

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 authors did lots of efforts to correct the manuscript and most of the reviews' comments have been addressed. However, I have a serious concern about the calculation of the fuel sulfur content. They keep calculating the FSC from peak values instead of calculating the ratios of the areas under the S and CO₂ time plot. This input a huge uncertainty in the method. I am, as an experimentalist, not fully convinced that I could believe in those ratios even if the calculated FSC is close to the laboratory given value.

For this reason, I can't recommend the paper for publication until the authors can prove that the peak value calculation is correct, for example, all the peak values in one measurement give the same FSC with an acceptable statistic.

Yes, I agree with this viewpoint. Our approach is exactly to use "the ratios of the areas" rather than "peak values". As is written in the manuscripts:

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

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 SO₂ and CO₂. 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.

$\int(SO_{2,peak} - SO_{2,bkg})dt [ppb]$ are just the area under the S time plot, and the interval of integration is 10 s.

Then, formula (1) can be rewritten as

$$FSC[\%] = 0.232 \frac{\int(SO_{2,peak} - SO_{2,bkg})dt[ppb] / 10}{\int(CO_{2,peak} - CO_{2,bkg})dt[ppm] / 10} [\%] \approx 0.232 \frac{AVG(SO_{2,peak}) - AVG(SO_{2,bkg})}{AVG(CO_{2,peak}) - AVG(CO_{2,bkg})} [\%] \quad (2)$$

AVG () is the calculate function for average measurement value. Therefore, the data in the manuscript are the average value of measurement in 10s. Through formula (2), it is just "the ratios of the areas under the S and CO₂ time plot".

In fact, we have also proved this viewpoint in the experiment. We increased the sampling rate to 1 s and send the original value of measurement to the receiver in the second-generation pod (as shown in Figure 1) in April, 2019.



Figure 1 The second-generation pod (the volume gets smaller)

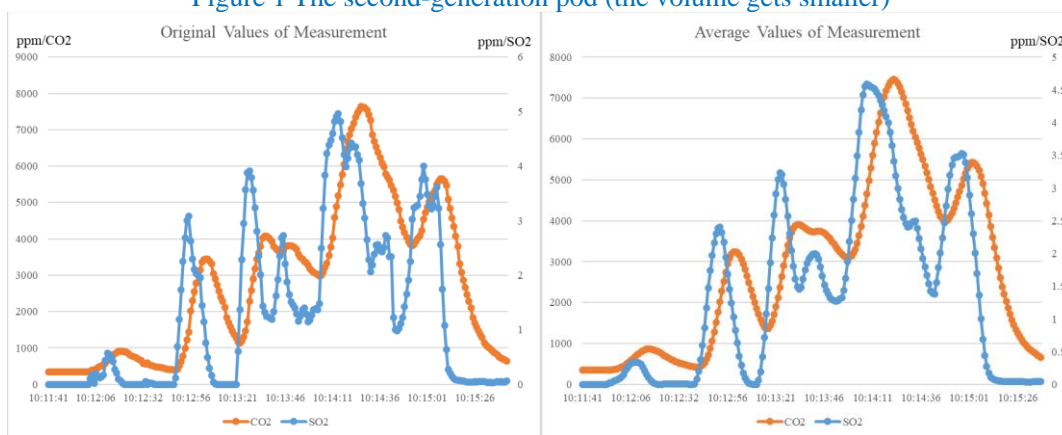


Figure 2 Measurement value of the second-generation pod

5 As shown in Fig. 2, the average values are obviously more stable than the original values, and it is easier to determine the peak value. If the original value is used, the data within certain time range needs to be selected to reduce uncertainty. But the result came out the same as that calculated with the average value.

10 In fact, in the calculation method with area ratio, the start time and end time of the area are also need to be confirmed. The problem of uncertainty remains. It is a process similar to that of peak value selection method and confirming the time span of integral described in this manuscript. Therefore, "the ratios of the areas" is necessary and can be calculated by average values of measurement. Nevertheless, we used the flight procedure given in section 3.1 and the selection method of peak values to minimize the impact of uncertainties. We believe the sharing of these results could helpful to other researchers. As the

15 comments of Referee #2, "It should be noted that the accuracy of the results of monitoring is a difficult issue and the accuracy estimates in literature may not always be comparable."

Finally, if necessary, I can provide all the data and flight recorders (stored in a database) to demonstrate the effect of this method in 23 experiments.

In the end, thanks again for the positive 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

Page 1 Line 20 Perhaps there are more recent data (references) on the contribution of shipping than those from 2010. Especially since there is mention of “rapid development”

Two references have been added.

Liu et al. (2016) reported that East Asia accounted for 16% of global shipping CO₂ emissions in 2013, which was an increase compared to only 4–7% in 2002–2005. In the research of Russo et al. (2018), who evaluated the contribution of shipping to overall emissions over Europe, this sector was found to represent on average 16%, 11%, and 5% of the total NO_x, SO_x, and PM₁₀ emissions, respectively.

Reference:

Russo, M. A. , Leitão, J. , Gama, C. , Ferreira, J. , & Monteiro, A. . (2018). Shipping emissions over europe: a state-of-the-art and comparative analysis. *Atmospheric Environment*, 177, 187-194.

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.

Page 2 line 11 English: the FSC content rather than FSC limit. And perhaps it should not say must not exceed but may not exceed.

This sentence has been rewritten.

The FSC limit was set to 0.1% (m/m) beginning in 2015.

Page 2 line 26 English: check the sentence starting with Optical methods. Unclear.

This sentence has been rewritten and added the reference.

Optical methods analyze the variation of the light properties after interaction with the exhaust plume and allow, if the local wind field is known, to determine the emission rate of SO₂. The simultaneous measurement of CO₂ and SO₂ emissions at a routine basis with these systems is unrealistic at the moment (Balzani Lööv et al., 2014).

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.

Page 2 Line 29 English: for calculating change to: to calculate

Page 2 Line 32 English: ship's plume rather than ship plume

Page 3 English: more accuracy estimation changes to more accurate estimation

These sentences have been rewritten.

Page 3 lines 10 etc. It should be noted that the accuracy of the results of monitoring is a difficult issue and the accuracy estimates in literature may not always be comparable. Please make a note on that.

Yes, I have added a note as fellow in the manuscript.

It is important to note that the accuracy of the results of monitoring is a difficult issue to address, and the accuracy of estimates in the literature may not always be comparable. For ideal comparison results, one would need to board the ship to take fuel samples, which is particularly difficult for sailing ships.

Page 3 the section starting line 15 with Ship emission measurements. It is perhaps not the best option to start here with the distinction between land-based methods and others. I would suggest to start with this subject (i.e. land based), add fuel sampling and then address all issues on accuracy etc. that's to me seems a more logic order.

This paragraph is the introduction of ship emission measurements according to five different platforms. Land-based and airborne-based methods are the two main platforms at present. As such, the most suitable approach for monitoring compliance is to employ “sniffer” measurements taken by airborne. However, the cost of airborne is high. Therefore, it is useful to research the ship emission measurements by UAV.

It is the writing logic of this paragraph.

I'm not sure I fully understand the suggestion. Because the accuracy has been talked about when introducing optical methods and “sniffing” methods in the previous paragraph (introduction of ship emission measurements according to optical methods and “sniffer” methods).

However, I think the logic of the section may not be very clear, and I have modified this paragraph to make it more logical.

Ship emission measurements can be divided into land-based (Kattner et al., 2015, Yang et al., 2016), airborne-based (Beecken et al., 2014, Aliabadi et al., 2016), marine-based (Cappa et al., 2014), 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. Airborne-based measurements can approach ship's plume and collect exhaust gas from the target ship. 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. 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. 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 aircraft. 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).

Reference:

- 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.
- 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.
- Beecken, J., Mellqvist, J., Salo, K., Ekholm, J., and Jalkanen, J.P.: Airborne emission measurements of SO₂, 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.
- 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.
- Ding, J., van der A, R. J., Mijling, B., Jalkanen, J.-P., Johansson, L., and Levelt, P.F.: Maritime NO_x emissions over Chinese seas derived from satellite observations. *Geophysical Research Letters*, 45, 2031–2037, doi:10.1002/2017GL076788, 2018.
- 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.
- 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.

Malaver Rojas, J. A., Gonzalez, L. F., Motta, N., Villa, T. F., Etse, V. K., and Puig, E.: Design and flight testing of an integrated solar powered UAV and WSN for greenhouse gas monitoring emissions in agricultural farms, International Conference on Intelligent Robots and Systems, Big Sky, Montana, USA, 2015 IEEE/RSJ, 1–6, 2015.

5 Mori, T., Hashimoto, T., Terada, A., Yoshimoto, M., Kazahaya, R., Shinohara, H., & Tanaka, R. Volcanic plume measurements using a UAV for the 2014 Mt. Ontake eruption. Earth, Planets and Space, 68(1). doi:10.1186/s40623-016-0418-0, 2016.

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

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

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.

15 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.

Page 3 line 21 English: collect exhaust gas. This change is also needed in other parts of the Text.

Page 3 line 26 English: taken from aircraft and not airborne.

20

These sentences have been rewritten.

Figure 1 legend: filter to remove water vapor? or is that a dryer (nafion)?

25 It is a gadget to remove water vapor, as follows:



Figure 1 Different types of gadget

Figure 2 is a nice picture, but it is not very illustrative. There is more than one exhaust and there is no visible plume. Please consider using another picture.

30 I have checked my phone and some pictures are as Fig. 2. Because the laboratory is located inside the Shanghai Maritime University, exhaust gas is cleaned before discharge in most cases. It is rare to see a plume. The picture used in the manuscript seems to be the best choice.



35

Figure 2 Pictures of the automatic engine room laboratory of Shanghai Maritime University.

Page 5 If the Matrice instrument is modified, perhaps something should be said on what has been done. Or say: small modifications.

5

Modifications mainly includes add the bracket and power interface for the pod, “small modifications” are appropriate, it has been modified.

In the experiment, we used the MATRICE 600 UAV (SZ DJI Technology Co., Ltd.) with a few small modifications.

10

Page 6 I thought the SO₂ sensor was rather new. Yet reference is made to Hodgson 1999. Is that correct? Please check.

15 According to the original suggestion requirements that “give more specific information on how the electrochemical cell mechanism works for SO₂”. The reference herein is used to illustrate the characteristics of electrochemical sensors of SO₂. It is not for this particular sensor that we used. I have modified the sentence as:

20 The SO₂ electrochemical sensor has the advantages of low power consumption, small size, light weight, and high precision. In addition, **this type of sensor** is capable of measuring SO₂ at a low ppb range (Hodgson et al., 1999).

Hodgson, A. W. E., Jacquinet, P., and Hauser, P. C.: Electrochemical Sensor for the Detection of SO₂ in the Low-ppb Range. Analytical Chemistry, 71(14), 2831–2837, doi:10.1021/ac9812429, 1999.

25

Page 7: at what distance from the ship is the background determined? This may vary but what are ranges.

30 More than 50 m away from the ship's smoke. As described in the manuscript: “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 SO₂ and CO₂.”. I have added information as fellow.

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 SO₂ and CO₂. The take-off position is usually on the dock and is more than 50 m away from the ship's smoke.

35

Page 8 line 2: English: flied change to flew

40

It has been modified.

Pag 2 line 3. Why not determine the background after the peak as well? And see how it changes

the results (or not). Please discuss.

It seems to be at Pag 8 line 3.

It's been discussed in the manuscript as fellow:

- 5 “Observed SO₂ and CO₂ 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”.

Page 8 discussion on sampling interval. It does not seem to be completely solved by setting all intervals to 10 second. This is an outcome of the experiments.

- 10 According to the first round of comments. I added this discussion on sampling interval to explain “how can we synchronize the time variations of two different measurements (SO₂ and CO₂) in order to calculate their ratio.”

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

The interval of integration is 10 s in Eq. (1), which is a parameter.

15

As discussed in section of experimental results. “After the measurement of plume 5, we adjusted 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).” But the interval of integration in Eq. (1) is still 10 s.

- 20 I agreed to the fact that “It does not seem to be completely solved by setting all intervals to 10 second”. In fact, we increased the sampling rate to 1 s and send the original value in the second-generation Pod in April, 2019. After a few experiments of changing the interval of integration, the accuracy did not seem to improve significantly. The interval of 10 s is appropriate to achieve accurate results relatively.

- 25 **Page 8 discussion on what is called “exhaust uncertainty” is difficult. In my opinion this is a completely different uncertainty and should be discussed on a different location in the paper.**

-We measure the composition of the exhaust gas and derive from that the S content in the fuel. There are errors: caused by inadequate sampling, calibration, errors subtraction of the background etc. An attempt should be made to estimate the total error in the measured fuel content.

30

That is one side of the comparison:

-on the other side there is fuel sampling with its errors (perhaps), and the representativity of the sample. Is the fuel sample representative for the exact moment of the measurement with the UAV? Is there a chance that the ship may have switched fuel.? These are additional uncertainties (that play a role in European harbors: switching of fuels. But no need to go into that. Ok we assume that the sample is representative is the right sample.

35

Then the last question is:

-is all Sulphur converted to SO₂ or also to other species. In Balzani et al a number of 6 % Sulphur s mentioned that is not converted. A correct sniffer method would then automatically underestimate the fuel content by 6%. This was at a S content of 1%. It is unclear whether this percentage is still to be expected at 0.5 % Sulphur. Please make a note of that. The 6% is rater

40

uncertain and probably depends on several parameters.

Therefore, I think that the exhaust uncertainty should not be discussed in the series of errors because it is not an error. It may cause unexpected differences, but it is not an error. A flawless monitoring method would discover this difference. Discuss this issue in the final comparison

5

Yes, I agree with the viewpoint that exhaust uncertainty is different. I have put the introduction of “exhaust uncertainty” at the end of section 3.3 (uncertainties), and the discussion of “exhaust uncertainty” in our experiment at section 4.2 (FSC estimation).

In section 3.3:

10 Exhaust uncertainty arises because not all the sulfur in the fuel is emitted as SO₂, which is a completely different uncertainty. Preliminary studies showed that 1-19% of the sulfur in the fuel is emitted in other forms, possibly SO₃ or SO₄ (Schlager et al., 2006, Balzani Lööv et al., 2014). Hence, the assumption that all sulfur is emitted as SO₂ 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.

15 In section 4.2:

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). Overall, the estimated FSC is smaller than the true value in many cases. This could be due to the exhaust uncertainty that not all the sulfur in the fuel is emitted as SO₂. 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 tendency of underestimation has also been found in previous studies (Johan, R et al. 2017).

25 “switching of fuels” is an important factor. In our experiment, one person operated the UAV to monitor the plume. Two maritime law enforcement officers board the ship to collect fuel samples at the same time. Both processes took about 10-20 minutes. As a result, the sample can be thought as the right sample.

30 **Pag 9. Put the remark on the absence of ships with exhaust cleaning in the sample somewhere else. Perhaps in the conclusion.**

This statement is also intended to remove uncertainty of this factor. I put it in the uncertainty analysis.

35

Page 10 line 4 English: ship smoke: change to the ships plume

It has been modified.

40

Page 10 selection method: number 3.....No problem ruling out those cases, but I wonder how exhaust uncertainty could explain this. As was mentioned earlier in the text: this is because only

94 % of the Sulphur is oxidized to SO₂.

“The peak values resulting from dramatic changes” means that the percentage of SO₂ and CO₂ in the plume is not stable at the time of measurement. “Exhaust uncertainty arises because not all the sulfur in the fuel is emitted as SO₂. Preliminary studies showed that 1-19% of the sulfur in the fuel is emitted in other forms, possibly SO₃ or SO₄ (Schlager et al., 2006, Balzani Lööv et al., 2014)”.

Therefore, I guess the dramatic changes in the percentage of SO₂ and CO₂ is probably due to changes in the conversion rates (S to SO₂ or C to CO₂).

But that's just a guess of me. So, I used the word “may”.

This **may** be because of exhaust uncertainty.

In addition, sensor uncertainty and unstable concentrations of SO₂ or CO₂ in the atmosphere may also cause such case. I have added.

This may be because of sensor uncertainty, exhaust uncertainty or unstable concentrations of SO₂ or CO₂ in the atmosphere.

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.

Balzani Lööv, J. M., Alföldy, 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.

Page 11 first sentence This will make the FSC relatively larger that of CO₂. Unclear what is meant.

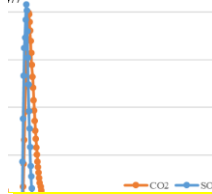


Fig. 3 screenshots

As shown in Fig. 3:

There is a small deviation between the time point of the peak values for SO₂ and CO₂. Therefore, there is two ways to select the time point of peak value: (1) the time point of SO_{2,peak}. (2) the time point of CO_{2,peak}. Different selection methods result in small differences in FSC estimates.

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

As in Eq. (1), The S in the numerator, C (fuel) in the denominator. We select the time point at peak of SO₂. “This will make the FSC value relatively larger than that of CO₂. As in Eq. (1), a higher SO₂ peak leads to a higher FSC estimate, while a higher CO₂ peak leads to a lower FSC estimate. As discussed in section 3.3, not all the sulfur in the fuel is emitted as SO₂, which will result in a lower estimate value. This selection allows the estimate to be relatively close to the true value.”

Figure 5 This is the core of the paper. I have many comments, but will not all give them:

(1)-Plume 1. There is an additional SO₂ source (and not CO₂ obviously) That is strange. Often SO₂ comes from fossil fuel burning. So if the SO₂ rises often CO₂ rises as well. Where does the SO₂ come from?. Or is it a drift or cross sensitivity in the SO₂ sensor? Please comment. Nevertheless, I think the interpretation is OK.

5 (2)-Plume 2 Same on the SO₂ background. Poor quality expected

(3)-Plume 6 seems good quality. But the maximum value is reached, and it is rejected(?)

(1-2) I made more comments in the manuscript as follow:

10 In some cases, background values seemed to fluctuate greatly. This was mainly because the UAV took off from the dock, where multiple ships were berthed, and wind speeds were high. In addition, the drift or cross sensitivity in the sensors also may have caused interference. Therefore, we used the flight procedure given in section 3.1 and the selection method of picking values to minimize this impact.

(3) The peak values of CO₂ at the full range of the SO₂

General remarks:

15 -(1) I would consider not using auto scaling of the Y axis to give the reader the chance to compare the different plumes

-(2) The different peaks in the concentrations are related to the distance to the funnel. If you go a bit further away from the funnel perhaps the plumes are more mixed, and no individual peaks are observed and interpretation is easier. Please discuss.

20 -(3) Although the level of detail is nice to inform the reader some structure could still help. Perhaps make a distinction between: good quality examples and poor-quality examples and rejected plumes, cases with high Sulphur content, case with low Sulphur content and then don't use autoscaling of the y axis. Also show how well the selection procedure works in the presented cases.

25 -(4) It is stated that the deviation is within 300 ppm is. But it is not entirely clear how the best value is selected. Please discuss the selection of the best value in relation to the error estimates on page 9 (equation 2 and the outcome of that)

30 (1) I tried to not use the auto scaling of the Y, but the results didn't seem to be ideal. Take the plume 2 and plume 6 as examples: Y axis of CO₂ is 800 ppm and SO₂ is 800 ppb in plume 2. In plume 6, the value is 6000 ppm and 4000 ppb, respectively. If we set the Y axis as fixed axes (6000 ppm and 4000 ppb for CO₂ and SO₂, respectively), then the peak value of plume 2 is difficult to distinguish as show in Fig 4.

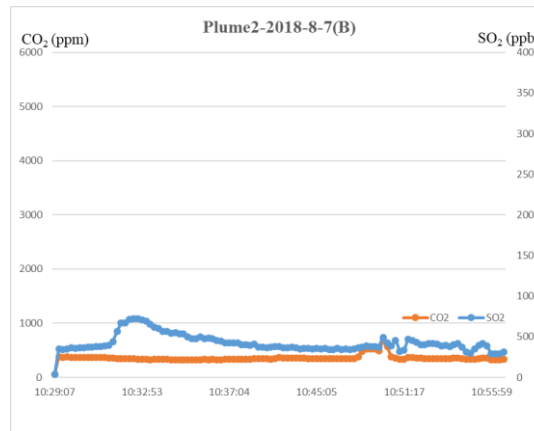


Fig 4. Plume 2 in fixed axes

(2) According to our experience, the gas concentration may exceed the range of sensor when the UAV is very close to the funnel. However, it is difficult to establish a clear correspondence between distance and measurement value. Wind speed and direction may cause interference. In addition, different ships have different flue lengths. I guess this problem may be testable in the laboratory.

5 (3) Good advice, I have made corresponding modifications in section of experiment.

(4) I'm not sure I understand the question exactly.

How to select the peak and background values are listed in Table 2, Fig. 5 and corresponding discussion in the manuscript. When the peak and background values are determined, the best value is confirmed.

10 Whether this means that there are still many options after the peak or background is confirmed? No, it is confirmed. As written in the manuscript:

“According to the flight record, the minimum values of SO₂ and CO₂ collected at the first measurement point are selected as the background values.” “The occurrence time of peak values in SO₂ and CO₂ 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 SO₂ and CO₂, we select the time point at peak of SO₂.”

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

$$\Delta SO_{2,peak} = TrueValue(SO_{2,peak}) - MeasurementValue(SO_{2,peak}) \quad (3)$$

20 Although the true value of FSC and the measurement value of SO₂ can be obtained. However, the true value of SO₂ is unknown. It is often replaced by the average of multiple measurements which can be got in simulations or laboratory experiments. In our experiment, formula 2 only shows the relationship between the deviation in the measurement values and that in the FSC.

25 **Page 13 line 15. Please rephrase to: This could be due to Sulphur emitted in other forms. This is not proven or so in this paper.**

-(1) The choice of peaks that are selected in the end remains a bit arbitrary. Picking the highest perhaps leads to the best comparison but why? The authors should defend this choice. Or illustrate what happens when other choices are made

30 **-(2) Perhaps in the table the three peaks could be averaged, and a standard deviation in the calculated Sulphur content could be presented. Just to illustrate how large errors may be.**

-(3) And it would be very nice if the error calculated from equation 2 is given for each sample. This could show why difference between measurements by UAV and fuels samples are sometimes larger than other times

35 **-(4) If the background values for CO₂ can be used it is perhaps better to carry out a recalibration afterwards. Presenting CO₂ concentrations far below 400 is not wanted. Or use an arbitrary scale (?). pleas change.**

I have made corresponding modifications.

(1) “Picking the highest perhaps leads to the best comparison but why?”

40 My guess is as follows:

The maximum values are likely to have been measured in the center of the ship's plume. At that location, the measurement value is relatively stable, and the probability of interference from other factors is lower.

5 I think this can be tested by detailed experiments in the laboratory. Nevertheless, the results of other choices are list in Table 2 in the manuscript. It can be seen that this approach can indeed achieve more accurate results according to the experiments. This idea has been proved to some extent.

10 (2) "how large errors" is listed in table 2. It can be seen that the number of peaks is usually 1-3. According to the selection method of peak values, some peaks need to be ruled out., only one or two peaks can be retained. Standard deviation is hard to calculate.

If there are multiple alternative results in one experiment, then if the deviation between the results is small, then it seems that the results are relatively stable; otherwise, a large deviation may occur.

However, based on the current experimental results (23 samples, only one peak in sometimes), it does not seem that the average value can get more accurate results.

15 (3) Yes, the biggest obstacle is that the cost of experiment is too high. "the accuracy of the results of monitoring is a difficult issue and the accuracy estimates in literature may not always be comparable."

(4) Yes, after the measurement of plume 5, the sensors were consequently recalibrated by standard mixture gas.

20 **Page 12 line 10. It is not clear what is meant with interpolation ratio.**

The sentence was modified as follows:

25 Nonetheless, Eq. (1) was used to calculate the ratio of sulfur dioxide difference to carbon dioxide difference, and it therefore does not affect the final calculation results.

30 **Pag 14 line 13. Significant contribution to the literature. I would refrain from such statements. That is for the readers to decide. Perhaps state that this research is part of a process towards reliable equipment to monitor FSC in SECA.**

Yes, I have made corresponding modifications.

35 **Page 15 point 2. Is it possible to sate when errors are large? This could be used to identify poor measurements. And it would be helpful.**

Yes, I have made corresponding modifications.

40 Poor-quality data or rejected plumes may derive from these situations.

I think the battery life is important but a rather detail and perhaps not for the conclusions but somewhere in the technical descriptions.

Yes, I have made corresponding modifications.

5

Abstract:

- change the order of first two sentences.
- describe the principles of the method in a few words/lines
- Use 23 samples rather than more than 20

10

Yes, I have rewritten the abstract.

Air pollution from ship exhaust gas can be reduced by the establishment of Emission Control Areas (ECAs). Efficient supervision of ship emissions is currently a major concern of maritime authorities. In this study, an Unmanned Aerial Vehicle (UAV)-based measurement system for exhaust gas from ships was designed and developed. Sensors were mounted on the UAV to measure the concentrations of SO₂ and CO₂ in order to calculate the fuel sulfur content (FSC) of ships. Waigaoqiao port in the Yangtze River Delta, an ECA in China, was selected for monitoring compliance with FSC regulations. Unlike in situ or airborne measurements, the proposed measurement system could be used to determine the smoke plume at about 5 m from the funnel mouth of ships, thus providing a means for estimating the FSC of ships. In order to verify the accuracy of these measurements, fuel samples were collected and sent to the laboratory for chemical examination, and these two types of measurements were compared. After 23 comparative experiments, the results showed that, in general, the deviation of the estimated value for FSC was 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.

20

25

Suggestions

Please identify weak points in the current approach and provide suggestions for further research and improvements

30

It has been added in the conclusion.

3. Currently, the pod can only carry two sensors. In subsequent tests, we will modify the pod to carry more sensors. The use of different types of UAVs also needs to be evaluated. In addition, our experiments mainly involved the monitoring of berthing ships, and experiments on ships at sea are needed in the future.

35

In the end, thanks again for the positive comments.

40

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

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Abstract. Air pollution from ship exhaust gas can be reduced by the establishment of Emission Control Areas (ECAs). Efficient supervision of ship emissions is currently a major concern of maritime authorities.~~Efficient supervision of ship emissions is currently a major concern of maritime authorities. A potential solution is the establishment of Emission Control~~
10 ~~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. Sensors were mounted on the UAV to measure the concentrations of SO₂ and CO₂ in order to calculate the fuel sulfur content (FSC) of ships. Waigaoqiao port in the Yangtze River Delta, an ECA in China, was selected for monitoring compliance with ~~fuel sulfur content~~FSC regulations.
15 Unlike in situ or airborne measurements, the proposed measurement system could be used to determine the smoke plume at about 5 m from the funnel mouth of ships, thus providing a means for estimating the FSC of ships. In order to verify the accuracy of ~~this~~these measurements, fuel samples were collected and sent to the laboratory for chemical examination, and these two types of measurements were compared. After ~~more than 2023~~ comparative experiments, the results showed that, in
20 (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. Estimations show that ships contribute 4-9% of global SO₂ emissions and 15% of NO_x (Eyring et al., 2010).
25 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 SO₂ 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
30 Mexico and Caribbean Sea, and along the western coast of North America (Johansson et al., 2017). Liu et al. (2016) reported

that East Asia accounted for 16% of global shipping CO₂ emissions in 2013, which was an increase compared to only 4–7% in 2002–2005. In the research of Russo et al. (2018), who evaluated the contribution of shipping to overall emissions over Europe, this sector was found to represent on average 16%, 11%, and 5% of the total NO_x, SO_x, and PM₁₀ emissions, respectively.

- 5 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 ~~was set to~~ 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. 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 the variation of the light properties after interaction with the exhaust plume and allow, if the local wind field is known, to determine the emission rate of SO₂. The simultaneous measurement of CO₂ and SO₂ emissions at a routine basis with these systems is unrealistic at the moment

~~(Balzani Lööv et al., 2014). Optical methods analyze variations in light properties after interactions with the exhaust plume, and the local wind field before determining the SO₂ emission rate is observed. The simultaneous measurement of CO₂ and SO₂ 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~~to calculate 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 SO₂ and NO₂ 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” methods are based on simultaneous measurement of elevated SO₂ and CO₂ concentrations in the exhaust plume from the target ship and comparing them with the background. The measurement of CO₂ allows for relating the measurement of SO₂ to the amount of fuel burned at a given time, thus enabling the calculation of FSC directly. The concentration of SO₂ in plumes was generally measured using UV fluorescence sensors, and CO₂ 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 ~~accurate~~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 SO₂ 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). ~~It is important to note that the accuracy of the results of monitoring is a difficult issue to address, and the accuracy of estimates in the literature may not always be comparable. For ideal comparison results, one would need to board the ship to take fuel samples, which is particularly difficult for sailing ships.~~

~~Ship emission measurements can be divided into land-based (Kattner et al., 2015, Yang et al., 2016), airborne-based (Beecken et al., 2014, Aliabadi et al., 2016), marine-based (Cappa et al., 2014), 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. Airborne-based measurements can approach ship's plume and collect exhaust gas from the target ship. 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. 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. 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 aircraft. 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). 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. 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. 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 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 gas than airborne-based measurements. Waigaoqiao port in the Yangtze River Delta was selected as the study site. By Using using this measurement system, we analyzed more than 2023 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 funnel mouth of ships and design an accurate measurement of FSC.

2. Measurement

2.1 UAV



5 **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), gas circuit, a filter (to remove water vapor), sensors for SO₂ and CO₂, 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.) with a few small modifications, ~~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 15 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.



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

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

5 2.2 Sensors

In the measurement process, the ship exhaust gas 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 SO₂ and CO₂ measurements. These sensors were purchased from HH Feuerungstechnik GmbH, Germany.

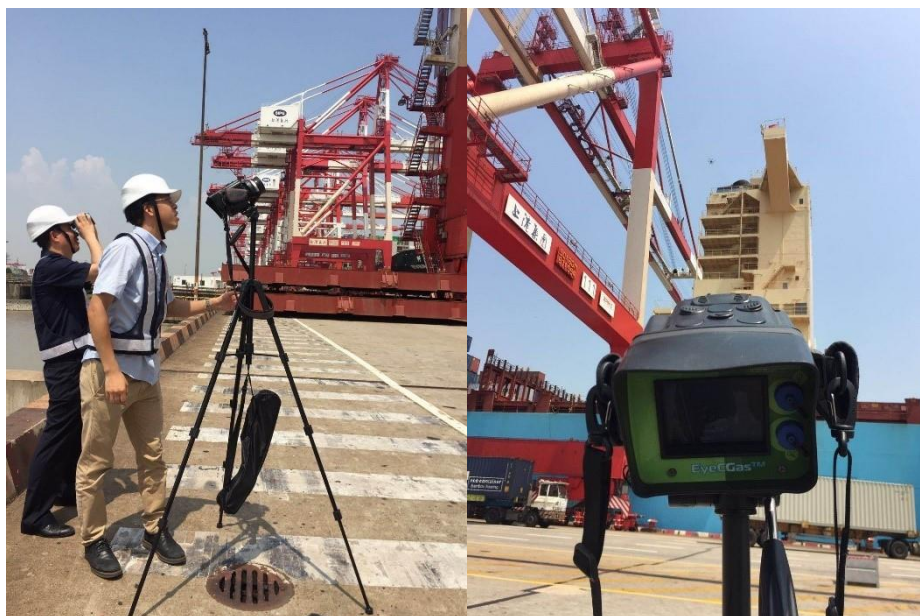
For SO₂, the sensor is based on the electrochemical method. An electrochemical sensor determines the concentration of a gas via a redox reaction, producing an electrical signal proportional to the concentration of the gas. In previous measurements of ship exhaust gas, SO₂ sensors are mainly based on 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 SO₂ electrochemical sensor has the advantages of low power consumption, small size, light weight, and high precision. In addition, ~~this type of~~ the sensor is capable of measuring SO₂ at a low ppb range (Hodgson et al., 1999). Therefore, we used the electrochemical sensor to measure SO₂ 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 ± 0.25 ppm. t_{90} is defined as the time it takes to reach 90% of the stable response after a step change in the sample concentration.

10 For CO₂, the sensor is based on the non-dispersive infrared analyzer method. This type of sensor is often used to measure the CO₂ 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
15 (t_{90}) is less than 1 s, and its accuracy is ± 50 ppm.

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. 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.

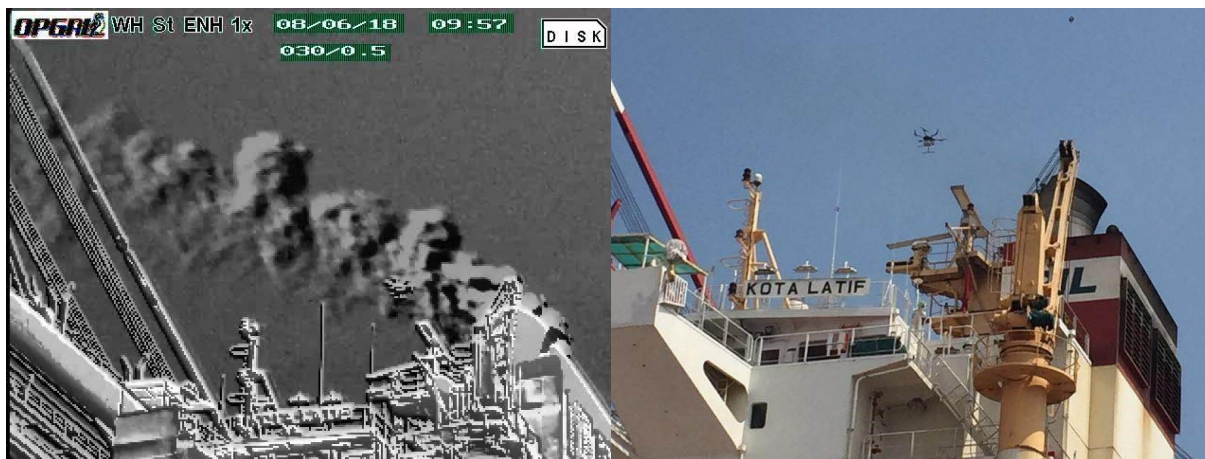
3. Methods

3.1 Flight procedures



(a)

(b)



5

(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 sensors
10 flew close to the funnel of ship, 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 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.
- 5 2. Check the equipment to ensure that: the power is sufficientenough, 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 SO₂ and CO₂. The take-off position is usually on the dock and is more than 50 m away from the ship's
- 10 smoke.
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 funnel for 5 min, respectively.
5. Lift the UAV and then return it to the starting point.

During the process, real-time observations of SO₂ and CO₂ were sent to receiving end. The operator adjusted the UAV's

15 position according to the observations to keep the sensors 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 flew into the ship plume, the peak areas of the SO₂ and CO₂ measurements were determined, and the

20 background was subtracted. The background value of SO₂ and CO₂ can be obtained when the UAV hovers at the first measurement point. The peak values of SO₂ and CO₂ 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 SO₂ and CO₂, respectively, the FSC mass

25 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 SO₂ and CO₂, respectively. 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).

30 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 SO₂ and CO₂. 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.

5 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, ~~exhaust uncertainty~~, and so on.

As for sensor uncertainty, the linear error is negligible and the nonlinearity of the two sensors should be no more than $\pm 1\%$.

10 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 SO₂. Preliminary studies showed that 1–19% of the sulfur in the fuel is emitted in other forms, possibly SO₃ or SO₄ (Schlager et al., 2006, Balzani Lööv et al., 2014). Hence, the assumption that all sulfur is emitted as SO₂ 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 SO₂. This tendency of underestimation has also been found in previous studies (Johan, R et al. 2017).~~

15
20

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.

25 Calculation uncertainty lies in selecting the background and peak values of SO₂ and CO₂. 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:

$$30 \Delta FSC = \frac{\partial f}{\partial SO_{2,peak}} \Delta SO_{2,peak} + \frac{\partial f}{\partial SO_{2,bkg}} \Delta SO_{2,bkg} + \frac{\partial f}{\partial CO_{2,peak}} \Delta CO_{2,peak} + \frac{\partial f}{\partial CO_{2,bkg}} \Delta CO_{2,bkg} \quad (2)$$

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

Exhaust uncertainty arises because not all the sulfur in the fuel is emitted as SO₂, which is a completely different uncertainty. Preliminary studies showed that 1-19% of the sulfur in the fuel is emitted in other forms, possibly SO₃ or SO₄ (Schlager et al., 2006, Balzani Lööv et al., 2014). Hence, the assumption that all sulfur is emitted as SO₂ 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.

- 5 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 300 ppm. In addition, none of the monitored ships were fitted with exhaust cleaning equipment.

4. Results

10 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.

- 15 Fig. 4 shows the UAV platform with sensors flying close to the ship's smokeplume. It hovered to collect exhaust gas, and detection information was subsequently sent back. Generally, changes in SO₂ and CO₂ observations can be divided into three stages: (1) The UAV took off and approached the ship funnel for about 3 min. The SO₂ and CO₂ observations were relatively low, and the background value was obtained in this stage. (2) The UAV was gradually flown to the plume center, and data were collected. Rapid increases in SO₂ and CO₂ concentrations, reaching their peaks, were observed, which took
20 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 SO₂ and CO₂ concentrations relative to the observation when the UAV was in

the plume center were observed. Observed SO₂ and CO₂ 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 2023~~ 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 SO₂ and CO₂ values simultaneously reach their peaks, it is easier to select the background and peak value ~~for calculating to calculate~~ the FSC. However, the actual data collected are sometimes not ideal, and there is calculation uncertainty when selecting the background and peak values of SO₂ and CO₂. 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 SO₂ and CO₂ in the receiving end were synchronized. Therefore, the background and peak values for SO₂ and CO₂ that we selected to calculate the FSC were observed at the same time point.

According to the flight record, the minimum values of SO₂ and CO₂ 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 SO₂ and CO₂ are selected as the peak values. The selection method is as follows:

1. The peak values in the observations of SO₂ and CO₂ are determined at the second and third measurement points, respectively.

2. The peak values at the full range of the SO₂ or CO₂ sensors are ruled out.

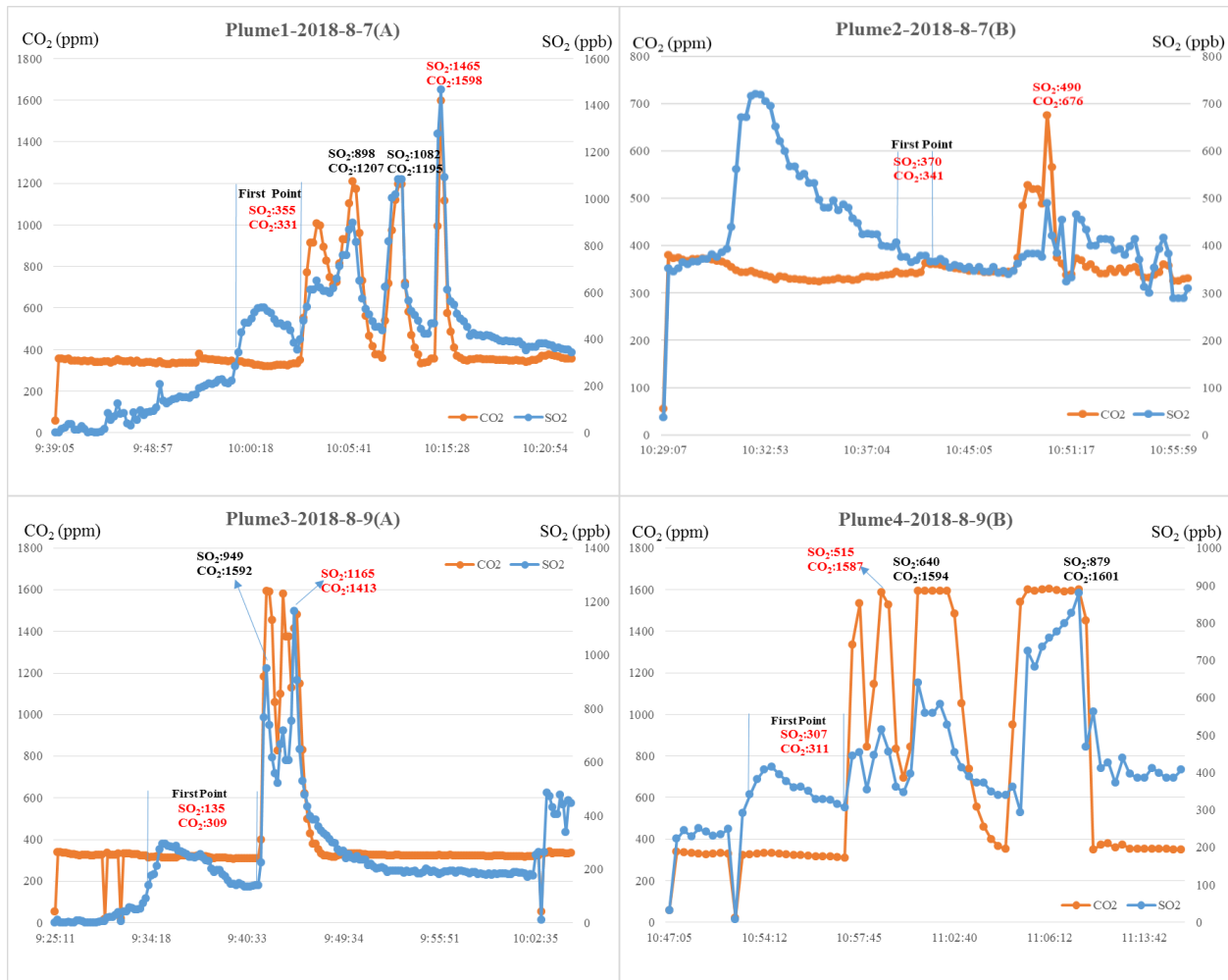
3. The peak values resulting from dramatic changes (for instance, if the change in CO₂ exceeded 500 ppm, or if the change in SO₂ exceeded 500 ppb) in continuous observations are ruled out. because these changes may have been related to sensor uncertainty, exhaust uncertainty, or unstable concentrations of SO₂ or CO₂ in the atmosphere. ~~This may be because of exhaust uncertainty.~~

4. The occurrence time of peak values in SO₂ and CO₂ 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 SO₂ and CO₂, we select the time point at peak of SO₂. This will make the FSC value relatively larger than that of CO₂. As in Eq. (1), a higher SO₂ peak leads to a higher FSC estimate, while a higher CO₂ peak leads to a lower FSC estimate. As discussed in section 3.3, not all the sulfur in the fuel is emitted as SO₂, which will result in a lower estimate value. This selection allows the estimate to be relatively close to the true value.

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 to calculate~~ the FSC. The maximum values are likely to have been measured in the center of the ship's plume. At that location, the measurement value is relatively stable, and the probability of interference from other factors is lower.

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 SO₂ and CO₂, marked as plumes 1–6, along with the time and serial number, are shown in Fig. 5. In addition, we made a distinction between good-and poor-quality data and rejected some plumes. Good-quality data for a plume meant that the peak values were obvious and easy to distinguish, whereas poor-quality data for a plume meant that the peak values were less obvious but still able to produce a result. When results could not be obtained, the plumes were rejected. An FSC of 0.01% (m/m) was used as the dividing line, and the plumes were divided into high-sulfur content and low-sulfur content samples.



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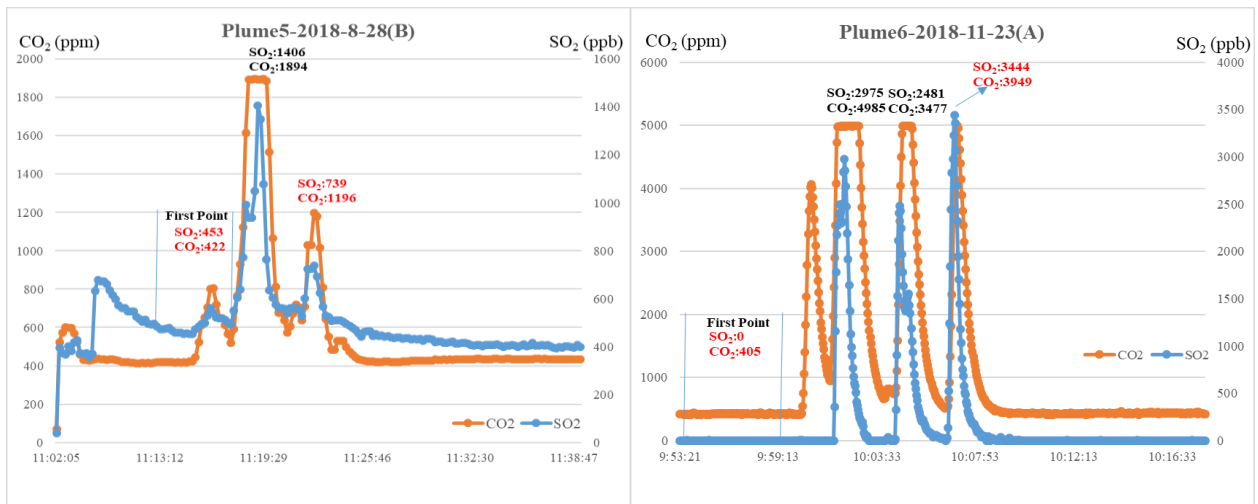


Figure 5. Six sets of plume measurement data for SO₂ and CO₂, marked as plumes 1–6, along with the time and serial number. The background and peak values of SO₂ and CO₂ were used to estimate the FSC. In each plume, the time range of the first monitoring point is marked by two vertical lines. The selected background and peak values of SO₂ and CO₂ 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 SO₂ and CO₂ peak values, and the global maximum peak values of SO₂ and CO₂ were selected. In plume 2, there was a peak for SO₂ at 10:32, but there was none for CO₂ at the same time. We used the data from the simultaneous peaks of SO₂ and CO₂ 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 SO₂ measurements reached the peak values, but the CO₂ 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, CO₂ measurements did not increase any further owing to the full range of the CO₂ sensor at 10:02 and 10:04. This happens in rare cases when the UAV is too close to the 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 were consequently recalibrated by standard mixture gas. Therefore, the background values of plumes 1–5 were different from those of plume 6 were not the same as those of plume 6. Nonetheless, Eq. (1) was used to calculate the ratio of sulfur dioxide difference to carbon dioxide difference—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 SO₂ 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.

The background and peak values of SO₂ and CO₂ 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 CO₂ in

plumes 1–4 exceeded 300 ppm, but the global background CO₂ was approximately 400 ppm. Meanwhile, the background value of SO₂ 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 seemed to fluctuate greatly. This was mainly because the UAV took off from the dock, where multiple ships were berthed, and wind speeds were high. In addition, the drift or cross sensitivity in the sensors also may have caused interference. Therefore, we used the flight procedure given in section 3.1 and the selection method of peak values 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.

- 10 **Table 2 Comparison and verification of the estimated and true values of FSC. We present the selected background and peak values of SO₂ and CO₂ 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	×		949		1592	1472		-641
6		√	135	1165	309	1413	2164	2113	51
7		√		515		1587	378		-18
8	Plume4	×	307	640	311	1594	602	396	206
9		×		879		1601	1029		633
9	Plume5	√		739		1196	857		-11
10		×	453	1406	422	1894	1502	868	634
11		√		3444		3949	2255		-132
12	Plume6	×	0	2481	405	3477	1874	2387	-513
13		×		2975		4985	1507		-880

- 15 ~~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 SO₃ or SO₄.~~

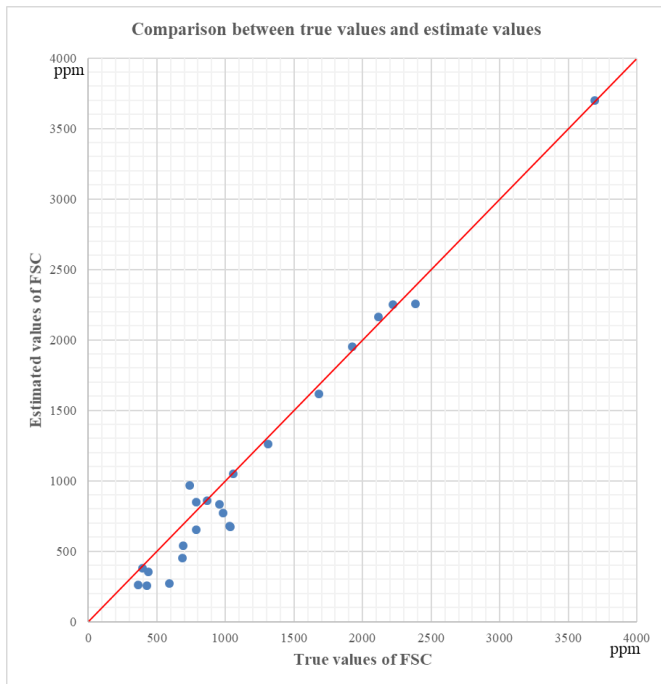


Figure 6. Comparison between the true values of FSC (x-axis) against the estimated values of FSC (y-axis) of 23 times measurement.

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). Overall, the estimated FSC is smaller than the true value in many cases. This could be due to the exhaust uncertainty that not all the sulfur in the fuel is emitted as SO₂. 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 tendency of underestimation has also been found in previous studies (Johan, R et al. 2017).

Finally, 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.

5. Conclusions

In this study, we performed close monitoring of ship smoke plumes using UAV. Observation data of SO₂ and CO₂ were collected at close range (5–10 m) of ship funnel 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 SO₂, 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.

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). The proposed method can be used for monitoring of ECAs for compliance with FSC standards.~~This study makes a significant contribution to the literature because the proposed method can be used for 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), and even more than 0.05% (m/m) in rare cases. Unstable concentrations of SO₂ or CO₂ in the atmosphere just before the measurement may cause such errors. Furthermore, uncertainties, such as sensor uncertainty, ~~measurement~~~~exhaust~~ uncertainty, ~~calculation~~~~measurement~~ uncertainty, and ~~exhaust~~~~calculation~~ uncertainty, may hinder accurate measurement. Poor-quality data or rejected plumes may derive from these situations.

3. Currently, the pod can only carry two sensors. In subsequent tests, we will modify the pod to carry more sensors. The use of different types of UAVs also needs to be evaluated. In addition, our experiments mainly involved the monitoring of berthing ships, and experiments on ships at sea are needed in the future.

~~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 authored the article. SP, WC and XN contributed to 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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