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

Fan Zhou[1], Shengda Pan[1], Wei Chen[2], Xunpeng Ni[2], Bowen An[1]

[1]College of Information Engineering, Shanghai Maritime University
[2]Pudong Maritime Safety Administration of the People's Republic of China

Correspondence to: Fan Zhou (fanzhou_cv@163.com)

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 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.

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.

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. 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, 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 analyse 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 analyse 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 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%) (Van Roy and Scheldeman, 2016a, 2016b, 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).

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) 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. As such, the most suitable approach for monitoring

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compliance is to employ "sniffer" measurements. 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, 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 analysed more than 20 ship plumes and compared the results with the FSC of entering ships determined from fuel samples analysed at certified laboratories. Through these experiments, we investigated and analysed the emission process of SO_2 and CO_2 very close to the smoke stack and design a high precision 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).

In the experiment, we used the MATRICE 600 UAV (SZ DJI Technology Co., Ltd.), and modified it. Pod equipment were installed underneath the UAV to carry pumps, "mini-sniffer" sensors, communication circuit boards, 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. 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

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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

2.2 "Mini-sniffer" sensors

The "mini-sniffer" sensors included instrumentation for both SO_2 and CO_2 measurements. For the former, the sensor was based on the electrochemistry method. An electrochemical sensor determines the concentration of a gas via a redox reaction,

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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 is less than 1 s, and the accuracy is ±5% full scale. For CO_2, the sensor is based on the non-dispersive infrared analyser 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, its resolution level is 1 ppm, its response time is less than 1 s, and its accuracy is ±1% full scale. 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)

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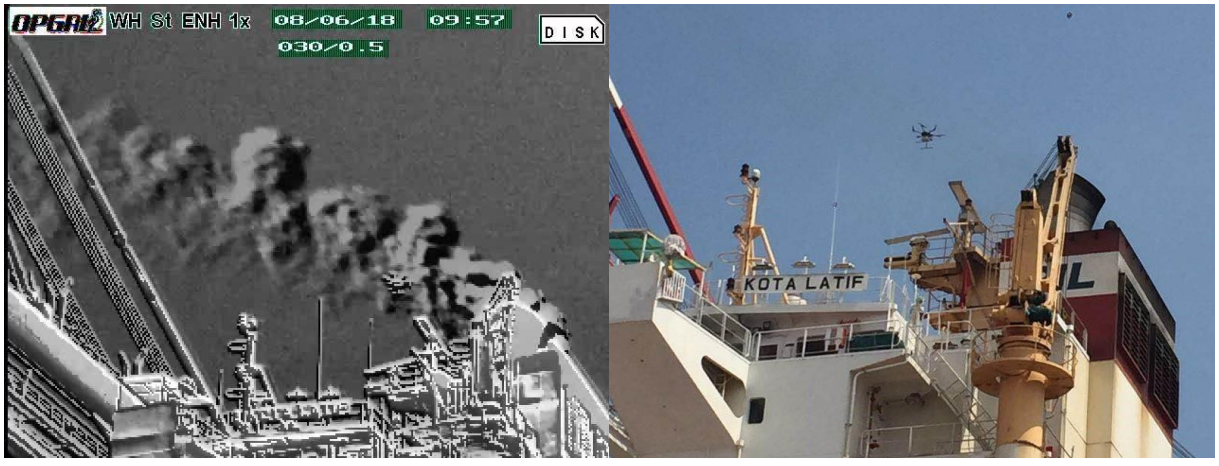

(c) (d)

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 measurement of the ship smoke plume are as shown in Fig. 3. The UAV platform with "mini-sniffer" sensors flew close to the 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 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 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).

3.3 Uncertainties

Because measurements taken inside the ship plumes are analysed 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 correction by frequent calibrations with standard gases and gradually establishing a quality management system.

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

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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, all the monitored ships are not fitted with exhaust cleaning equipment.

4. Results

(a) (b)

Figure 4. Photographs showing the flight of the UAV during measurements. The UAV platform was flown close to the ship smoke 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 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.

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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 were synchronized. Therefore, the background and peak values for SO_2 and CO_2 that we selected 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.

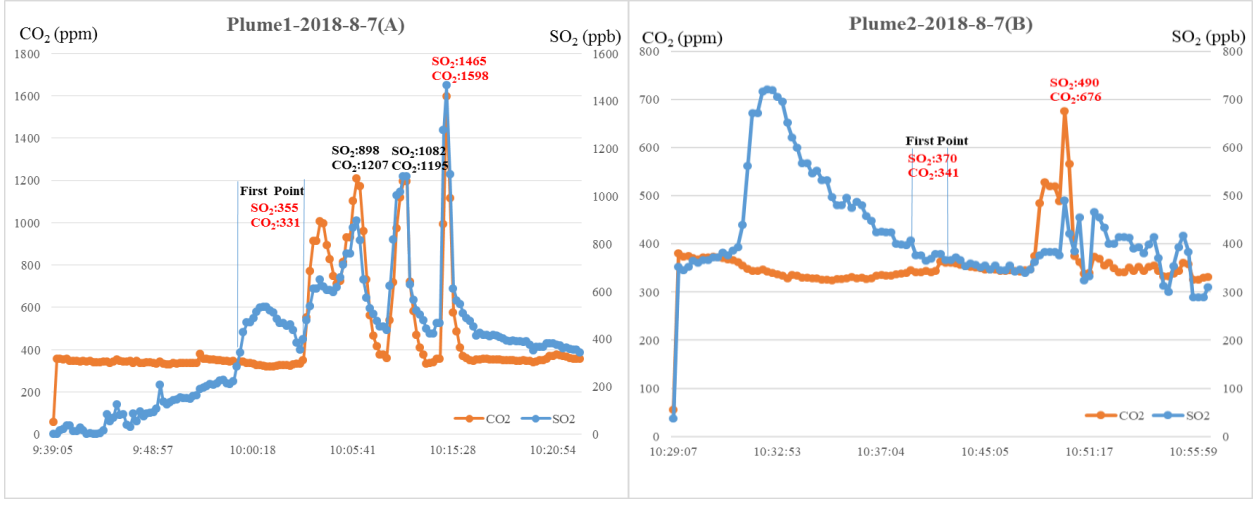
2. The peak values at the full range of the SO_2 or CO_2 sensors are culled.

3. The peak values resulting from dramatic changes 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, 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.

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.



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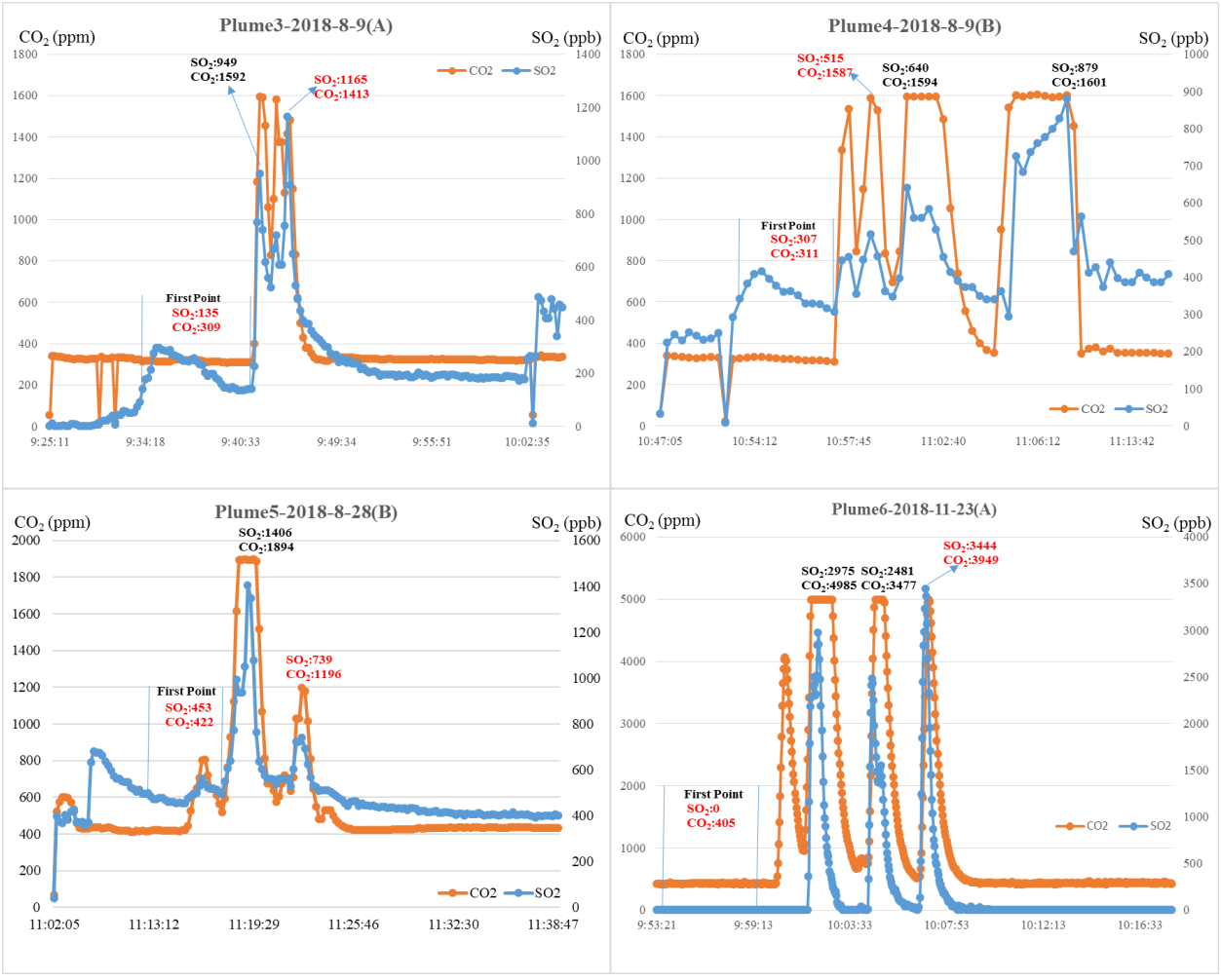

Figure 5. Six sets of plume measurement data for SO_2 and CO_2, marked as plumes 1–6, along with the time and serial number. The background and peak values of SO_2 and CO_2 were used to estimate the FSC. In each plume, the time range of the first monitoring point is marked by two vertical lines. The 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 (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, and the sensor was 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 plumes 1-4 exceeded 300 ppm, but the global background CO_2 was approximately 400 ppm. 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.

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 vessel chimney mouths, 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 300 ppm, and even more than 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.

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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] 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.

[3] 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.

[4] Cooper, D. A.: Exhaust emissions from ships at berth. Atmospheric Environment, 37(27), 3817–3830. doi:10.1016/s1352-2310(03)00446-1, 2003.

[5] 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.

[6] 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, 2018. Doi:10.1002/2017GL076788.

[7] Eyring, V., Kohler, H. W., van Aardenne, J., and Lauer, A.: Emissions from International Shipping: 1. The Last 50 Years, J. Geophys. Res., 110, D17305, doi:10.1029/2004JD005619, 2005.

[8] Fan, S., Liu, C., Xie, Z., Dong, Y., Hu, Q., Fan, G., Chen, Z., Zhang,T., Duan, J., Zhang, P., Liu, J: Scanning vertical distributions of typical aerosols along the Yangtze River using elastic lidar, Science of The Total Environment., 628-629,631–641, doi:10.1016/j.scitotenv.2018.02.099, 2018.

[9] Hodgson, A. W. E., Jacquinot, P., and Hauser, P. C.: Electrochemical Sensor for the Detection of SO_2 in the Low-ppb Range. Analytical Chemistry, 71(14), 2831–2837,1999. doi:10.1021/ac9812429

[10] 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.

[11] IMO, Emission Control Areas (ECAs) designated under MARPOL Annex VI. http://www.imo.org/en/OurWork/Environment/PollutionPrevention/AirPollution/Pages/Emission-Control-Areas-(ECAs)-designated-under-regulation-13-of-MARPOL-Annex-VI-(NOx-emission-control).aspx, 2017.

[12] 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.

[13] MARPOL: International Convention for the Prevention of Pollution from Ships, 1973 as modified by the Protocol of 1978–Annex VI: Prevention of Air Pollution from Ships, International Maritime Organization (IMO), 1997.

[14] MEPC.184(59), Guideline for exhaust gas cleaning systems: http://www.imo.org/blast/blastDataHelper.asp?data_id=26469&filename=184(59).pdf

[15] Prata, A. J.: Measuring SO_2 Ship Emissions with an Ultraviolet Imaging Camera, Atmos. Meas. Tech., 7, 1213-1229, https://doi.org/10.5194/amt-7-1213-2014, 2014.

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

[17] 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.

[18] Seyler, A., Wittrock, F., Kattner, L., MathieuÜffing, Barbara, Peters, E., Richter, A., Schmolke, S., P. Burrows, J.: Monitoring shipping emissions in the German Bight using MAX-DOAS measurements. Atmos. Chem. Phys., 17, 10997–11023, https://doi.org/10.5194/acp-17-10997-2017, 2017.

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

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

[21] 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.