In addition, the discussion about underestimate of FSC has also add in the conclusion.

In abstract:

After more than 20 comparative experiments, the results show that, in general, the deviation of the estimated value for FSC is less than 0.03% (m/m) at an FSC level ranging from 0.035% (m/m) to 0.24% (m/m). Hence, UAV measurements can be used for monitoring of ECAs for compliance with FSC regulations.

In result:

As shown in Fig 6, the FSC in our experiments was mainly at a level of 0.035% (m/m) to 0.24% (m/m) (only one measurement of 0.37% (m/m), not enough for reference). The deviation of the estimated FSC value calculated using the proposed method was within 300 ppm (0.03% (m/m)), although there was

some uncertainty.

In conclusion:

In general, the deviation of the estimated FSC value was within 0.03% (m/m) at an FSC level of 0.035% (m/m) to 0.24% (m/m). Because not all the sulfur in the fuel is emitted as SO_2, the estimated FSC is smaller than true value in many cases. Therefore, if the maritime department wants to take the estimated value as the basis for the preliminary judgment regarding whether the ship exceeds the emission standard, it needs to set an appropriate threshold and a confidence interval.

How many non-conformities were encountered.

In the result, we discuss that:

In addition, when the FSC of the target ship is low, for example, when the fuel used is light diesel fuel, the SO_2 observation values were mostly 0. When this happened, according to our experience, the FSC was generally lower than 200 ppm, and the ship was likely to meet the emission requirements.

In the conclusion, we discuss that:

1. In about 10% of the cases, the UAV did not measure the effective background value and peak value. This is mainly caused by the UAV missing the plume during its flight. Therefore, effective methods for finding and navigating to plumes using real-time sensor feeds need to be explored.
2. In about 10% of the cases, the absolute error was more than 0.03% (m/m), and even more than 0.05% (m/m) in rare cases. Unstable concentrations of SO_2 or CO_2 in the atmosphere just before the measurement may cause such errors. Furthermore, uncertainties, such as sensor uncertainty, exhaust uncertainty, measurement uncertainty, and calculation uncertainty, may hinder accurate measurement.

Paper: This could be a very useful paper with lots of detail. Especially the level of detail is useful since this is an area with a lot of development and sharing of these new results could very helpful to other scientists. I provide some comments that could help to make the paper a bit clearer in some areas. See my specific comments below.

Thank you for the comments, we are very encouraged.

Figure 1: I am not familiar with UAVs and in a first glance I thought the black box mentioned in the text was the large flight case black box below the drone. Page 3 line 16. Not everybody may be familiar with the word "Pod".

Yes, this "Pod" was designed and customized by us. It's not a commercial product. At first glance it may indeed seem puzzling. We have explained it in more detail in the title of figure 1 and text.

Figure 1. Image of the modified UAV platform. The black box installed under the UAV is a pod which was designed and customized by us. It carries a gas pump (to collect the ship's exhaust), gas circuit, a filter (to remove water vapor), sensors for SO_2 and CO_2, a small motor (to provide energy for pumping), a camera, and communication modules.

In the experiment, we used the MATRICE 600 UAV (SZ DJI Technology Co., Ltd.), and modified it. We designed and customized a special pod, which was installed underneath the UAV, to carry sensors, communication circuit boards, gas circuit systems, and other modules, as shown in Fig.1.

Page 4 last sentence: electrochemistry method. Electrochemical method?

Electrochemical method, the term has been rewritten.

Page 5 line 12-13. These sentences are rather unclear. What is meant with 180 working hours apart? Each 180 working hours? It is not entirely clear what the actual accuracy is if it is 1% full scale.

This sentence has been rewritten. The accuracy is written as $\pm$0.25 ppm for SO_2 and ±50 ppm for CO_2, respectively.

Sensor calibration is required when the equipment is used daily. The time interval for sensor calibration is three months or when the accumulated working time of the sensor exceeds 180 h. If either of these conditions is met, calibration will be carried out.

Page 7 line 16: correction should be corrected. Gradually establishing a quality management system.... Is rather vague what is meant. Please rephrase.

These sentences have been rewritten.

As for sensor uncertainty, the linear error is negligible and the nonlinearity of the two sensors should be no more than ±1%. It can be corrected through frequent calibrations with standard gases and gradually establishing a quality management system comprising sensor linearity, sensitivity, repeatability, hysteresis, resolution, stability, drift, and other attributes of the minimum requirements.

Page 7 line 22. Here 200 ppm is mentioned where in other places in the text 0.03 % (300 ppm) is mentioned. This should be explained or there should rather be only one number. Same place: the deviations mentioned in Balzani et al. (2014) were determined at FSC of 1%. It is not clear whether these deviations are still the same at 0.1% FSC. They could be lower at 0.1% FSC content. The authors should mention that or provide more information (which would be useful)

Yes, the measurement range of the FSC is very important information when discussing the measurement results. We supplement the information of measuring range when we discuss the relative precision. We have made the following description for the "200ppm".

Exhaust uncertainty arises because not all the sulfur in the fuel is emitted as SO_2. Preliminary studies showed that 1-19% of the sulfur in the fuel is emitted in other forms, possibly SO_3 or SO_4 (Schlager et al., 2006, Balzani Lööv et al., 2014). Hence, the assumption that all sulfur is emitted as SO_2 yields an underestimation of the true sulfur content in the fuel. Accordingly, this factor needs to be considered when setting the alarm threshold of the FSC. In our experiments, this uncertainty factor led to low FSC estimation results, and the deviation was generally not more than 200 ppm. This prediction is based on the fact that several measurements of some plumes were taken at particular times. Similar calculation results for FSC were obtained, but they were all less than the real value of 100–200 ppm. This is probably because not all the sulfur in the fuel is emitted as SO_2. This tendency of underestimation has also been found in previous studies (Johan, R et al. 2017).

[1] Balzani Lööv, J. M., Alfoldy, B., Gast, L. F. L., Hjorth, J., Lagler, F., Mellqvist, J., Beecken, J., Berg, N., Duyzer, J., Westrate, H., Swart, D. P. J., Berkhout, A. J. C., Jalkanen, J.-P., Prata, A. J., vander Hoff, G. R., and Borowiak, A.: Field test of available methods to measure remotely SOx and NOx emissions from ships, Atmos. Meas. Tech., 7, 2597–2613, doi:10.5194/amt-7-2597-2014, 2014.
[2] Johan, R., Conde, V., Beecken, Jörg and Ekholm, J.: Certification of an aircraft and airborne surveillance of fuel sulfur content in ships at the SECA border, CompMon (https://compmon.eu/), 2017.
[3] Schlager, H., Baumann, R., Lichtenstern, M., Petzold, A., Arnold, F., Speidel, M., Gurk, C., and Fischer, H.: Aircraft-based Trace Gas Measurements in a Primary European Ship Corridor, proceedings TAC-Conference, 83–88, 2006.

Page 7 last paragraph. To me it is not clear how errors in determination of the peak height is propagated in the total error and it is not clear how this is done. The error of 300 ppm is (it seems) related to the comparison with the on-board samples. And not from error propagation analysis as far as I can tell. It would be nice to show the error propagation numbers as well and see how well these two approaches match. In general, I think that the uncertainty discussion could be more

quantitative.

The results of the FSC are derived from the calculation of four data. Therefore, errors or incorrect selection of these four values can affect the results of the FSC. Therefore, the law of error propagation can explain the uncertainty. I have supplemented the error propagation formula of the FSC formula to illustrate this problem. Currently, the data we can obtain are FSC estimates (derived from four measurements) and FSC true values (derived from chemical validation of the fuel). Currently, only multiple measurements of the same plume or multiple peaks using the same measurement can be used to analyze its uncertainty.

Calculation uncertainty lies in selecting the background and peak values of SO_2 and CO_2. According to the law of error propagation (widely used in surveying, mapping, and statistics), the relationship between the deviation in the measurement values and that in the FSC can be obtained. The FSC calculation results are functions of independent observations $SO_{2,peak}$, $SO_{2,bkg}$, $CO_{2,peak}$, and $CO_{2,bkg}$ as in formula 1. The relationship between the observation error ($\Delta SO_{2,peak}$, $\Delta SO_{2,bkg}$, $\Delta CO_{2,peak}$, and $\Delta CO_{2,bkg}$) and function error (ΔFSC) can be approximated using the full differential of the function as follows:

$$\Delta FSC = \frac{\partial f}{\partial SO_{2,peak}}\Delta SO_{2,peak} + \frac{\partial f}{\partial SO_{2,bkg}}\Delta SO_{2,bkg} + \frac{\partial f}{\partial CO_{2,peak}}\Delta CO_{2,peak} + \frac{\partial f}{\partial CO_{2,bkg}}\Delta CO_{2,bkg} \quad (2)$$

In our study, this deviation was generally in the order of hundreds of ppm, as explained in section 4.

Page 9 line 16: "this makes the FSC value relatively larger than that of CO_2". It is not clear what is meant here.

These sentences have been rewritten.

$$FSC[\%] = \frac{S[kg]}{fuel[kg]} = \frac{SO_2[ppm]\cdot A(S)}{CO_2[ppm]\cdot A(C)} \cdot 87[\%] = 0.232\frac{\int(SO_{2,peak}-SO_{2,bkg})dt[ppb]}{\int(CO_{2,peak}-CO_{2,bkg})dt[ppm]}[\%] \quad (1)$$

As in Eq. (1), a higher SO_2 peak leads to a higher FSC estimate, while a higher CO_2 peak leads to a lower FSC estimate. As discussed in section 3.3, not all the sulfur in the fuel is emitted as SO_2, which will result in a lower estimate value. This selection allows the estimate to be relatively close to the true value.

Page 9 line 6: were synchronized is rather vague. Please explain Page 9 In general, the data treatment is unclear to me. Why are peak values taken to compare SO_2 and CO_2? Or is it the surface area? The S-content may be derived from any set of concentrations. Taking the pea area ins just of way of averaging. It seems to me now that the peak position and its height is depending on the performances of the sensors (especially response time) and the accidental position in the plume. This could lead to uncertainties especially if the peak height only is used. This should be explained better. Especially the "approach" could be elaborated more. Sometimes I am in doubt whether peak means the highest point in the concentration or the peak area.

We made the following explanation in the manuscript:

The response time of both sensors is less than 1s. Even if the sampling rates of the two sensors are set to be consistent, the two sensors cannot be completely synchronized. This makes it difficult to calculate the ratio of SO_2 and CO_2. Our approach is that the sensor sends the average measurement value of the last 10 s to the receiver at an interval of 10 s. Therefore, the interval of integration in formula (1) is 10 s. We determined that taking the mean of measurements directly or at shorter intervals leads to too many narrow peaks in one measurement process. This makes it difficult to select the peak value, and the calculation results are unstable. At the same time, the interval should not be set too long, which will make the crest very inconspicuous or too flat. Therefore, we selected 10 s as the empirical parameter value after several experiments.

Also, in the description of result:

After the measurement of plume 5, the communication module was fault when we wanted to adjust sampling rate. We consequently replaced the communication protocol "HTTP protocol" with the "TCP/IP protocol". The main changes involved adjusting the data sampling rate from 10 to 2 s to make it easier to find the peak value (the sensor sends the average measurement value of the last 10 s to the receiver at an interval of 2 s), and the sensors were consequently recalibrated by standard mixture gas.

Page 9 in general: what exactly is "selected". This should be made clear. Now it seems a bit arbitrary. Of course, full range values are not used. But what are dramatic changes? Would be useful to explain.

These sentences have been rewritten:

The global maximum values are selected as peak values for calculating the FSC.

The peak values resulting from dramatic changes (for instance, the change in CO_2 exceeded 500 ppm, or SO_2 changes by more than 500 ppb) in continuous observations are ruled out. This may be because of exhaust uncertainty.

Page 9 line 21: 300 ppm at what level??

These sentences have been rewritten:

However, the final deviation generally does not exceed 0.03% (m/m) at an FSC level of 0.04% (m/m) to 0.24% (m/m).

Page 10: Figure 5. Sometimes background values of SO_2 are 400 ppb? That is very high. Why not subtract the background? Also in plume 6 the background seems to fluctuate very much. makes interpretation of peaks uncertain. please discuss.

We have made the following discussion in the manuscript:

The background value of CO_2 in plumes 1-4 exceeded 300 ppm, but the global background CO_2 was approximately 400 ppm. Meanwhile, the background value of SO_2 exceeded 400 ppb at some time. This was due to sensor calibration, which did not affect the final result. This kind of situation did not happen again after we recalibrated the sensors by standard mixture gas. In some cases, background values seem to fluctuate very much. This is mainly because the UAV took off from the dock, where multiple ships were berthed and wind speeds were high. Therefore, we used the flight procedure given in section 3.1 to minimize this impact.

Page 11 table. Why is not a graph provided? Such as true value (x-axis) against estimated value (y-axis). Then also a correlation coefficient could be calculated. Also a good measure of quality. In general: The results section could improve in clarity if some structure was used: data treatment; FSC observed etc. For example, the issues with sampling rate etc. (page 1 top) are perhaps important but mixed here with the results. To increase clarity this could be treated separately Conclusions High precision is not reasonable to state in view of the rather large underestimations.

This suggestion is very helpful; we added the result graph of all the data.

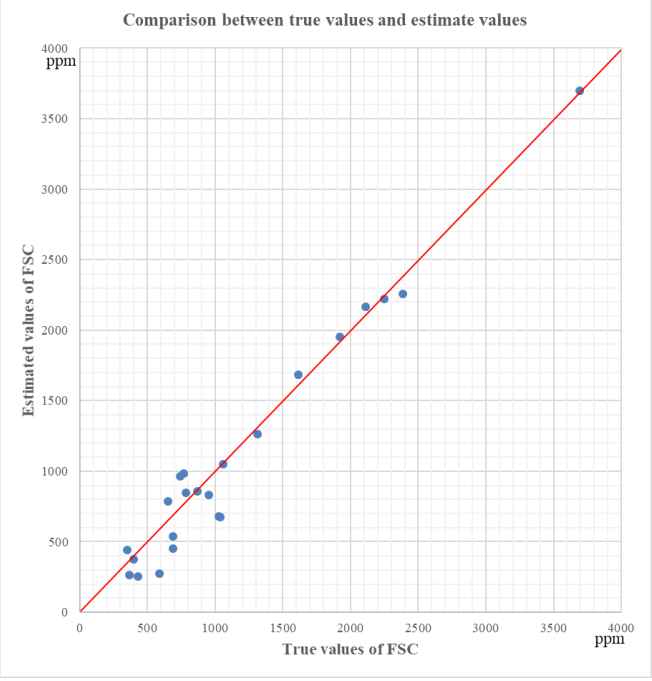

Figure 6. Comparison between the true values of FSC (x-axis) against the estimated values of FSC (y-axis) of 23 times measurement.

As shown in Fig 6, the FSC in our experiments was mainly at a level of 0.04% (m/m) to 0.24% (m/m) (only one measurement of 0.37% (m/m), not enough for reference). The deviation of the estimated FSC value calculated using the proposed method was within 0.03% (m/m), although there was some uncertainty. Considering the uncertainties listed in section 3.3, the proposed method provides accurate results. Overall, the estimated FSC is smaller than the true value in many cases. This is because 1–19% of the sulfur in the fuel is emitted in other forms, possibly SO_3 or SO_4.

In general: The results section could improve in clarity if some structure was used: data treatment; FSC observed etc. For example, the issues with sampling rate etc. (page 1 top) are perhaps important but mixed here with the results. To increase clarity this could be treated separately

The structure has been adjusted. 4.Results: 4.1 Data treatment; 4.2 FSC estimation.

Page 12: in Conclusions something might be said on the effect of SO_3 and SO_4 Specific comments:

As mentioned above, we explained it in the results. At the same time, we make the following explanation in the conclusion.

The estimated results were compared with the FSC values determined at certified laboratories. In general, the deviation of the estimated FSC value was within 0.03% (m/m) at an FSC level of 0.035% (m/m) to 0.24% (m/m). Because not all the sulfur in the fuel is emitted as SO_2, the estimated FSC is smaller than true value in many cases. Therefore, if the maritime department wants to take the estimated value as the basis for the preliminary judgment regarding whether the ship exceeds the emission standard, it needs to set an appropriate threshold and a confidence interval.

I am not a native speaker, but the English seems fine with me in general. Some specific text could be altered: - on ships the "chimney" is often called the "funnel" - "ship" is normally "vessel". - Culled is not a word that is often used Page 8 line 3: English: none of the monitored ships were fitted with exhaust cleaning equipment

The overall language of the manuscript has been enhanced; thus, any language and grammar mistakes have been corrected to the greatest extent possible.
Some words were changed as follows:

"Chimney" or "funnel": funnel.

"ship" or "vessel": In the relevant literatures, "ship" and "vessel" has both been used, "ship" seems to be used more frequently.
I believe that the use of "ship" or "vessel" generally depends on the size of the vehicle. The term "vessel" is more common and can be used for all sizes of vehicles, be they boats or ships. However, "ship" refers to seafaring vessels that are generally large enough to carry heavy loads or cargo and travel long distances. Because our research objects are mainly ocean-going ships, rather than inland river vessels. "ship" seems more appropriate.

“Culled”: Replace with “be ruled out”

“none of the monitored ships were fitted with exhaust cleaning equipment”: It has been changed.

In the end, we thank the Referee #2 for his/her positive and constructive comments.

Monitoring of compliance with fuel sulfur content regulations through UAV measurements of ship emissions

~~High-precision monitoring of compliance with fuel sulfur content through UAV measurements of ship emissions~~

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Abstract. Efficient supervision of ship emissions is currently a major concern of maritime authorities. A potential solution is the establishment of Emission Control Areas (ECAs), through which pollution from ship exhaust gas can be reduced. Nevertheless, ECAs should be strictly monitored to control ship emissions and maintain a healthy environment. In this study, an Unmanned Aerial Vehicle (UAV)-based measurement system for exhaust gas from ships was designed and developed. Waigaoqiao port in the Yangtze River Delta, an ECA in China, was selected for monitoring compliance with fuel sulfur content. Unlike in situ or airborne measurements, the proposed measurement could be used to determine the smoke plume at about 5 m from the ~~chimney~~ funnel mouth of ships, providing a means for estimating the fuel sulfur content (FSC) of ship. In order to verify the accuracy of this measurement, fuel samples were collected and sent to the laboratory for chemical examination, and these two types of measurements were compared. After more than 20 comparative experiments, the results show that, in general, ~~the deviation of the estimated value for FSC is less than 0.03%(m/m). Hence, UAV measurement can be used for high-precision monitoring of ECAs for compliance with FSC.~~the deviation of the estimated value for FSC is less than 0.03% (m/m) at an FSC level ranging from 0.035% (m/m) to 0.24% (m/m). Hence, UAV measurements can be used for monitoring of ECAs for compliance with FSC regulations.

1. Introduction

With the rapid development of international shipping in recent years, air pollution caused by ship emissions has become serious. ~~International shipping is responsible for approximately 5 to 8% of global anthropogenic SO_2 emissions (Eyring et al., 2005). SO_2 can cause severe health and environmental problems, is important in atmospheric chemistry studies as a principal air pollutant.~~ Estimations show that ships contribute 4-9% of global SO_2 emissions and 15% of NO_x (Eyring et al., 2010). According to the United Nations Conference on Trade And Development (UNCTAD, 2017), the volume of the world's seaborne trade grew by 66% between 2000 and 2015. As global commerce expands, ocean-going ships consume more fuels, generally low-quality residual fuels containing high concentrations of sulfur and heavy metals (Lack et al., 2011). From the viewpoint of spatial distribution, the highest emissions of SO_2 per unit area occur in the eastern and southern China seas, sea

areas in south-eastern and southern Asia, Red Sea, Mediterranean Sea, North Atlantic near the European coast, Gulf of Mexico and Caribbean Sea, and along the western coast of North America. (Johansson et al., 2017). Ship-emitted pollutants influence air quality, human health, and climate. They not only affect the air quality in coastal areas but even influence the inland areas hundreds of kilometers away from the emission sources (Liu et al., 2016).

In order to limit hazards caused by ship emissions, the International Maritime Organization (IMO) extended the MARPOL 73/78 International Convention for the Preventions for Pollution of Air Pollution from Ship (MARPOL, 1997). In 2005, some regulations went into effect after being received by appropriate laws of the signatory states (at the European level it was received with the directives 1999/32/EC, 1999, and 2005/33/EC, 2005), and introduces limits to marine fuel sulfur content and engine performance to reduce SO_x and NO_x emissions. Further amendments to Annex VI were adopted in 2008 and entered into force in 2010. Fuel sulfur content (FSC) is normally given in units of percent sulfur content by mass; in the following written as % (m/m). Following the IMO regulation, the global cap for FSC in marine fuel was set in 2012 at 3.5% (m/m), and it will be reduced to 0.5% (m/m) by 2020. In addition, the IMO provides for the establishment of Emission Control Areas (ECAs) to control ship emissions, where there are more stringent controls on ship emissions. At present, the Baltic Sea, the North Sea, the North American area, and the United States Caribbean Sea are designated as ECAs (IMO, 2017). The FSC limit must not exceed 0.1% (m/m) beginning in 2015.

China is one of the world's busiest and fastest-growing shipping regions. In 2016, China accounted for seven of the world's top 10 ports and 11 of the top 20. ~~To cope with the current international situation of energy conservation and emission reduction~~ In order to reduce the air pollution caused by ship emissions, the Atmospheric Pollution Prevention and Control Law of the People's Republic of China was promulgated in 2015 (Standing Committee of the National People's Congress, 2015). Three domestic emission control areas (DECA) were set up, which include the Yangtze River Delta, the Pearl River Delta, and Bohai Rim (Beijing-Tianjin-Hebei Region). The current stage of the plan requires that the FSC does not exceed 0.5% (m/m).

With the above regulations in place, the main question remains on how to efficiently verify compliance of ships in the ECAs with the regulation. At present, the most accurate method for checking compliance is to collect fuel samples from ships at berth by state port control authorities, and then analyze the samples at certified laboratories or by portable detectors. However, it is time consuming and few ships are effectively controlled. Another problem is that sailing ships within the ECAs cannot be checked.

Several studies have suggested inferring FSC by monitoring ship emissions, and then identifying ships with excessive FSC. According to the available literature, these approaches include optical methods (LIDAR (Fan et al., 2018), Differential Optical Absorption Spectroscopy (DOAS) (Seyler et al., 2017), UV camera (Prata, 2014)) or “sniffer” methods (Balzani Lööv et al., 2014, Beecken et al., 2015). Optical methods analyze variations in light properties after interactions with the exhaust plume, and the local wind field before determining the SO_2 emission rate is observed. The simultaneous measurement of CO_2 and SO_2 emissions on a routine basis is unrealistic at present. Thus, the amount of fuel burned at the time of measurement is unknown and has to be estimated via modeling for calculating the FSC. For instance, the model

STEAM (ship traffic emission assessment model), developed by the Finnish Meteorological Institute (Jalkanen et al., 2009) was used in the research for estimating FSC by Balzani Lööv et al. (2014). In addition, using the ratio of SO_2 and NO_2 measured via DOAS in the ship plume can be used as an indicator of FSC (Johan, R et al. 2017, Cheng, Y et al, 2019). The advantage of the optical method is that it can detect ship emissions at a long distance (thousands of meters away), but it is limited in that it can only distinguish between a high FSC (>1% (m/m)) and a low FSC (<1% (m/m)) (Johan et al., 2017). ~~The "sniffing" method is based on simultaneous measurement of the elevated concentrations of SO_2 and CO_2 in the exhaust plume from the target ship and comparing them with the background. In the process, SO_2 and CO_2 concentrations are measured by IR radiometer and UV fluorescence instruments in an airflow provided through a probe. The advantage of the "sniffing" method is that it offers measurement accuracy between 0.1–0.2%(m/m) FSC, which can be further increased up to 0.05–0.1%(m/m) FSC if combined with an additional NO_x sensor. However, the instrument must be placed in the plume exhausted by the target ship to obtain accurate results (Johan et al., 2017).~~ The "sniffing" method is based on simultaneous measurement of elevated SO_2 and CO_2 concentrations in the exhaust plume from the target ship and comparing them with the background. The measurement of CO_2 allows for relating the measurement of SO_2 to the amount of fuel burned at a given time, thus enabling the calculation of FSC directly. The concentration of SO_2 in plumes was generally measured using UV fluorescence sensors, and CO_2 was measured using a non-dispersive infrared analyzer (NDIR) or cavity ring down spectrometer (CRDS). The advantage of the "sniffing" method is that it offers more accuracy estimation for FSC. However, the instrument must be placed in the plume exhausted by the target ship. In some studies (Van Roy and Scheldeman, 2016a, 2016b), the "sniffing" method offers a measurement accuracy between 0.1–0.2% (m/m) FSC, which can be further increased up to 0.05–0.1% (m/m) FSC if combined with an additional NO_x sensor. This is because the response of SO_2 analyzers (fluorescence) has cross sensitivity to NO. Deviations are not the same at different FSC levels, with an estimated relative uncertainty of 20% (m/m) for ships with 1% (m/m) FSC and a relative uncertainty of 50–100% at 0.1% (m/m) FSC. Balzani Lööv et al. (2014) obtained the following FSC measurements based on the "sniffer" principle: 0.86±0.23% (m/m) from land, 1.2±0.15% (m/m) from an on-board stack, and 1.13±0.18% (m/m) from a mobile platform. There was a 6% relative uncertainty for an FSC of 1% (m/m) but a 60% relative uncertainty for an FSC of 0.1% (m/m).

Ship emission measurements can be divided into land-based (Kattner et al., 2015, Yang et al., 2016), marine-based (Cappa et al., 2014), airborne-based (Beecken et al., 2014, Aliabadi et al., 2016), ~~and satellite-based (Ding et al., 2018)~~ satellite-based (Ding et al., 2018) and Unmanned Aerial Vehicle (UAV)-based (Villa et al., 2019) according to different platforms. Land-based measurements provide continuous observation but are greatly affected by wind speed, wind direction, and the distance between the ship and equipment. Marine-based measurements are suitable for studying the discharge from individual ships. The monitoring equipment is generally installed and used by research institutions or ship owners. This is not subjected to FSC inspection by government regulatory authorities. Airborne-based measurements can approach ship plumes and collect exhaust from the target ship. Satellite-based measurements are suitable for large-scale observation and mainly used to observe the NO_x emissions of ships. UAV-based measurements have gradually increased in the research regarding the atmosphere (Malaver Rojas et al., 2015, Mori et al., 2016). However, to date, there are relatively few applications of these

measurements in ship emissions. As such, the most suitable approach for monitoring compliance is to employ "sniffer" measurements taken by airborne. However, the cost of airborne platforms is high, and it requires active sampling of ship exhaust plumes at low altitude. The closer the detector is to the ship's plume, the more accurate the results. However, safety risks are also relatively high near the plume. Optical measurements and "sniffer" measurements of gases in the exhaust plume of ships and more details on such measurements can be found in several related papers (Balzani Lööv et al., 2014, Van Roy and Scheldeman, 2016a, 2016b, Johan et al., 2017).

Based on the experience from those studies, we established ~~"mini sniffer"~~ sensors mounted on a UAV to measure the concentrations of SO_2 and CO_2 in order to calculate the FSC. The UAV can collect samples closer to the exhaust than airborne-based measurements. Waigaoqiao port in the Yangtze River Delta was selected as the study site. Using this measurement, we analyzed more than 20 ship plumes and compared the results with the FSC of entering ships determined from fuel samples analyzed at certified laboratories. Through these experiments, we investigated and analyzed the emission process of SO_2 and CO_2 very close to the ~~smoke stack~~funnel mouth of ships and design ~~a high precision~~an accurate measurement of FSC.

2. Measurement

2.1 UAV

~~**Figure 1. Image of the modified UAV platform. The black box installed under the UAV is a pod carrying sensors, a camera, communication modules, and a gas pump (to collect the ship's exhaust).**~~

Figure 1. Image of the modified UAV platform. The black box installed under the UAV is a pod which was designed and customized by us. It carries a gas pump (to collect the ship's exhaust), gas circuit, a filter (to remove water vapor), sensors for SO_2 and CO_2, a small motor (to provide energy for pumping), a camera, and communication modules.

Figure 2. UAV platform flying close to the smoke stack for collecting exhaust gas in the automatic engine room laboratory of Shanghai Maritime University.

In the experiment, we used the MATRICE 600 UAV (SZ DJI Technology Co., Ltd.), and modified it. We designed and customized a special pod, which was installed underneath the UAV, to carry sensors, communication circuit boards, gas circuit systems, and other modules, as shown in Fig.1. After the successful assembly of the UAV platform, we first carried out preliminary experiments in the automatic engine room laboratory of Shanghai Maritime University. Through the preliminary test, we verified the stability and security of the whole UAV system. At the same time, it also allowed the UAV operator to practice how to operate the UAV for sampling close to the smoke stack. Fig.2 shows a photograph of the process of collecting exhaust gas from near the smoke stack. The UAV can fly near the smoke for the collection and detection of exhaust gas. The detection information can be sent to the receiving end in real time. Table 1 presents the parameters of the UAV. The weight of the pod is about 3 kg and the UAV can fly for about 25 min. Therefore, measurements can be taken from 1–2 ships using one set of batteries.

Table 1. Parameters of the UAV

Parameter	Value
Symmetrical motor wheelbase	1133 mm
Size	1668 mm × 1518 mm × 727 mm
Weight	9.5 kg
Recommended maximum take-off weight	15.5 kg
Hovering accuracy(P-GPS)	Vertical: ±0.5 m, Horizontal: ±1.5 m
Maximum rotational angular velocity	pitch axis: 300°/s, Heading axis: 150°/s
Maximum pitch Angle	25°
Maximum rising speed	5 m/s

Maximum rate of descent	3 m/s
Maximum sustained wind speed	8 m/s
Maximum horizontal flight speed	65 km/h (no wind environment)
Hover time	non-loaded:32 min, load 6 kg:16 min

2.2 ~~"Mini-sniffer" s~~Sensors

In the measurement process, the ship exhaust is pumped into the pod by the gas pump. After the filter removes the water vapor, the sensors react and the communication module sends the measurement results to the receiving end. The sensors included instrumentation for both SO_2 and CO_2 measurements. These sensors were purchased from HH Feuerungstechnik GmbH, Germany.

~~The "mini-sniffer" sensors included instrumentation for both SO_2 and CO_2 measurements.~~ For ~~the former~~SO_2, the sensor ~~was~~ is based on the electrochemical~~electrochemistry~~ method. An electrochemical sensor determines the concentration of a gas via a redox reaction, producing an electrical signal proportional to the concentration of the gas. In previous measurements of ship exhaust gas, SO_2 sensors are mainly based on the UV-fluorescence method (Balzani et al., 2014, Beecken et al., 2014, Kattner et al., 2015, Johan et al., 2017), which is not appropriate for the UAV due to weight limitations. The SO_2 electrochemical sensor has the advantages of low power consumption, small size, light weight, and high precision. In addition, the sensor is capable of measuring SO_2 at a low ppb range (Hodgson et al., 1999). Therefore, we used the electrochemical sensor to measure SO_2 concentration. The measuring range of the sensor is 0–5 ppm, the resolution level is 0.001 ppm, response time (t_{90}) is less than 1 s, and the accuracy is ±~~5%~~0.25 ppm~~ full scale~~. t_{90} is defined as the time it takes to reach 90% of the stable response after a step change in the sample concentration.

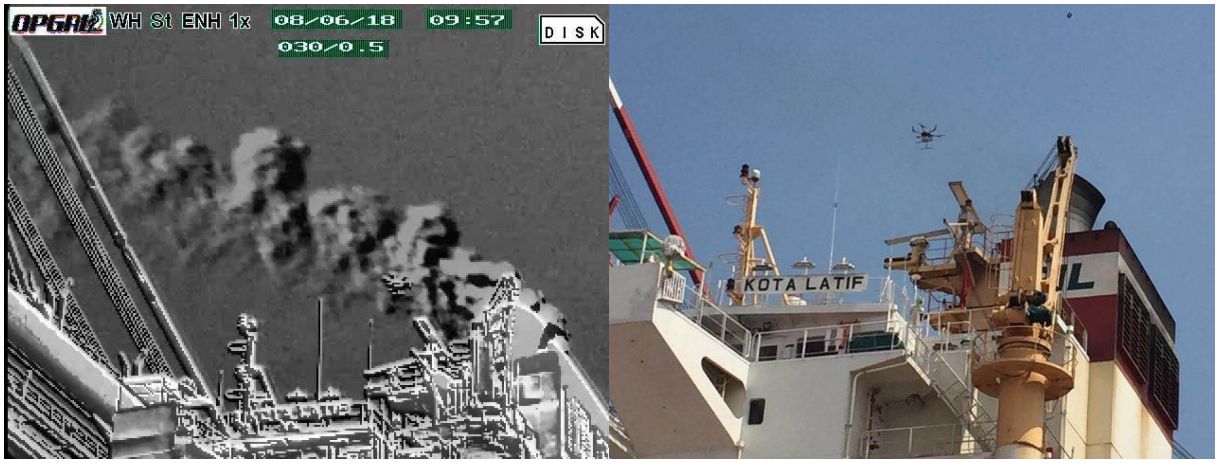
For CO_2, the sensor is based on the non-dispersive infrared analyzer method. This type of sensor is often used to measure the CO_2 concentration of ship exhaust gas (Balzani et al., 2014, Beecken et al., 2014, Kattner et al., 2015, Johan et al., 2017). An infrared beam passes through the sampling chamber, and each gas component in the sample absorbs infrared rays at a specific frequency. The concentration of the gas component is determined by measuring the infrared absorption at the corresponding frequency. The measuring range of the used sensor is 0–5000 ppm, resolution level is 1 ppm, response time (t_{90}) is less than 1 s, and its accuracy is ±50 ppm~~1% full scale~~.

Sensor calibration is required when the equipment is used daily. The time interval for sensor calibration is three months or when the accumulated working time of the sensor exceeds 180 h. If either of these conditions is met, calibration will be carried out.~~Sensor calibration is required before the equipment is put into daily use. It is typically calibrated three months or 180 working hours apart.~~ The zero and full scales are usually calibrated by standard mixture gas. Before each mission, sensors are activated and residual gas in the airway is discharged by the gas pump.

3. Methods

3.1 Flight procedures

(a) (b)



(c) (d)

Figure 3. Photographs showing the setup of the experiment. An infrared camera is set up for locating the smoke plume (a), (b). The target plume is imaged by the infrared camera (c). The UAV takes off towards the smoke plume (d).

The preliminary positioning measurements of the ship smoke plume are as shown in Fig. 3. The UAV platform with ~~"mini-sniffer"~~ sensors flew close to the funnel of ship ~~smoke stack~~, hovered for collecting exhaust gas, and then detection

information was sent back. This procedure is not without risk and a well-considered flight approach is recommendable. We ~~summarise~~ summarize the experiment steps as follows:

1. Determine the position of the plume according to the wind speed, wind direction, height gauge, infrared camera, and other factors.

2. Check the equipment: the power is enough, the GPS signal is normal (it is recommended that the number of satellites is more than 13), the electrochemical sensor is activated, and the residual gas is discharged in the air path of the pod.

3. The UAV takes off vertically and rises to an altitude of 100 m (the first measurement point) for 3 min to determine the background value of SO_2 and CO_2.

4. Fly the UAV towards the plume and hover to collect exhaust gas from about 10 m (the second measurement point) and 5 m (the third measurement point) away from the ~~chimney~~ funnel for 5 min, respectively.

5. Lift the UAV and then return it to the starting point.

During the process, real-time observations of SO_2 and CO_2 were sent to receiving end. The operator adjusted the UAV's position according to the observations to keep the sensor in the plume. Therefore, in general, the UAV confirmed the approximate location of the plume at a distance of 10 m, and then gradually approached the location of about 5 m for collection.

3.2 Calculation of FSC

When the UAV flied into the ship plume, the peak areas of the SO_2 and CO_2 measurements were determined, and the background was subtracted. The background value of SO_2 and CO_2 can be obtained when the UAV hovers at the first measurement point. The peak values of SO_2 and CO_2 are determined when the UAV hovers at the second measurement point or the third measurement point (main observation point). In the calculation, the molecular weights of carbon and sulfur are 12 g mol^{-1} and 32 g mol^{-1}, respectively, and the carbon mass percent in the fuel is 87±1.5% (Cooper et al., 2003). With the assumption that 100% of the sulfur and carbon contents of the fuel are emitted as SO_2 and CO_2, respectively, the FSC mass percent can be expressed as follows:

$$FSC[\%] = \frac{S[kg]}{fuel[kg]} = \frac{SO_2[ppm]\cdot A(S)}{CO_2[ppm]\cdot A(C)} \cdot 87[\%] = 0.232\,\frac{\int(SO_{2,peak}-SO_{2,bkg})dt[ppb]}{\int(CO_{2,peak}-CO_{2,bkg})dt[ppm]}[\%] \quad (1)$$

where *A(S)* is the atomic weight of sulfur and *A(C)* the atomic weight of carbon. $SO_{2,peak}$, $SO_{2,bkg}$, $CO_{2,peak}$, and $CO_{2,bkg}$ are the peak and background values of SO_2 and CO_2, respectively. ~~The time interval of the integral is 10 s in our experiment.~~ This calculation method is consistent with that described in the MEPC guidelines 184(59) and previous studies (Beecken et al., 2014, Kattner et al., 2015, Johan et al., 2017).

The response time of both sensors is less than 1s. Even if the sampling rates of the two sensors are set to be consistent, the two sensors cannot be completely synchronized. This makes it difficult to calculate the ratio of SO_2 and CO_2. Our approach is that the sensor sends the average measurement value of the last 10 s to the receiver at an interval of 10 s. Therefore, the interval of integration in Eq. (1) is 10 s. We determined that taking the mean of measurements directly or at shorter intervals

leads to too many narrow peaks in one measurement process. This makes it difficult to select the peak value, and the calculation results are unstable. At the same time, the interval should not be set too long, which will make the crest very inconspicuous or too flat. Therefore, we selected 10 s as the empirical parameter value after several experiments.

3.3 Uncertainties

Because measurements taken inside the ship plumes are analyzed relative to the background, offset errors can be neglected. Nevertheless, there are certain uncertainties in the estimation process of the FSC. They can be summed up as sensor uncertainty, exhaust uncertainty, measurement uncertainty, calculation uncertainty, and so on.

As for sensor uncertainty, the linear error is negligible and the nonlinearity of the two ~~"mini-sniffer"~~ sensors should be no more than ±1%. It can be corrected through frequent calibrations with standard gases and gradually establishing a quality management system comprising sensor linearity, sensitivity, repeatability, hysteresis, resolution, stability, drift, and other attributes of the minimum requirements.

Exhaust uncertainty arises because not all the sulfur in the fuel is emitted as SO_2. Preliminary studies showed that 1-19% of the sulfur in the fuel is emitted in other forms, possibly SO_3 or SO_4 (Schlager et al., 2006, Balzani Lööv et al., 2014). Hence, the assumption that all sulfur is emitted as SO_2 yields an underestimation of the true sulfur content in the fuel. Accordingly, this factor needs to be considered when setting the alarm threshold of the FSC. In our experiments, this uncertainty factor led to low FSC estimation results, and the deviation was generally not more than 200 ppm. This prediction is based on the fact that several measurements of some plumes were taken at particular times. Similar calculation results for FSC were obtained, but they were all less than the real value of 100–200 ppm. This is probably because not all the sulfur in the fuel is emitted as SO_2. This tendency of underestimation has also been found in previous studies (Johan, R et al. 2017).

Measurement uncertainty is mainly attributable to inadequate sampling (the UAV did not fly into the plume). Moreover, shipborne antennae, dock facilities, and strong winds may cause interference in finding an appropriate sampling point and even lead to sampling failure. This uncertainty factor can lead to an incorrect estimation of the FSC. Therefore, we formulated the flight procedures as described in section 3.1.

Calculation uncertainty lies in selecting the background and peak values of SO_2 and CO_2. According to the law of error propagation (widely used in surveying, mapping, and statistics), the relationship between the deviation in the measurement values and that in the FSC can be obtained. The FSC calculation results are functions of independent observations $SO_{2,peak}$, $SO_{2,bkg}$, $CO_{2,peak}$, and $CO_{2,bkg}$ as in Eq. (1). The relationship between the observation error ($\Delta SO_{2,peak}$, $\Delta SO_{2,bkg}$, $\Delta CO_{2,peak}$, and $\Delta CO_{2,bkg}$) and function error (ΔFSC) can be approximated using the full differential of the function as follows:

$$\Delta FSC = \frac{\partial f}{\partial SO_{2,peak}}\Delta SO_{2,peak} + \frac{\partial f}{\partial SO_{2,bkg}}\Delta SO_{2,bkg} + \frac{\partial f}{\partial CO_{2,peak}}\Delta CO_{2,peak} + \frac{\partial f}{\partial CO_{2,bkg}}\Delta CO_{2,bkg} \quad (2)$$

In our study, this deviation was generally in the order of hundreds of ppm, as explained in section 4.

In any case, these uncertainties will occur during the measurement process. After the establishment of flight procedures as mentioned in section 3.1 and selection process as in section 4, we observed that the deviation between the estimated value of

FSC and true value of FSC was generally not more than 300ppm. In addition, none of~~all~~ the monitored ships ~~are~~ were ~~not~~ fitted with exhaust cleaning equipment.

4. Results

4.1 Data treatment

(a) (b)

Figure 4. Photographs showing the flight of the UAV during measurements. The UAV platform was flown close to the funnel of ship for collecting exhaust gas and detection at Waigaoqiao pier.

Figure 4 shows the UAV platform with ~~"mini sniffer"~~ sensors flying close to the ship smoke. It hovered to collect exhaust gas, and detection information was subsequently sent back. Generally, changes in SO_2 and CO_2 observations can be divided into three stages: (1) The UAV took off and approached the ship ~~chimney~~ funnel for about 3 min. The SO_2 and CO_2 observations were relatively low, and the background value was obtained in this stage. (2) The UAV was gradually flown to the plume centre, and data were collected. Rapid increases in SO_2 and CO_2 concentrations, reaching their peaks, were observed, which took approximately 10–15 min. The peak data were obtained in this stage. (3) The UAV completed the gas collection and returned, which took about 5 min. Decreased SO_2 and CO_2 concentrations relative to the observation when the UAV was in the plume centre were observed. Observed SO_2 and CO_2 values returned to background levels, but they were not used as background values. Residual gas in the airway needed to be discharged by the gas pump before the next collection.

Numerous measurements have been made in the Waigaoqiao wharf since January 2018. After the adjustment of various technical parameters and the accumulation of UAV flight experience, this method could provide accurate results. From August 2018 to January 2019, more than 20 plumes exhausted by ships have been detected. Fuel samples, which are

considered as the true value of FSC, were taken and sent for laboratory chemical examination. Finally, the results of the UAV method were compared with those of the laboratory tests.

According to Eq. (1), if the observations of SO_2 and CO_2 values simultaneously reach their peaks, it is easier to select the background and peak value for calculating the FSC. However, the actual data collected are sometimes not ideal, and there is calculation uncertainty when selecting the background and peak values of SO_2 and CO_2. In previous studies, to the procedures for selecting background and peak values are not discussed in detail. As the number of experiments increased, we gradually developed a selection process. In our experiment, observations of SO_2 and CO_2 in the receiving end were synchronized. Therefore, the background and peak values for SO_2 and CO_2 that we selected to calculate the FSC were observed at the same time point.

According to the flight record, the minimum values of SO_2 and CO_2 collected at the first measurement point are selected as the background values. There is generally greater uncertainty in selecting the peak values. The synchronous, stable, obvious, and maximal values in observations of SO_2 and CO_2 are selected as the peak values. The selection method is as follows:

1. The peak values in the observations of SO_2 and CO_2 are determined at the second and third measurement points, respectively.

2. The peak values at the full range of the SO_2 or CO_2 sensors are ~~culled~~ruled out.

3. The peak values resulting from dramatic changes (for instance, the change in CO_2 exceeded 500 ppm, or SO_2 changes by more than 500 ppb) in continuous observations are ruled out. ~~in observations are culled.~~ This may be because of exhaust uncertainty.

4. The occurrence time of peak values in SO_2 and CO_2 are compared, and then the simultaneous peaks and almost simultaneous peaks (no more 20 s) are retained. If there is a small deviation between the time point of the peak values for SO_2 and CO_2, we select the time point at peak of SO_2. This will make the FSC value relatively larger than that of CO_2. As in Eq. (1), a higher SO_2 peak leads to a higher FSC estimate, while a higher CO_2 peak leads to a lower FSC estimate. As discussed in section 3.3, not all the sulfur in the fuel is emitted as SO_2, which will result in a lower estimate value. This selection allows the estimate to be relatively close to the true value.

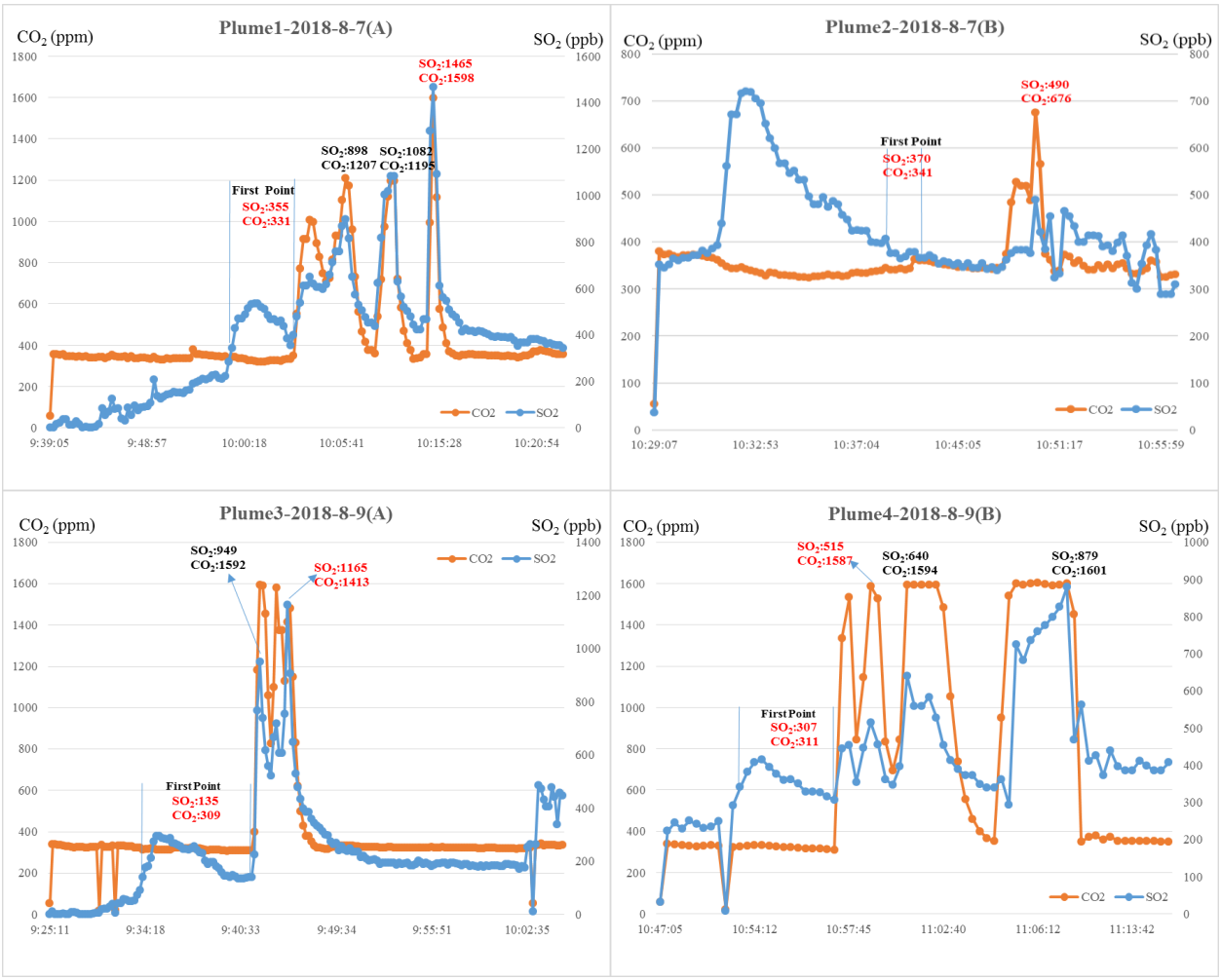
~~This will make the FSC value relatively larger than that of CO_2, the reason for this is the exhaust uncertainty discussed in section 3.3 (not all the sulfur in the fuel is emitted as SO_2).~~

5. After the above filtration, approximately 1-4 time points will be left as the selection points for peak values. The global maximum values are selected as peak values for calculating the FSC.

~~4~~.2 FSC estimation

In our experience, using the above method can provide the FSC value that is closest to the real value in most cases. In a few cases, it may be suboptimal rather than optimal. However, the final deviation generally does not exceed 0.03% (m/m) at an FSC level of 0.035% (m/m) to 0.24% (m/m). To illustrate this selection method, six typical sets of plume measurement data for SO_2 and CO_2, marked as plumes 1–6, along with the time and serial number, are shown in Fig. 5.

~~In our experience, using the above method can provide the FSC value that is closest to the real value in most cases. In a few cases, it may be suboptimal rather than optimal. However, the final deviation generally does not exceed 300ppm. Six typical sets of plume measurement data for SO_2 and CO_2, marked as plumes 1–6, along with the time and serial number, are shown in Fig. 5.~~



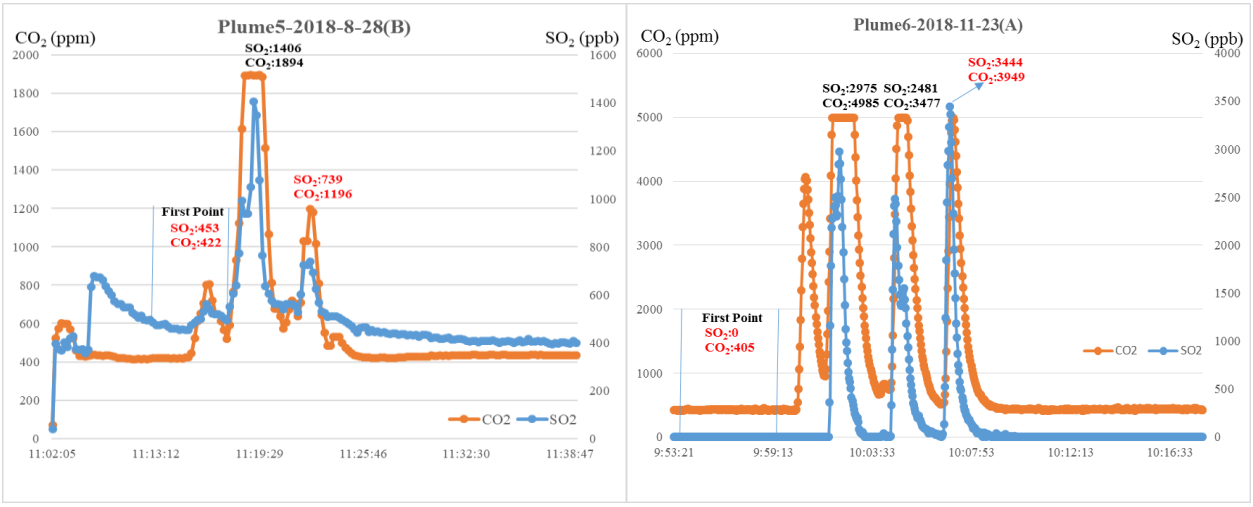


Figure 5. Six sets of plume measurement data for SO_2 and CO_2, marked as plumes 1–6, along with the time and serial number. The background and peak values of SO_2 and CO_2 were used to estimate the FSC. In each plume, the time range of the first monitoring point is marked by two vertical lines. The selected background and peak values of SO_2 and CO_2 are written in red and alternative peak values are written in black.

As shown in Fig. 5, the observations of plumes 1 and 3 simultaneously reached the peak value. However, these were multiple SO_2 and CO_2 peak values, and the global maximum peak values of SO_2 and CO_2 were selected. In plume 2, there was a peak for SO_2 at 10:32, but there was none for CO_2 at the same time. We used the data from the simultaneous peaks of SO_2 and CO_2 for the calculations. The observations of plumes 4 and 5 also simultaneously reached the peak value at multiple time points. However, at 11:02 and 11:07 in plumes 4 and 11:19 in plume 5, the SO_2 measurements reached the peak values, but the CO_2 measurements reached plateau levels above which they did not increase any further. Therefore, the data in this period were not used as peak values of the plumes. In plume 6, CO_2 measurements did not increase any further owing to the full range of the CO_2 sensor at 10:02 and 10:04. This happens in rare cases when the UAV is too close to the ~~chimney~~ funnel (less than 5 m), and these data cannot be used as peak values. After the measurement of plume 5, the communication module was fault when we wanted to adjust sampling rate. We consequently replaced the communication protocol “HTTP protocol” with the “TCP/IP protocol”. The main changes involved adjusting the data sampling rate from 10 to 2 s to make it easier to find the peak value (the sensor sends the average measurement value of the last 10 s to the receiver at an interval of 2 s), and the sensors ~~was~~ were consequently recalibrated by standard mixture gas. Therefore, the background values of plumes 1–5 were not the same as those of plume 6. Nonetheless, Eq. (1) was used to calculate the interpolation ratio, and it therefore does not affect the final calculation results. In addition, when the FSC of the target ship is low, for example, when the fuel used is light diesel fuel, the SO_2 observation values were mostly 0. When this happened, according to our experience, the FSC was generally lower than 200 ppm, and the ship was likely to meet the emission requirements.

Table 2 Comparison and verification of the estimated and true values of FSC. We present the selected background and peak values of SO_2 and CO_2 and alternative peak values (mentioned in Figure 5). The FSC results and deviations of these different values are also listed for comparison purposes. They are distinguished as follows in the column titled "Selected": the selected peak values are marked “√” indicates the selected peak values, and “×” indicates alternative peak values (which is not selected as the calculated value in the final result of FSC).

ID	Plume ID	Selected	SO_2 (ppb)		CO_2 (ppm)		Estimated value of FSC (ppm)	True value of FSC (ppm)	Deviation (ppm)
			Bkg	Peak	Bkg	Peak			
1		√		1465		1598	2033		110
2	Plume1	×	355	1082	331	1195	1952	1923	29
3		×		898		1207	1438		-485
4	Plume2	√	370	490	341	676	831	954	-123
5	Plume3	×	135	949	309	1592	1472	2113	-641
6		√		1165		1413	2164		51
7		√		515		1587	378		-18
8	Plume4	×	307	640	311	1594	602	396	206
9		×		879		1601	1029		633
9	Plume5	√	453	739	422	1196	857	868	-11
10		×		1406		1894	1502		634
11		√		3444		3949	2255		-132
12	Plume6	×	0	2481	405	3477	1874	2387	-513
13		×		2975		4985	1507		-880

The background and peak values of SO_2 and CO_2 were selected from plumes 1–6, and the FSC was calculated according to Eq. (1). The comparison results of the estimated FSC values are presented in Table 2. The background value of CO_2 in plumes 1-4 exceeded 300 ppm, but the global background CO_2 was approximately 400 ppm. Meanwhile, the background value of SO_2 exceeded 400 ppb at some time. This was due to sensor calibration, which did not affect the final result. This kind of situation did not happen again after we recalibrated the sensors by standard mixture gas. In some cases, background values seem to fluctuate very much. This is mainly because the UAV took off from the dock, where multiple ships were berthed and wind speeds were high. Therefore, we used the flight procedure given in section 3.1 to minimize this impact. By comparing the results and deviations of the different calculated values, it can be seen that appropriately selecting the peak value is important. In general, the optimal value can be selected using the selection method with the exception of plume 1. However, the deviation is not large.

~~This was due to sensor calibration, which did not affect the final result. By comparing the results and deviations of the different calculated values, it can be seen that appropriately selecting the peak value is important. In general, the optimal value can be selected using the selection method with the exception of plume 1. However, the deviation is not large. In our experiment with more than 20 plumes, the deviation of the estimated FSC value calculated using the proposed method was within 300 ppm (0.03% (m/m)), although there was some uncertainty. Considering the uncertainties listed in section 3.3, the proposed method provides accurate results.~~As shown in Fig 6, the FSC in our experiments was mainly at a level of 0.035% (m/m) to 0.24% (m/m) (only one measurement of 0.37% (m/m), not enough for reference). The deviation of the estimated

FSC value calculated using the proposed method was within 300 ppm (0.03% (m/m)), although there was some uncertainty. Considering the uncertainties listed in section 3.3, the proposed method provides accurate results. Overall, the estimated FSC is smaller than the true value in many cases. This is because 1–19% of the sulfur in the fuel is emitted in other forms, possibly SO_3 or SO_4.

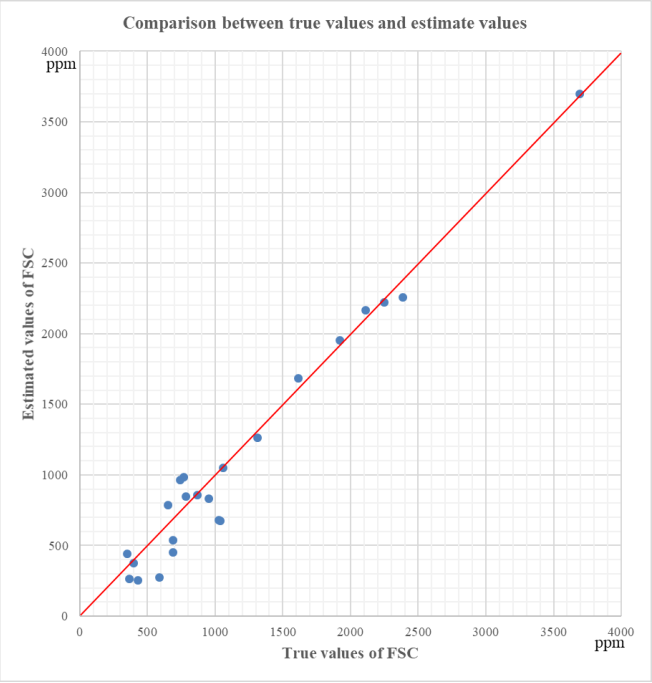


Figure 6. Comparison between the true values of FSC (x-axis) against the estimated values of FSC (y-axis) of 23 times measurement.

5. Conclusions

In this study, we performed close monitoring of ship smoke plumes using UAV. Observation data of SO_2 and CO_2 were collected at close range (5–10 m) of ship funnel ~~chimney~~ mouths. The estimated results were compared with the FSC values determined at certified laboratories. In general, the deviation of the estimated FSC value was within 0.03% (m/m) at an FSC level of 0.035% (m/m) to 0.24% (m/m). Because not all the sulfur in the fuel is emitted as SO_2, the estimated FSC is smaller than true value in many cases. Therefore, if the maritime department wants to take the estimated value as the basis for the preliminary judgment regarding whether the ship exceeds the emission standard, it needs to set an appropriate threshold and a confidence interval.

~~, and they were used to estimate the FSC with high precision. The estimated results were compared with the FSC value determined at certified laboratories. In general, the deviation of the estimated FSC value was within 300 ppm (0.03% (m/m)).~~

At present, the FSC limit in China's emission control requirements is 0.5% (m/m), and that for ECAs is 0.1% (m/m). This study makes a significant contribution to the literature because the proposed method can be used for ~~high-precision and rapid~~ monitoring of ECAs for compliance with FSC standards. However, after more than one year of testing and experiment, we found that there are still many issues that remain to be resolved:

1. In about 10% of the cases, the UAV did not measure the effective background value and peak value. This is mainly caused by the UAV missing the plume during its flight. Therefore, effective methods for finding and navigating to plumes using real-time sensor feeds need to be explored.

2. In about 10% of the cases, the absolute error was more than 0.03% (m/m)~~300 ppm~~, and even more than 0.05% (m/m)~~500 ppm~~ in rare cases. Unstable concentrations of SO_2 or CO_2 in the atmosphere just before the measurement may cause such errors. Furthermore, uncertainties, such as sensor uncertainty, exhaust uncertainty, measurement uncertainty, and calculation uncertainty, may hinder accurate measurement.

3. Limited by the battery life, each flight could only last about 30 min. Therefore, after measuring 1−2 ships, the UAV was required to return for battery replacement. Nevertheless, we believe that with the development of battery technology and the improvement of lightweight sensors, the battery life will be extended much further.

Data availability. Requests for data sets and materials please address to Fan Zhou (fanzhou_cv@163.com).

Author contributions. FZ designed the study, analyzed the experimental data and wrote the article. SP, WC and XN contributed the experiments. BA provided constructive comments on this research.

Competing interests. The authors declare that they have no conflict of interest.

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