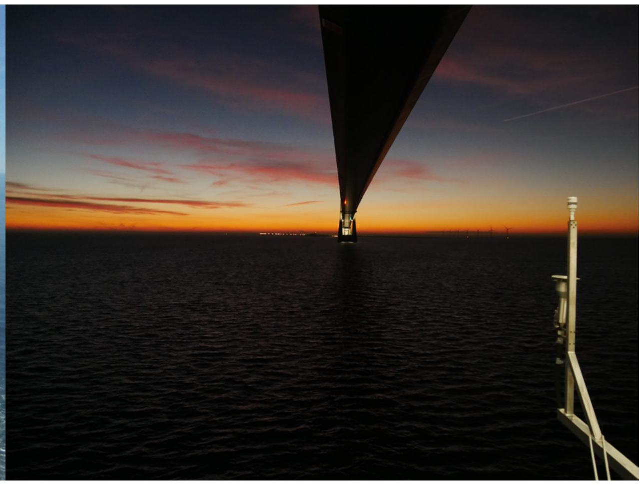
Surveillance of Sulfur Emissions from Ships in Danish Waters

Johan Mellqvist, Jörg Beecken, Vladimir Conde and Johan Ekholm
Department of Space, Earth and Environment,
Chalmers University of Technology, Göteborg, Sweden

Report to the Danish Environmental Protection Agency

Tender no: 2015/S 004-004391

Surveillance of Sulfur Emissions from Ships in Danish Waters

Date
30/12/2017

Title of publication **Surveillance of Sulfur Emissions from Ships in Danish Waters**	
Author(s) Johan Mellqvist, Jörg Beecken, Vladimir Conde and Johan Ekholm	
Commissioned by, date December 30, 2017.	
Keywords Sulfur surveillance, ship emissions, compliance, SECA, Denmark, Airborne, fixed	
Contact person Johan Mellqvist	Language of the report English

In 2015 new rules from the IMO and legislation from EU (Sulfur directive) requires ships to run with maximum fuel sulfur content (FSC) of 0.1 % m/m in northern European waters. In order to promote a level playing field within the shipping sector, there is a need for measurement systems that can make effective compliance control. This report describes the results from ship emission measurements on the waters surrounding Denmark from June 2015 to July 2017 on behalf of the Danish Environmental Protection Agency. The overall aim was to carry out operational surveillance of ships with respect to the EU sulfur directive and particularly the sulfur limits for marine fuel in the European Sulfur Emission Control Area (0.10 %), which entered into force on January 1st 2015, as well as to guide further port state control of ships at the destination harbors of the ships, both in Denmark and other ports. During the project the FSC of individual ships was estimated by performing spot checks of exhaust plumes of individual ships. This was conducted by automatic gas sniffer measurements at the Great Belt Bridge and airborne surveillance measurements using sniffer and optical sensors. The data from the fixed system were transmitted in real time to a web database and alarms were triggered for high FSC ships in the form of emails. The report describes the technical systems and their performance and the general compliance levels of the measured ships. The measurement systems have been developed by Chalmers University of Technology through Swedish national funding and the EU project CompMon.

The airborne dataset corresponds to approx. 900 individual ships, measured by sniffer or/and optical sensor over 245 flight hours. The optical sensor has low precision and is therefore used as a first alert system to identify ships running on high sulfur fuel. The precision of the airborne FSC measurements by the sniffer system is better and it is estimated as ± 0.05 % m/m (1σ) with a systematic bias of - 0.045 % m/m. Therefore only ships running with FSC of 0.2 % m/m or higher can be detected as non-compliant ships with good confidence limit (95 %) by the airborne sniffer system. The airborne measurements during 2015 and 2016 on Danish waters show that 94 % of the ships complied with the EU Sulphur directive, at the 95 % confidence limit. The compliance rate was lower, 92 %, during the 2nd half of 2016.

.

In the period June 2015 to May 2017, 8426 sniffer measurements of individual ships were carried out at the Great Belt Bridge. However, there were technical problems in the first part of the project and the sniffer therefore had reduced sensitivity the first year and only high sulfur ships (> 1 % FSC) could be detected as non-complying vessels with appropriate statistical confidence.

The precision in the estimated FSC by the fixed sniffer system is estimated as ± 0.04 % m/m (1σ) with a systematic bias of - 0.055 % m/m**.** Therefore only ships running with FSC of 0.18 % or higher can be detected as non-compliant ships with good confidence limit (95 %) by the fixed sniffer system. The data for the period June 2016 to October 2016 show a compliance rate of 94.6 % which increased to 97.4 % in the period January 2017 to May 2017.

The compliance level during different time periods and platforms varied between 92-97 %. Here 1 - 2 % of the ships were in gross non-compliance with the EU sulfur directive with FSC values above 0.5 % m/m. There were differences over time, with the highest values in the summer of 2016. The compliance level was close to the values (95 %) measured by port state control authorities in Sweden and Denmark 2015 and 2016. When comparing ships measured by port state and the ones in this project it can be deduced that the efficiency of finding non-compliant vessels could be increased by at least a factor of 4, if the port state controls were guided by measurements. Most of the non-compliant ships (90 %) were measured high only once. But there were cases with individual ships and ship operators that were more abundant in the non-compliance statistics. The non-compliant ships that were seldom in the area around Denmark had higher emissions of SO_2 than the non-compliant ones that operated their more frequently. On several occasions during this study we encountered ships equipped with scrubbers that were non-compliant with respect to the EU sulfur directive.

.

Table of Contents

Acronyms

AIS	Automatic Identification System
DEPA	Danish Environmental Protection Agency
DOAS	Differential Optical Absorption Spectroscopy
FSC	Fuel Sulfur Content in mass percentage (m/m)
IGPS	Identification of Gross Polluting Ships
IMO	International Maritime Organization
MEPC	Marine Environment Protection Committee
MARPOL	Marine Pollution
PSC	Port State Control (authority)
SECA	Sulfur Emission Control Area
STC	Supplemental Type Certificate
UV	Ultraviolet

1 Introduction

In 2015 new rules from the IMO and legislation from EU (Sulfur directive) and the US requires ships to run with maximum fuel sulfur content (FSC) of 0.1 % m/m on northern European and US waters. The extra cost of this fuel is 50 %, or more. At present compliance monitoring of ships is carried out by port state control authorities that take fuel samples of ships at berth. This procedure is time consuming and only few ship are being controlled, and none while underway on open waters. The high extra cost for low sulfur fuel and the relatively small risk of getting caught, creates a risk that unserious ship operators will run cheaper high sulfur fuel. In order to promote a level playing field within the shipping sector there is hence a need for measurement systems that can make effective compliance control, without stepping on board the ships.

This report describes the results from ship emission measurements on the waters surrounding Denmark from June 2015 to July 2017 on behalf of the Danish Environmental Protection Agency. During the measurement period the fuel sulfur content (FSC) of individual ships was estimated by performing spot checks of exhaust plumes of individual ships. This was conducted by automatic gas sniffer measurements at the Great Belt Bridge and airborne surveillance measurements. The data from the fixed system were transmitted in real time to a web database and alarms were triggered for high FSC ships in the form of emails. The objective of the report is to describe the technical systems and their performance, but we will also discuss the general compliance levels of the measured ships.

The measurement systems have been developed in the Swedish project *Identification of Gross-Polluting Ships* (IGPS) (Mellqvist et al., 2014) and the EU project CompMon (https://compmon.eu/). This includes a portable and flight certified version of the sniffer system. As part of the CompMon project, fixed measurements were performed at the Göteborg ship channel and Öresund Bridge (Mellqvist et al., 2017b). In addition airborne ship emission measurements were performed at the SECA (Sulfur emission control area) border at the English Channel (Mellqvist et al., 2017a). Similar systems have been applied by the authors elsewhere including Baltic sea (Beecken et al., 2014a; Berg et al., 2012), Göteborg (Mellqvist et al., 2010 and 2014), Rotterdam (Alfoldy et al., 2011 and 2013; Balzani-Loov et al., 2014), Saint Petersburg (Beecken et al., 2014b) and Los Angeles (Mellqvist et al., 2017c).

2 Method

2.1 Sniffer System

With the sniffer system the FSC is directly obtained by sampling of the gas concentrations in the ship plumes. This is done with several commercially available gas analyzer instruments which in some cases have been modified to match measurement requirements especially concerning the response time and pressure dependence.

The FSC is obtained from the ratio between the pollutants and CO_2 inside of the plume. Eq. 1 shows a more general of this calculation, which is consistent with the on board method described in the MEPC guidelines 184(59).

$$FSC = 0.232 \frac{\int \left[SO_2 - SO_{2,bkg}\right]_{ppb} dt}{\int \left[CO_2 - CO_{2,bkg}\right]_{ppm} dt} \text{ [\% sulfur]} \qquad (1)$$

Here CO_2 and SO_2 corresponds to the gas concentrations expressed in ppm (parts per million) and ppb (parts per billion), respectively. The subscript bkg (background) corresponds to the ambient concentration neighboring the plume. The constant 0.232 corresponds to the sulfur-carbon atomic weight ratio multiplied with a factor of 87% that relates to the carbon content of the fuel, and a correction for different units.

The FSC as described on Eq.1 can be considered to be directly proportional to the sulfur to carbon content in the fuel, assuming that all sulfur is converted to SO_2. However, this is only partly true since some studies have shown that around 5 % of the sulfur is present as sulfate in particles (Moldanova et al., 2009; Petzold et al., 2008); hence, the apparent FSC obtained from the SO_2 to CO_2 ratio will be somewhat lower than the true FSC. The sniffer also measures NO_x which play an indirect role by correcting the SO_2 measurements, thus improving the accuracy of the FSC estimations. This additional correction will be further explained in the following sections.

In order to identify a particular emitter ship, the gas measurements must also include wind data and positional information. This is achieved through a meteorological station and, by tracking the name, speed and positional information of ships nearby the measurements area through an Automated Identification System receiver (AIS), Figure 1.

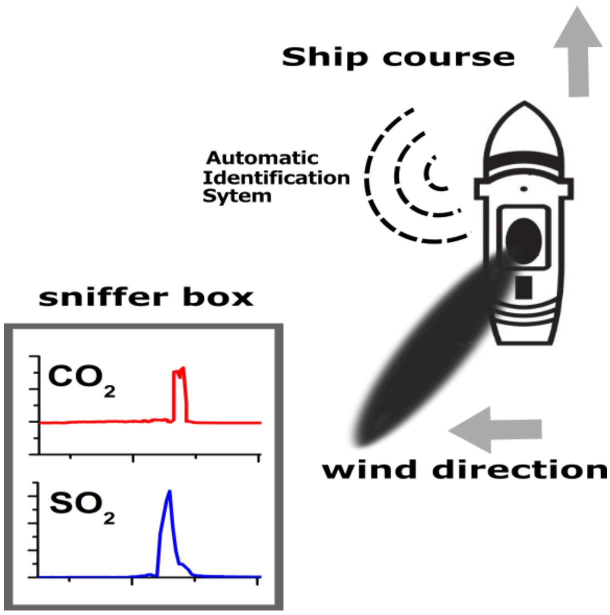


Figure 1. Schematic of the sniffer system and ship identification. An emitter ship is identified by combining wind measurements and the transponder signals through the Automatic Identification System AIS.

2.2 Optical System

In this study the airborne sniffer measurement have been complemented by optical remote sensing using several spectrometers that operate in the ultraviolet and visible wavelength region, respectively, for simultaneous gas column density measurements of SO_2 and NO_2 (Berg, 2012). This system is able to discriminate ships running on 1 % m/m FSC from 0.1 % m/m, and in this project it was used as a first alert system for high sulfur ships that were then further analyzed with a sniffer system. The results from the optical system can also be used directly to guide further control by port state control authorities.

The system measures solar light that has been reflected on the ocean through two telescopes that point 30^o below the horizon, Figure 2. The gas column densities are retrieved from the spectral measurements by applying Differential Optical Absorption Spectroscopy (DOAS) which is a technique widely used for atmospheric measurements from satellites and ground based instruments. From the optical measurements, combined with wind and vessel information, it is possible to estimate the absolute emission rate in gram per second of the retrieved gas species with an uncertainty of about 50 % (Berg et al., 2012). Combined with a model that predicts the instantaneous fuel consumption of a ship (STEAM) an estimate of the FSC can be made (Berg, 2012), following the principles in Eq. 1. The advantage with this method lies in the fact that it is possible to obtain the absolute emission rate. However, it is rather uncertain due to the difficulty of modelling the optical path of the light and uncertainties associated with modelling the fuel consumption. The method also requires that the full plume is transected more or less orthogonally. In this project we have applied a new more flexible variant, using the measured ratio of SO_2 and NO_2 in the ship plume as an indicator for the FSC. This method does not require knowledge of several of the parameters which cause large uncertainties in the method above (optical path, wind speed, ship speed and fuel consumption) and it is therefore considerable faster from an operational point of view. In Figure 3 an example of such optical measurements of SO_2 and NO_2 is shown. The peaks correspond to measurements of two ships using either low or high FSC, as can be deduced from the SO_2 to NO_2 ratio.

In more detail, ships typically emit 40-90 g NO_x per kg of fuel (Becken et al., 2014) and the emission depends on several factors such as age, type, size and load and possible emission abatement system. Most of the NO_x (90-95 %) is emitted as NO but in the air it is rapidly converted to NO_2 by reaction with ozone. Measurements at the Great Belt bridge site show that 15-50 % of the NO_x has been converted to NO_2, and that the amount depends on the distance to the ship. A high sulfur ship (1 % FSC) emits 20 g SO_2/kg and a low one (0.1 % FSC) 2 g/kg. This means that a 1 % FSC ship will typically have a SO_2/NO_2 mass ratio of 1 or higher while the ratio corresponding to a 0.1% FSC ship can be 10 times lower. Naturally, this approach has uncertainties mostly associated with the large variation in the NO_x emissions and in the NO/NO_2 ratio in the flue gas, as indicated above. We have therefore analyzed real measurements to assess how much information can actually be obtained. In Figure 4 is shown the correlation between the measured ratio of SO_2 and NO_2, obtained from the optical sensor, and FSC obtained from airborne sniffer measurements at the SECA border in the CompMon project (Mellqvist et al., 2017a). The data in the figure show that 83 % of 53 ships with a FSC below 0.2 % have an SO_2 to NO_2 ratio below 1. Here we use 0.2 % as a limit to account for uncertainties. The corresponding statistics for 32 high FSC ships (>1 % FSC) shows that 94 % of the ships have a SO_2 to NO_2 ratio higher than 1. Hence, both low and high FSC ships will be classified correctly with about 80 - 90 % probability when using an upper limit of 1 for the SO_2 to NO_2 ratio. Since the main idea is to guide further compliance controls we believe that this probability is sufficient. In this project we have used this approach and ships with a SO_2 to NO_2 ratio above 1 were assigned a FSC value of 1 % m/m in the emission database while ships with a ratio below 1 was assigned an FSC value of 0.1% m/m. In 2020 the FSC limit of all ships outside the SECA region will correspond to 0.5 % m/m. The optical method should be able to distinguish between ships running on FSC of 0.5 % m/m against 2.5 % m/m which is approximately the fleet average, with the same efficiency as distinguishing between ships running on 0.2 % m/m and 1 % m/m as presented here. However, further investigation is needed to assess the efficiency for the optical method to identify ships running on 1 % m/m FSC against 0.5 % m/m.

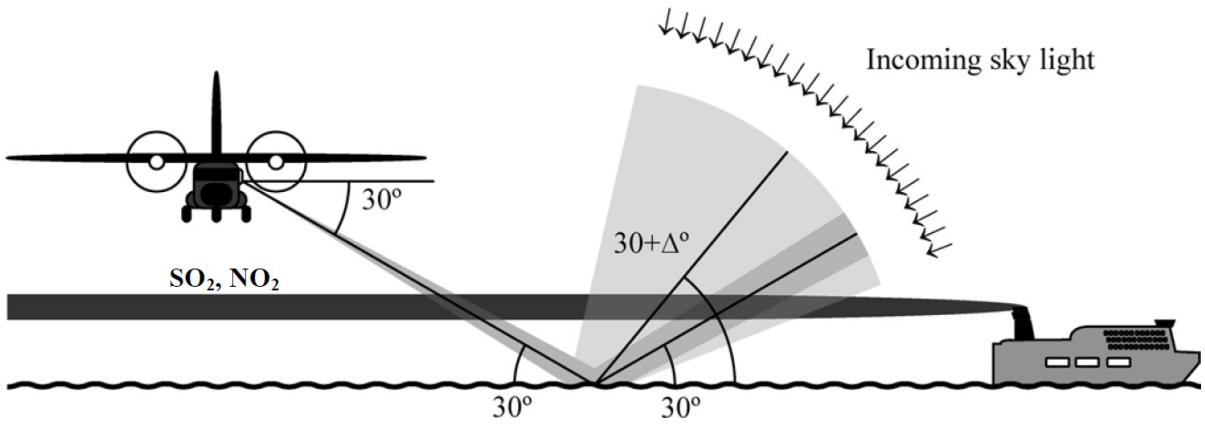


Figure 2. Schematic of the optical measurements of SO2 and NO_2 columns.

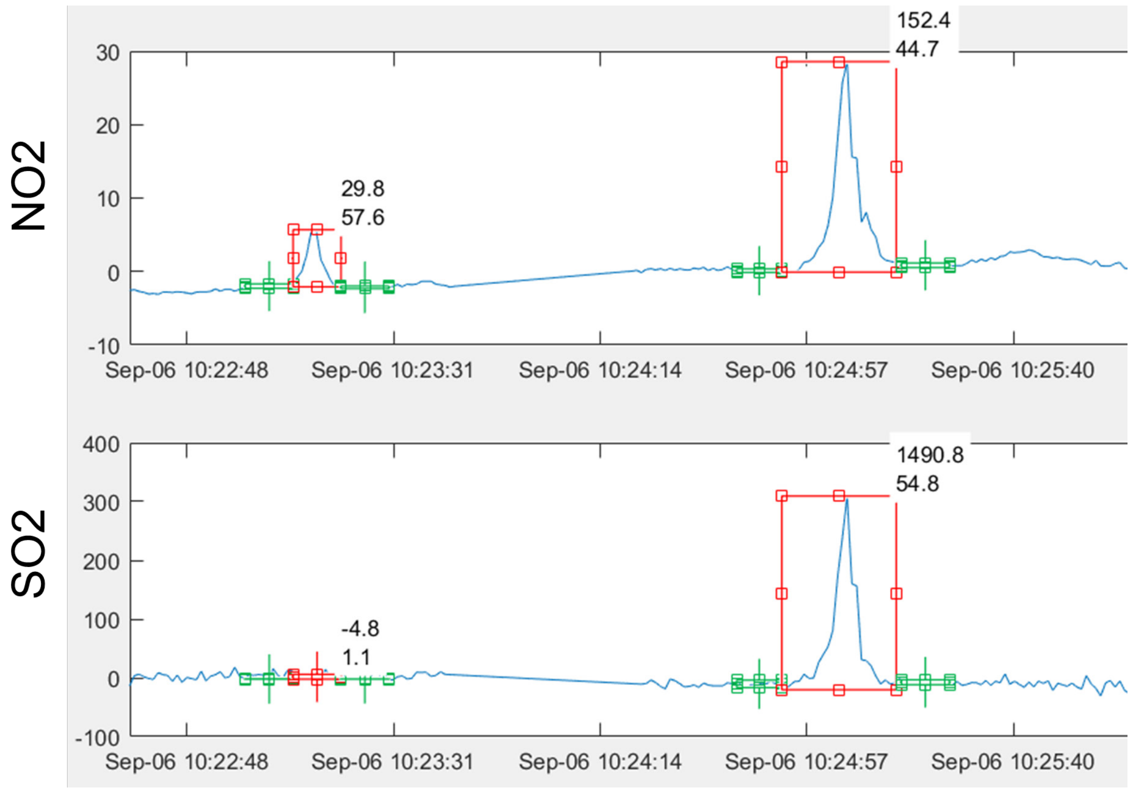


Figure 3. Optical measurements of NO_2 and SO_2 of two ships, one running on 0.1 % m/m fuel and another one running on 1 % m/m fuel is shown. The data were obtained at the SECA border as part of the CompMon project (Mellqvist et al., 2017 a).

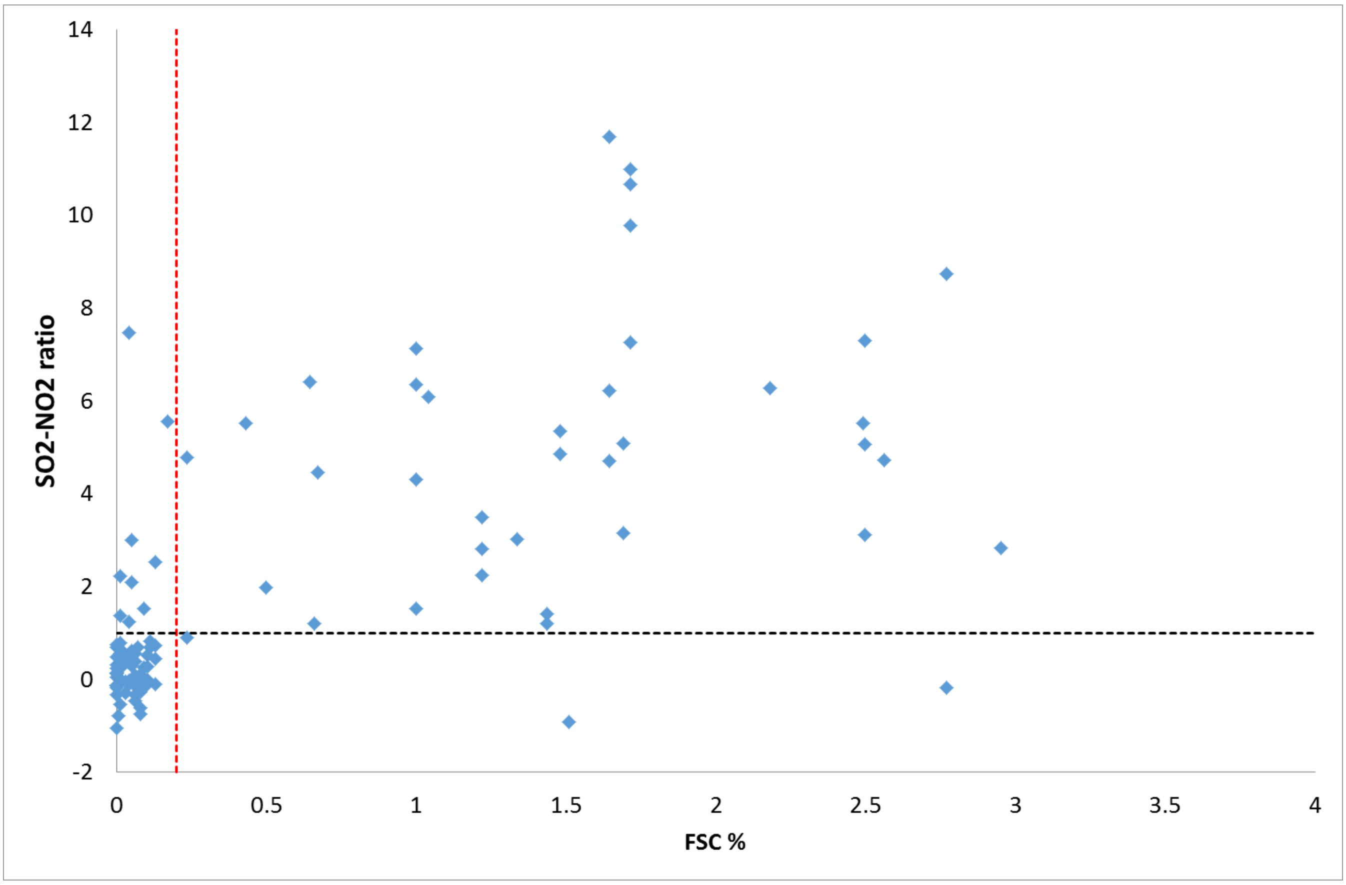


Figure 4. The measured ratio of SO_2 and NO_2 from the optical sensor and the FSC obtained from sniffer measurements. The data were obtained in the CompMon project at the SECA border (Mellqvist et al., 2017a).

2.3 Instrumentation and correction for cross interference

The sniffer and optical systems, respectively, are based on the instruments described in Table 1. The sniffer instruments are commercially available as state of the art instruments and they are being used worldwide as reference methods for air quality measurements. To fulfill flight requirements these instruments have been modified for fast response, smaller weight, smaller shape (form factor) and field robustness. For instance, to be able to obtain a fast response time the SO_2 instrument in the flight system is operated without the so called “kicker” which is a diffusion tube which removes organic substances from the sampling stream before the measurements chamber. Other adaptions correspond to replacement of toxic material (PVC) in the instruments and extra shielding of electromagnetic radiation (Mellqvist et al., 2014; Mellqvist et al., 2017a).

The optical method is based on two spectrometer (f.c. 303 mm/f.c. 160 mm) equipped with UV-sensitive cameras based on CCD (Charge Coupled Device) sensors. A pair of telescopes with 150 mm focal length are connected to the spectrometers through liquid guide fibers (Berg, 2010). In Table 1 the precision (basically same as half of the detection limit) of the instruments and their response times are also shown. The t_{90} parameter corresponds to the time that the instruments need to change from 10 % to 90 % of the signal when making a step change. It has been demonstrated that the instruments in Table 1, built into suitable boxes, are able to operate under harsh ambient conditions. For instance we have operated the instruments from 2 helicopters, two harbor vessels, and two aircraft.

Table 1. The instruments employed for ship surveillance. Response time (t90) and measurement resolution uncertainty (σ) is given.

Species	Quantity	Method	Model	t_{90}	1σ	Platform
CO_2	Mixing ratio (sniffer)	Cavity ring down spectrometer with custom hardware and sampling (sniffer)	Picarro G-2301m	<1 s	0.1 ppm	Air Fixed
CO_2	Mixing ratio (sniffer)	Non dispersive infra-red instrument, single cell with multiple filters.	LI-COR 7200	0.1 s	0.3 ppm	Air
SO_2	Mixing ratio (sniffer)	Fluorescence (modified)	Thermo 43i-TLE	2 s	5 ppb	Air Fixed
NO_x	Mixing ratio (sniffer)	Chemiluminescence (modified)	Thermo 42i-TL	1 s	1 ppb	Air Fixed
SO_2	Column (optical)	Optical meas (DOAS)	Andor: Shamrock SR–303i , Newton 920BU	1 Hz	20 ppb over 50 m	Air
NO_2	Column (optical)	Optical meas (DOAS)	Andor: Shamrock SR–303i, Newton 920BU	1 Hz	20 ppb over 50 m	Air

The SO_2 analyzer response has cross sensitivity to NO. For example our laboratory test shows that 200 ppb of NO will cause a 3 ppb response in the SO_2 analyzer (Alfoldy, 2014). This may lead to an overestimation of the FSC by up to 0.1 % if not accounted for. To remove the influence of NO on the measurements, the NO_x species are measured in parallel to the SO_2 measurements. However, NO_x consists of the two gas species NO and NO_2 and the SO_2 analyzer only has a cross sensitivity to the former one. One therefore have to assume a certain ratio between NO and NO_2. Based on previous experience we first assumed that 80 % of the NO_x was present as NO but as part of the data analysis for this report we have found that this has caused a 20 % overcompensation of the NO interference which caused a negative bias in the FSC values of approx. 0.04 %. In the dataset described in this report the new compensation is used. The new modified calculation of FSC when including the new NO interference is the one given in Eq 2, and here it is assumed that 71 % of the NO_x is present as NO, The latter is based on measurements of the NO to NO_2 ratio at the great Belt bridge as part of this project, as shown in Figure 5.

$$FSC = 0.232 \frac{\int \left[SO_2 - SO_{2,bkg}\right]_{ppb} dt - 0.0098 \int \left[NO_x - NO_{x,bkg}\right]_{ppb} dt}{\int \left[CO_2 - CO_{2,bkg}\right]_{ppm} dt} \text{ [\%sulfur]} \qquad (2)$$

A second measurements artifact in the flight SO_2 instrument is caused by the absence of the kicker, as mentioned above. This applies for the airborne system for the full measurement period and the fixed system for the period June to October 2015. The kicker removes the influence of organic substances such as aromatic volatile organic carbons. Generally these species are not present to any larger extent in the flue gas of the ships but we later found out, through laboratory tests, that the instrument without kicker is also sensitive to other organic species, vapors or particles, present in engine lubrication oil and that these species seem to condensate easily in the tubing of the instrument. This is supported by a recent engine laboratory study (Eichler et al., 2015 and 2017) in which they performed advanced measurement of organic particles in ship emission exhaust gas which showed that the mass spectra of many particles in the exhaust gas are similar to the ones from condensed lubrication oil and that they consist of long chained cyclic alkanes (C_{20}-C_{25}) with low volatility. It is likely that these species also cause a response in the SO_2 fluorescence instrument.

In real measurements when not using a kicker, especially at the inlet channel of Göteborg (Mellqvist, 2017b) significant tails were observed in the SO_2 time series of the ship plumes which could cause false FSC values of up to 0.3 % m/m. We believe these tails were caused by organic condensable material. The problem is usually mitigated by excluding the tail of the plume in the calculation of the FSC. The measurements in Göteborg (Mellqvist 2017b) during two years shows that running the sniffer instrument with and without a kicker, respectively, causes a 0.07 % m/m positive bias of the median FSC value and a significant increase in the random uncertainty from ± 0.04 % m/m to ± 0.1 % m/m (Mellqvist 2017b). A kicker effect has been observed also in the Great Belt data during the period June to October 2015 when running without kicker, but too lesser degree than at the Göteborg site. The kicker effect is probably stronger at the Göteborg site since it is positioned where the ships are changing speed and this causes transient emissions with generally are high on particulates. For the airplane measurements carried out in this project there is little evidence of a kicker artifact in the statistics even though this instrument has no kicker. The reason for this is presumably because the ships are operated at steady state conditions and higher load when measured in the open sea and then particle emissions are usually lower than during low and transient operation. For instance, one of the ships that regularly showed high readings in the sniffer measurements at Göteborg in 2015 due to the kicker effect, have been sampled low with the aircraft on 3 different occasions on the open sea. This applies to several other ships as well.

In Table 2 several measurement factors causing errors in the data are discussed. Part of the details in the table can be found in others sections of the report.

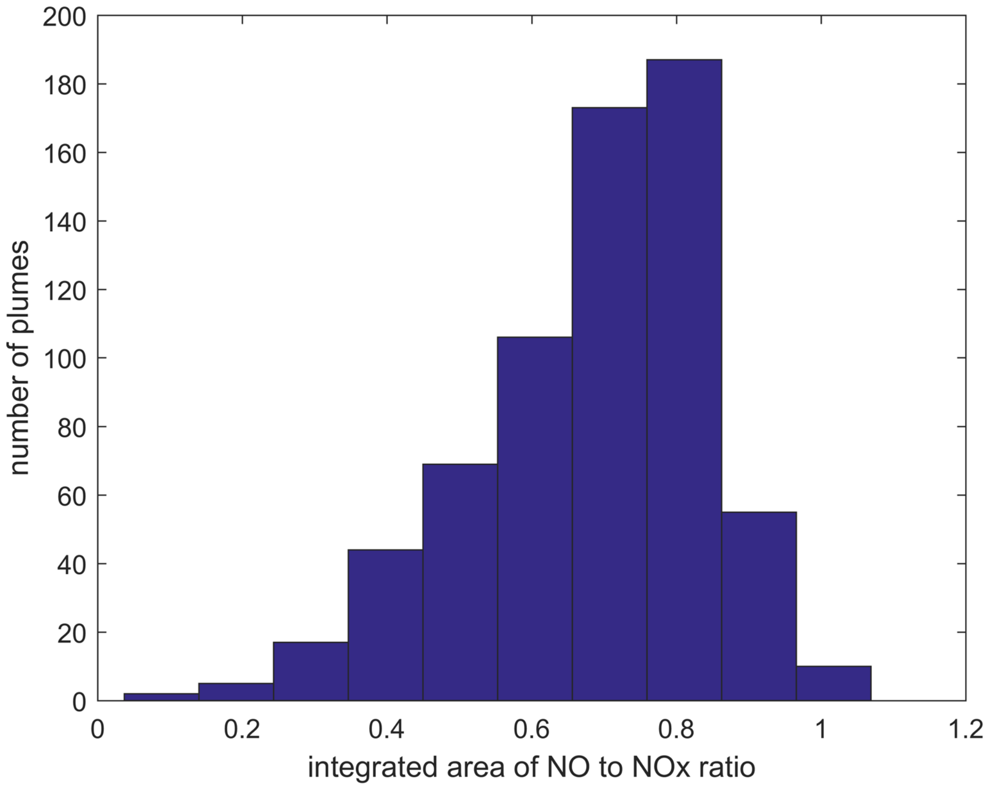


Figure 5. The measured ratio of NO and NO_x in ship plumes measured at the Great Belt bridge during January and February 2017, using two chemiluminiscence instruments. The median value corresponds to 0.71. The 10th, 25th,50th, 75th and 90th percentile values correspond to 0.44, 0.59, 0.71 , 0.80 and 0.86, respectively.

Table 2. The main error sources involved in the measurements are shown here.

Error source	**Description**	**Comment**
Correction for background	Done by statistical fitting of the baseline. This procedure is sometimes difficult when there is a variable background of CO2	Part of random noise.
Measurement noise	CO_2: 0.2 ppm SO_2: 2 ppb NO_x 1 ppb	Part of random noise but it is included in the quality flag assessment
Calibration gas uncertainty	CO_2: 0.5 % SO_2: 3 % NO_x : 3 %	Part of systematic uncertainty. Calibration certificate from gas manufacturing companies
Calibration interpolation error	Variation of instrument response between calibrations.	Part of random noise.
Cross interference i)	The SO_2 measurement is compensated for cross-interference with NO (0.98%). This is based on NO_x measurements assuming that 71% of NO_x is NO.	Part of measurement bias
Cross interference ii)	The fast responding SO_2 measurements (without kicker) exhibits skewed false SO_2 peaks presumably caused by organics particles	The effect is mitigated by using the first part of the plume.
Sampling error	Uncertainty when measuring short duration plumes (aircraft)	Test with a premixed gas shows a 13% precision and 10% accuracy
Sampling losses	SO_2 adsorption /absorption conversion on surfaces gas inlets, tubings and instrument.	Most measurements have a negative bias and this could be one of the causes.
Fuel carbon content uncertainty	Usually 87% is assumed	Causes 2% additional random uncertainty

2.4 General Uncertainty

In 2008, a joint study was carried out in Rotterdam with support from the EU (Alfoldy et al., 2011; Alfoldy et al., 2013; Balzani et al., 2014). The objective was to compare methods for the determination of FSC and NO_x emission factors based on remote measurements and comparison to direct stack emission measurements on a ferry. The methods were selected based on a review of the available literature on ship emission measurements and they were either optical (LIDAR, Differential Optical Absorption Spectroscopy-DOAS, UV-camera) or sniffer based ones. Using the latter method, three research groups participated with their own SO_2 and CO_2 instruments and one of the groups used a setup with double instruments measuring at different heights. Our group carried out both DOAS and sniffer measurements using an older instrument setup than the one used in this study (Mellqvist et al., 2010, Berg, 2011; Berg et al. 2012, Balzani et al. 2014). The measurements were performed from a land station, a boat and a helicopter together with on board measurements. It was found that the sniffer approach is the most convenient technique for determining mass specific emission factors of both SO_2 and NO_x remotely. The overall estimated uncertainty for SO_2 was 23 % (Alfoldy et al. 2013) at 1 % m/m FSC, based on comparison with on board sampling. In Figure 6 results are shown from a comparison between the Chalmers sniffer system measuring from a 3 m mast to a similar sniffer system by the Joint Research centre (JRC-Ispra) who ran measurements on a 20 m mast. There is a clear correlation between the two systems although there is a 20% systematic difference, similar to the estimated uncertainty. In another study (Beecken et al. 2014a) the measurement precision was estimated from the variability of multiple airborne measurements of the same ship, for a total of 158 different ships. A random uncertainty of ± 0.19 % m/m was obtained for ships with approx. 1 % m/m FSC.

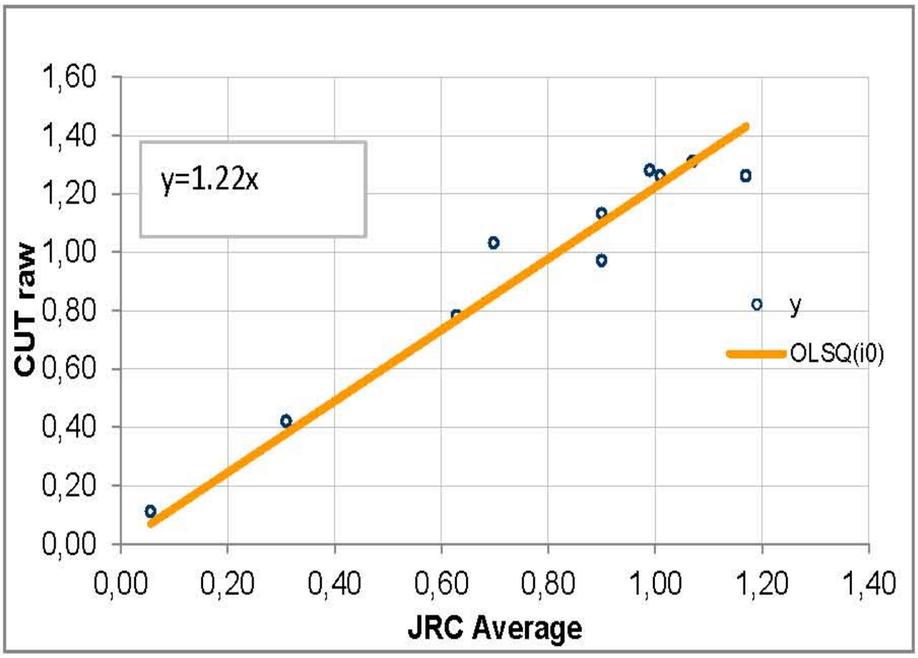


Figure 6. Field measurements in Rotterdam measuring individual ships during two days in the ship channel of Rotterdam (Balzani 2013). Two nearby systems, the Chalmers sniffer system(CUT) and the system developed by the Joint Research centre (JRC-Ispra) were compared.

3 Measurements

3.1 Fixed system

3.1.1 Installation

The fixed sniffer system was installed at the eastern pylon at the Great Belt Bridge, Figure 7. This is a very good measurement spot in view of the large volume of marine traffic (25000 ships per year) and predominant south westerly wind conditions.

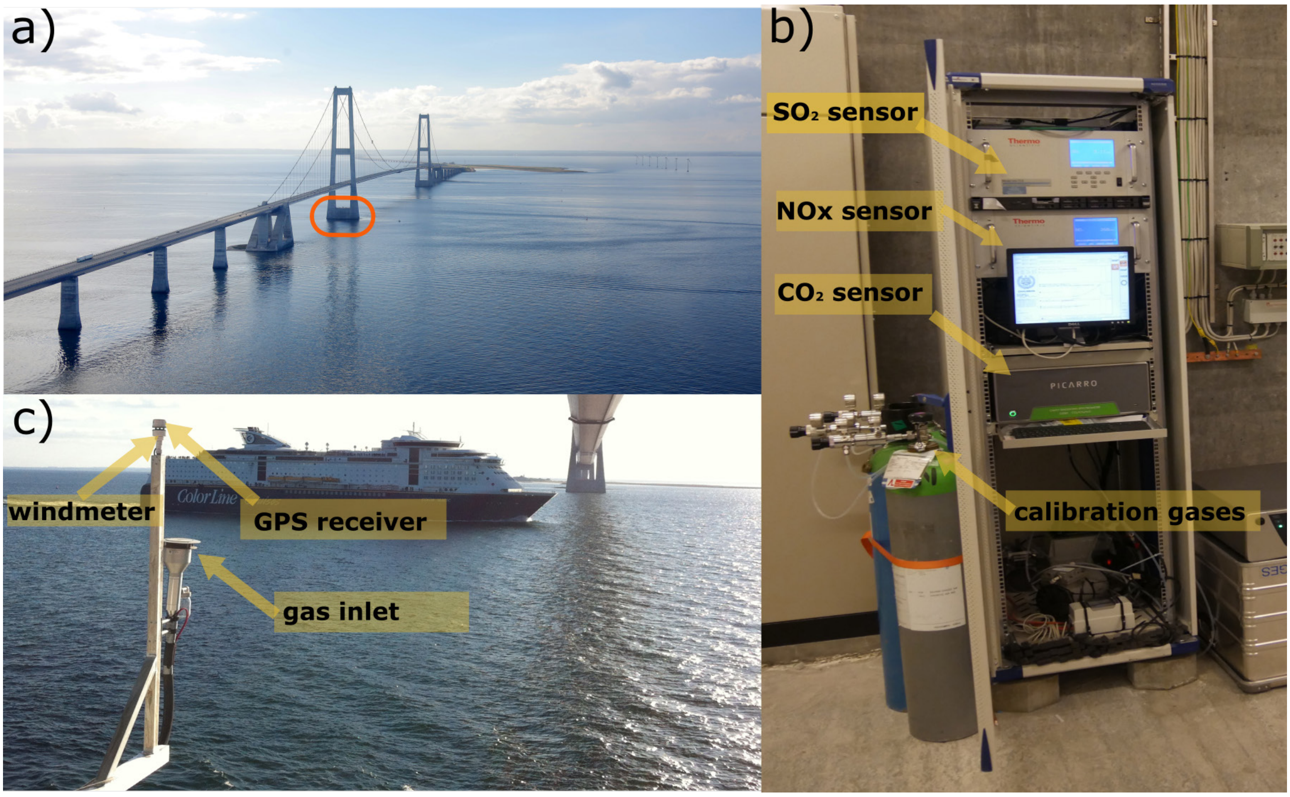


Figure 7. Fixed sniffer system installed at The Great Belt Bridge, Denmark. (a) Installation site at the Pylon 16 of the bridge. (b) Instrument rack inside the Pylon. (c) The GPS receiver and wind sensor mounted on a metallic angle structure. The gas inlet and the AIS antenna are in the same metallic angle (not showed on the picture).

The gas sensors and its components were installed in a rack inside the control room at the eastern Pylon (#16) of the bridge, while the AIS antenna, GPS receiver and inlet where mounted on a metallic angle just outside the bridge. The system has it's independent internet link through a 4 G modem.

The gas was extracted via a 10 meters long heated Teflon tubing that was connected to a metallic sampling inlet used as rain protection during the first year and then a U bent Teflon tube ending with a plastic cone the 2nd year, The total flow speed was approximately 12 lit/min. The sensors were regularly calibrated (every 5th day) by injecting reference gases through a 10 m gas tube that was connected in the beginning of the sampling line close to the main inlet to the sniffer system.

3.1.2 Data acquisition system and web data reporting

The optical and sniffer data are handled by a Data Acquisition System (DAS) which is a combination of three custom made software applications running unattended and continuously: *TCPlog*, *IGPSpresent* and the *IGPS mailer*.

The software *TCPlog* has the most critical task which is continuously logging all the available instruments with a sampling period of approximately one second. This includes data from the sniffer and optical sensors, wind meters, AIS receiver and in case of the airborne platform also information from the aircraft.

The *IGPSpresent* program analyses the data in near-real time, namely calculating the FSC through ratio measurements between the concentrations of SO_2 and CO_2. Moreover, the IGPSpresent identifies the presence of ship plumes and its corresponding source of origin. For the fixed station the program initiates a calibration every 5th day.

Finally, the *IGPSmailer* program automatically sends evaluated and compiled measurements to the database at Chalmers University of technology, see an example in Table 3 from the Älvsborg site in the ship channel of Göteborg obtained in the Compmon project. The database includes the FSC values as well as date, time, position and ship specific data.

The DAS also generates alerts as emails or SMSs when a high emitter ship has been detected, or when there is a possible system malfunction. These alert messages combined with regular remote logging, has been of key importance to ensure reliable measurements. .

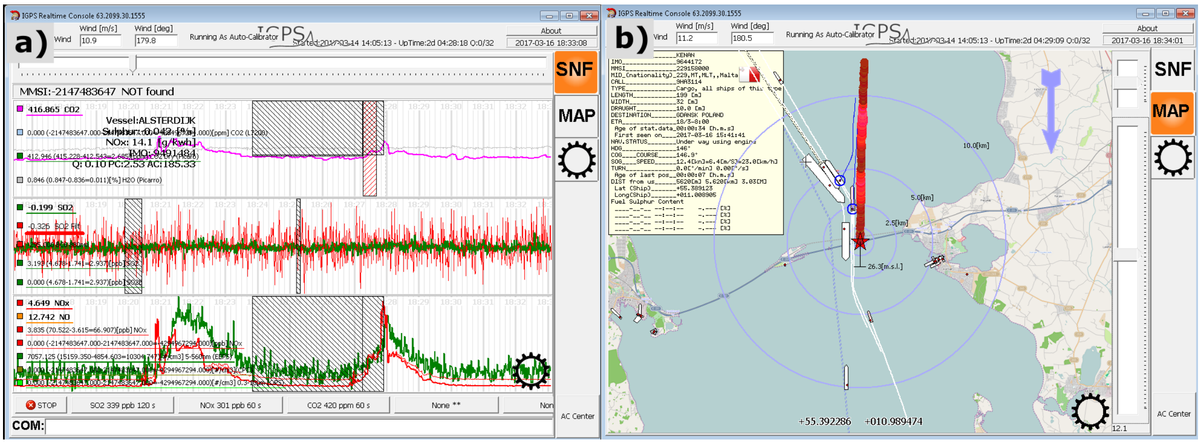


Figure 8. Example of the IPGSpresent software while performing a measurement at the Great Belt bridge. (a) Real-Time series of CO_2, SO_2 and NO_x concentrations. (b) Identification of plumes from the nearby ships.

Table 3. Example of the data base setup from the Göteborg site at the Älvsborg island (Mellqvist et al., 2017b). The data at the Great Belt Bridge are stored in the same manner.

Date	Record type	Value	Quality tag	Quality control	Platform	Platform type	Ship name	Ship type	IMO	MMSI	Ship coordina
03/09/2016 - 10:56	SC	0.00	Poor	Automatic	Älvsborg	Stationary	HANS	Other Type	8996956	230034540	11.848055,57
03/09/2016 - 10:49	SC	0.06	High	Automatic	Älvsborg	Stationary	ICE STAR	Cargo	9142631	244264000	11.846465,57
03/09/2016 - 10:25	SC	0.02	Medium	Automatic	Älvsborg	Stationary	GEORGE	Other Type	8634077	230034510	11.846706,57
03/09/2016 - 09:44	SC	0.04	Medium	Automatic	Älvsborg	Stationary	STENA DANICA	Passenger	7907245	265177000	11.825803,57
03/09/2016 - 09:18	SC	0.02	Medium	Automatic	Älvsborg	Stationary	ASTINA	Tanker » Hazardous category A	9320063	266220000	11.850048,57
03/09/2016 - 08:56	SC	-0.15	Poor	Automatic	Älvsborg	Stationary	DELFIN	Tanker	0	265505100	11.853978,57
03/09/2016 - 08:44	SC	0.02	Poor	Automatic	Älvsborg	Stationary	STENA SCANDINAVICA	Passenger » No additional information	9235517	266343000	11.860925,57
03/09/2016 - 08:40	SC	-0.04	High	Automatic	Älvsborg	Stationary	STENA SCANDINAVICA	Passenger » No additional information	9235517	266343000	11.843681,57
03/09/2016 - 08:17	SC	0.07	High	Automatic	Älvsborg	Stationary	THUN GLOBE	Tanker	9229051	245573000	11.834666,57
03/09/2016 - 08:08	SC	-0.02	Medium	Automatic	Älvsborg	Stationary	HANS	Other Type	8996956	230034540	11.837226,57
03/09/2016 - 07:51	SC	0.01	Medium	Automatic	Älvsborg	Stationary	LEXUS	Tanker » No additional information	9310317	219157000	11.856116,57

3.1.3 Hardware changes and practical problems

The fixed station at the Great Belt Bridge was gradually modified during the project due to malfunctioning instruments and observed measurement artifacts, as shown in Table 4.

Tails in the SO_2 plume measurement data were occasionally observed during the period June to October 2015. These, we believe, were caused by the kicker effect explained in section 2.3 and when calculating the apparent FSC several ships were actually above the compliance limit threshold, see section 4. To mitigate this problem we installed a kicker in the instrument on October 2015 and this removed the problem with tails.

To prevent water droplets and salt to enter the sniffer tube system, a metallic sampler was used as inlet, see Figure 7. This is an inlet with anodized aluminum which bends the air in such a way that only gases and small particles below 10 µm in size can enter. Even though aluminum is considered a poor material for gas sampling it was considered that the inlet would have little impact on the sampling of the gases given the high air flow combined with relatively little wall surface in the inlet. Several ships with high FSC were found in the beginning indicating that the system was working. However, suspicion were raised that there was a problem with the inlet, after comparing airborne and fixed measurements. We therefore acquired a premixed high concentration calibration gas of SO_2 and CO_2 which made it possible to "puff" the gas outside the inlet. The test showed that the probe reduced the SO_2 and the inlet was therefore changed to U bent Teflon tubing with a plastic funnel at the end. The main calibration did not pick this up since it could only be done by injecting calibration gas after the inlet. See implication in the section 4.3.

Table 4. Technical adjustments at the fixed measurement site.

Day	Change
June 16, 2015	Installation and start of measurements
Oct 16, 2015	Installation of hydrocarbon kicker in SO_2 instrument to reduce interference from aromatic VOCs, reduces noise of FSC and a positive bias of about 0.1% in FSC
Nov 20, 2015	Installation of pump for faster flow through inlet tube
May 27, 2016	Change of inlet from aluminum to plastic to reduce adsorption of SO_2 on probe
Dec 1, 2016	SO_2 pump broke during this month
Jan 16, 2017	Installation of sniffer without HC-kicker while other SO_2 system was taken for service
Jan 24, 2017	Installation of a new SO2 instrument with hydrocarbon kicker

3.1.4 Calibration

The quality assurance of the fixed sniffer instruments is obtained by repeated calibrations with about 5 days in between. The instruments at the Great Belt installation site were remotely calibrated using gas standards diluted in nitrogen with values ranging 200-450 ± (5 %) ppb, 210-300 ± (5 %) ppb and 380-420 ± (1 %) ppm for SO_2, NO_x and CO_2 respectively.

The calibration gas was injected just after the measurement inlet. Figure 9 shows the time series of the software calibration response which shows that the CO_2 has a stable response versus time while SO_2 and NO_x showed larger drift of a few percent per month.

In most cases the instrument were not recalibrated and instead the output from the instruments was validated and post-corrected using the calibration factors. However, when the instrument response deviated too much from the nominal value a hardware recalibration of the instrument was carried out. This can be seen as the sharp variations in the time series in Figure 9.

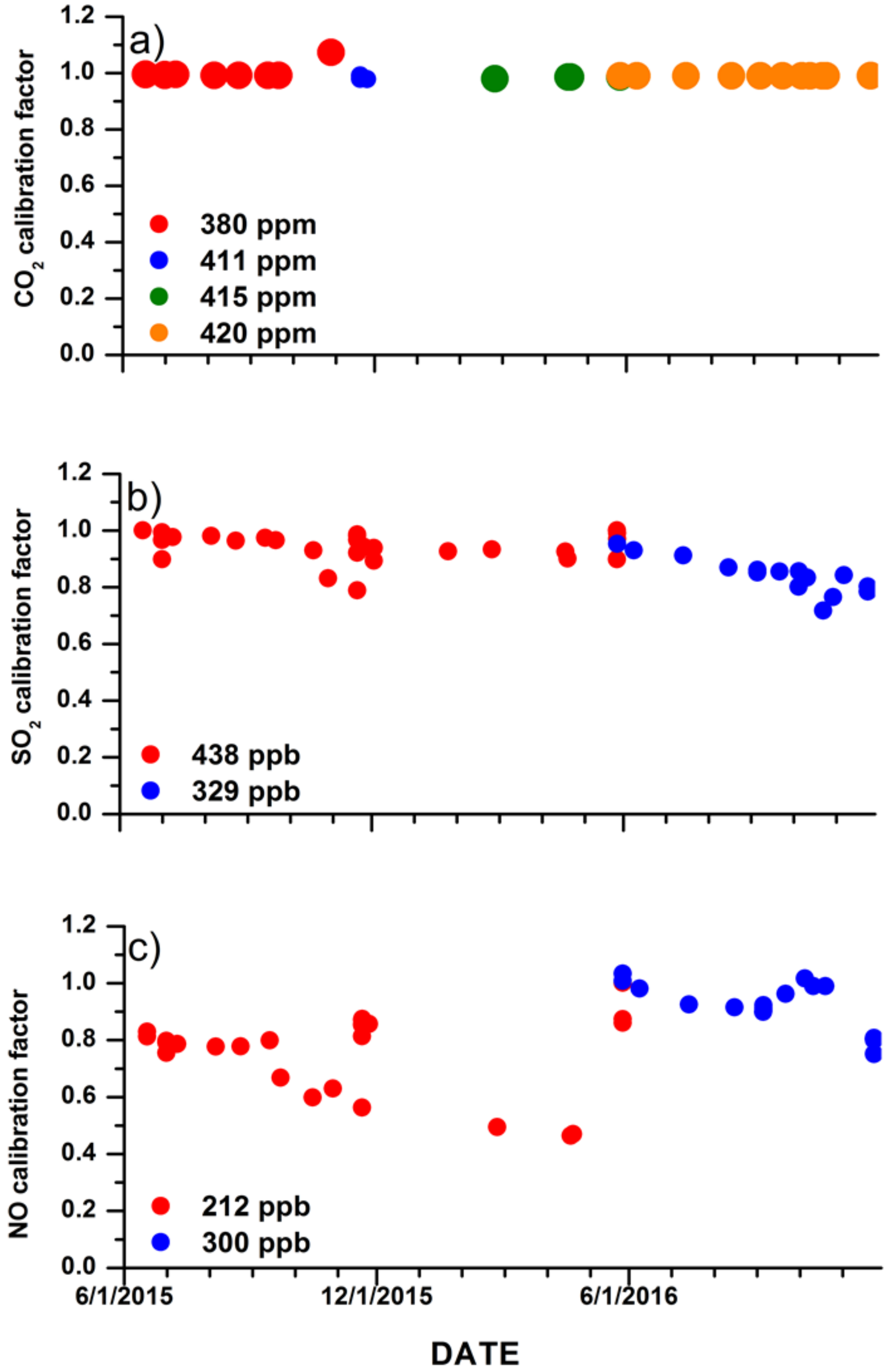


Figure 9. Time series of the correction factors obtained from calibration of the sniffers system at the Great Belt bridge: (a) CO_2, (b) SO_2 and (c) NO_x. The different colors correspond to different reference gases.

3.1.5 Quality assessment of data

In the data evaluation the quality of the measurements is expressed through a quality flag that can alternate between the following levels: HIGH, MEDIUM and POOR. This assessment is based on the parameters in Table 5 for the fixed station at the Great Belt Bridge. As can be seen in the table the quality flag is a combination of measured parameters such as CO_2 peak signal and empirical observations of conditions when the measurements are more certain. One important parameter here is the comparison of CO_2 in the ship plume against the variation of the ambient background CO_2, which comprises both variations of the background (upwind fixed source like a city) and the noise of the instrument. The quality level may also shrink if different hardware warning flags are raised while the instruments are operating. These flags are mostly associated to issues related to abnormal temperature, low voltages, flow interruptions, etc. In general the automatic data retrieval performed satisfactorily for high and medium quality measurements while only few of the poor quality data could be upgrade to medium quality after manual inspection.

Table 5. The quality criteria applied for the fixed measurements at the Great Belt bridge during 2015 to 2017. Some of the criteria suggested for future use are also given.

Criteria	Comment	High	Medium	Poor
Instrument operation	Warning flags for the hardware not set, such as high/low temperature, low voltages etc	Required	Required	Depends
$\Delta CO_{2\ peak}$	Peak height	>3 ppm	>2 ppm	0.5 ppm
$\Delta CO2_{plume}$	Integrated amount	>50 ppms	>25 ppms	3> ppms
$\Delta t_{CO2\ plume}$	Time duration in plume	<100 s	<150 s	<240 s
Wind direction	Wind relative to ship movement	± 30^o	± 60^o	± 60^o
Wind speed		3 - 8 m/s	2 -10 m/s	1 - 12 m/s
No of ships with overlapping plumes		1	1	1
FSC	Filtering out low values	>-0.2	>-0.2	>-0.2
ΔSO_2 in plume	Peak height	NA	NA	NA
ΔSO_2/ (1.5%*ΔNO)	Interference effect, If interference dominates uncertainty increases	NA	NA	NA

3.2 Airborne

3.2.1 Installation

A Piper Navajo stationed in Roskilde, operated by the company Aircraft Aps, is equipped with sniffer and optical equipment for ship surveillance measurements, Figure 10. The installation, including modification of the aircraft and the attachment and functioning of the instruments has been certified by the European air safety agency (Supplemental Type Certificate 10051623, European Air Safety Agency). This includes for instance an electromagnetic interference test based on the standard RTC DO 160/issue M/cat M/section 21 and section 15 (Mellqvist et al., 2014; Mellqvist et al., 2017a).

The flight sniffer system is a compact version of the previously described fixed system, but it is pressure compensated and the SO_2 fluorescence system is much faster responding but more sensitive to interfering species since it runs without kicker. Most of the system has been rebuilt into a common module, Figure 11, in a 19” wide box which holds a logging computer, a Thermo fluorescence instrument for SO_2, a LI-COR non dispersive infrared instrument for CO_2, an AIS, a GPS receiver and a power converter. The weight of the system is 47 kg and the power consumption is 15 A at 28 V DC.

The measurement strategy consists of performing optical measurements at about 200 m altitude and if these measurement indicate high FSC values a sniffer measurements through the plume is carried out at about 65 m altitude, Figure 12. Under good weather conditions it has been possible to measure 4 to 8 ships per hour in the waters around Denmark, mostly depending on the traffic density. Using the same equipment the Belgian institute MUMM is however able to measure up to 12 ships per hour in the English Channel where the traffic is even more dense (https://compmon.eu/reports)

The airborne data acquisition system is similar to the one described in section 3.1.2. The real time program *IGPSpresent* is an essential part of the flight operation since it is used to guide the aircraft and for realtime analysis of the FSC, Figure 12. The data can be sent directly to the web server and alerts can be sent out in the same way as for the fixed system, given that the aircraft is connected to internet. During this project this was not the case and instead the data were reported within a week, with exception for non-compliant ships which were reported within the same day of the flight.

Figure 10. Airborne measurements system: (a) Sniffer installation in the airplane cabin, (b) Spectrometers installed in the airplane cabin with their corresponding telescopes on the upper side, (c) Metallic structure holding the telescopes of the optical system and the gas inlet.

Figure 11. The central part in the measurement system corresponds to a 19" wide sniffer box including a logging computer, a UV fluorescence instrument for SO_2 , a NDIR for CO_2, and a AIS and GPS receiver and power converter (47 kg, 15 A at 28 V DC).

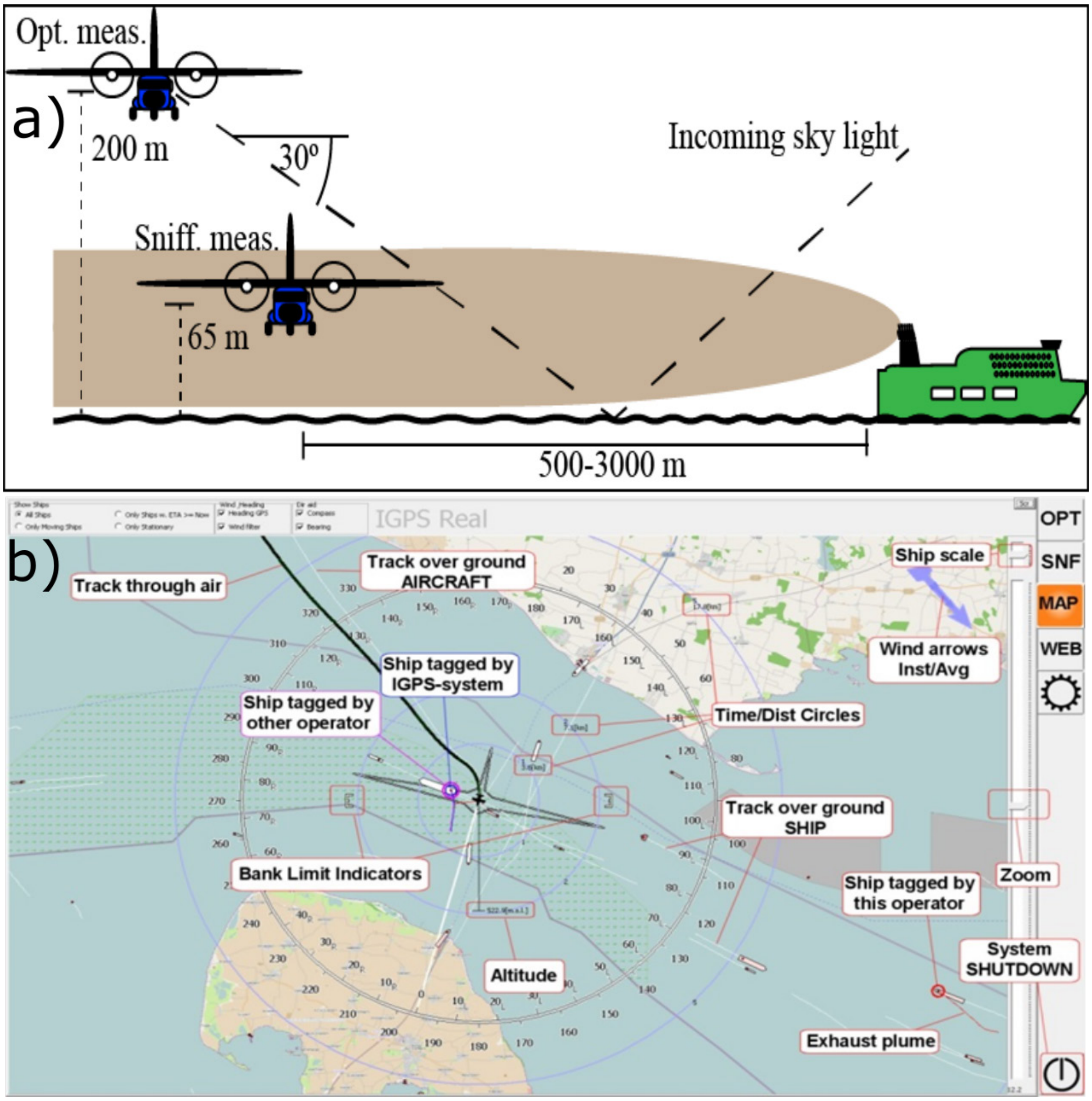


Figure 12. Airborne measurements strategy. (a) The optical measurements require flying above the ship plume at a around 200 m above the sea. The sniffer measurements require flying inside the plume at 65 m. (b) The airborne version of the IGPSpresent.

3.2.2 Calibration

The sniffer and optical instruments were calibrated before each flight mission on the ground, after preheating of at least an hour.

In Figure 13 a picture is shown where premixed calibration gas was flushed in front of the gas inlet using Teflon tubing. In addition can be seen a validation exercise of the optical measurement in which gas cells filled with SO_2 and NO_2, respectively, were held in front the optical telescopes.

The wavelength setting and the instrumental line shape of the optical instruments were calibrated before every measurement day using a mercury lamp. The sniffer instruments were calibrated against premixed gas standards with a typical accuracy of a few percent. The typical gas concentration values for SO_2, CO_2 and NO_x were 401 ± (3 %) ppb, 370 ± (0.5 %) ppm and 191 ± (3 %) ppb, respectively. The gas standard of NO_x was diluted in nitrogen while the other gases were diluted in synthetic air. From the calibration the correction factors were obtained which were used to post correct the flight measurement. In addition, to the standards above we used a multigas calibrator and zero air generator (Thermo 146i and Thermo 1160) together with more stable mixtures of high concentration calibration gases from AGA Special gas AB corresponding to 101 ± (0.5 %) ppm for both NO and SO2 gases. These calibrations were done a few times each year to check the stability of the calibration gases, and to bridge the gap when switching gases. In Figure 15 the obtained and used calibration factors are shown for all flights.

Figure 13. Quality control of the sniffer and optical sensors on the Navajo Piper from Aircraft Aps. The yellow plate includes two windows for optical sensors, a window for a video camera, two inlets for gases and particles, respectively, and one exhaust pipe. The optical system is checked by holding gas cells filled with known concentrations of SO_2 and NO_2 in front of the telescopes. The sniffer system is calibrated by flushing premixed calibration gas in front of the gas inlet using a Teflon tubing.

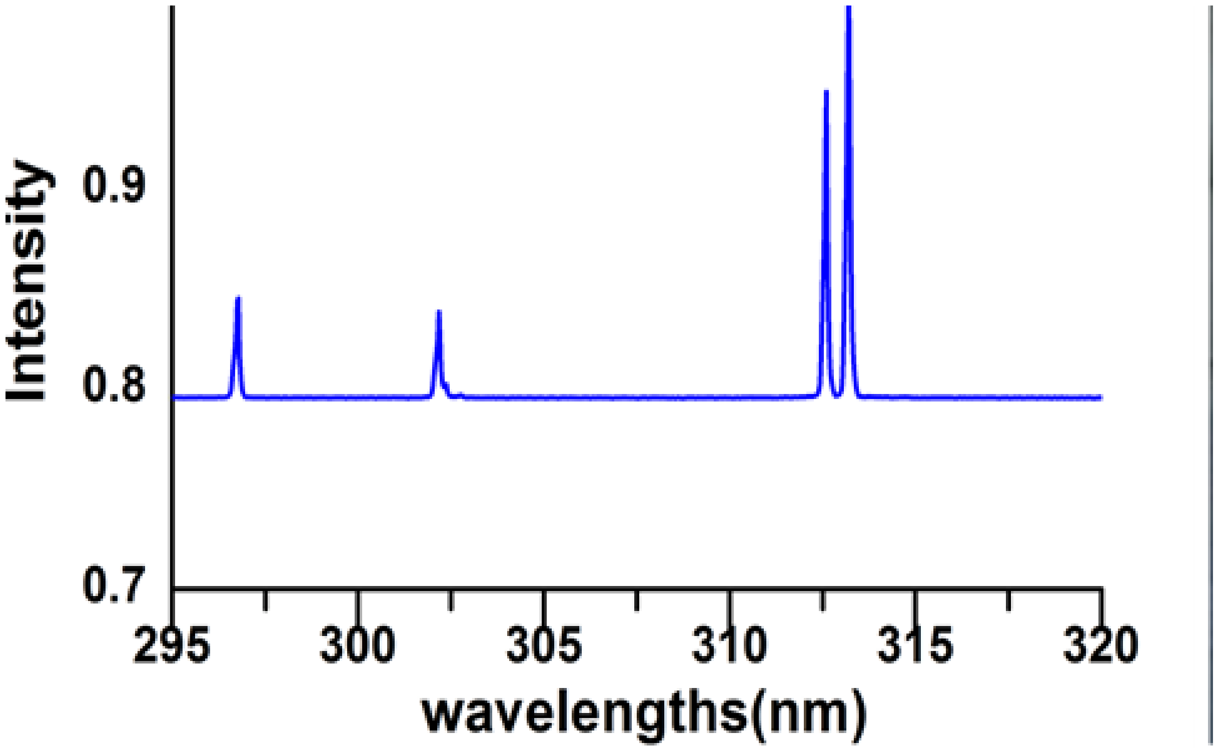


Figure 14. Calibration of the wavelength scale and line shape response function of the grating spectrometer used for the SO_2 measurements.

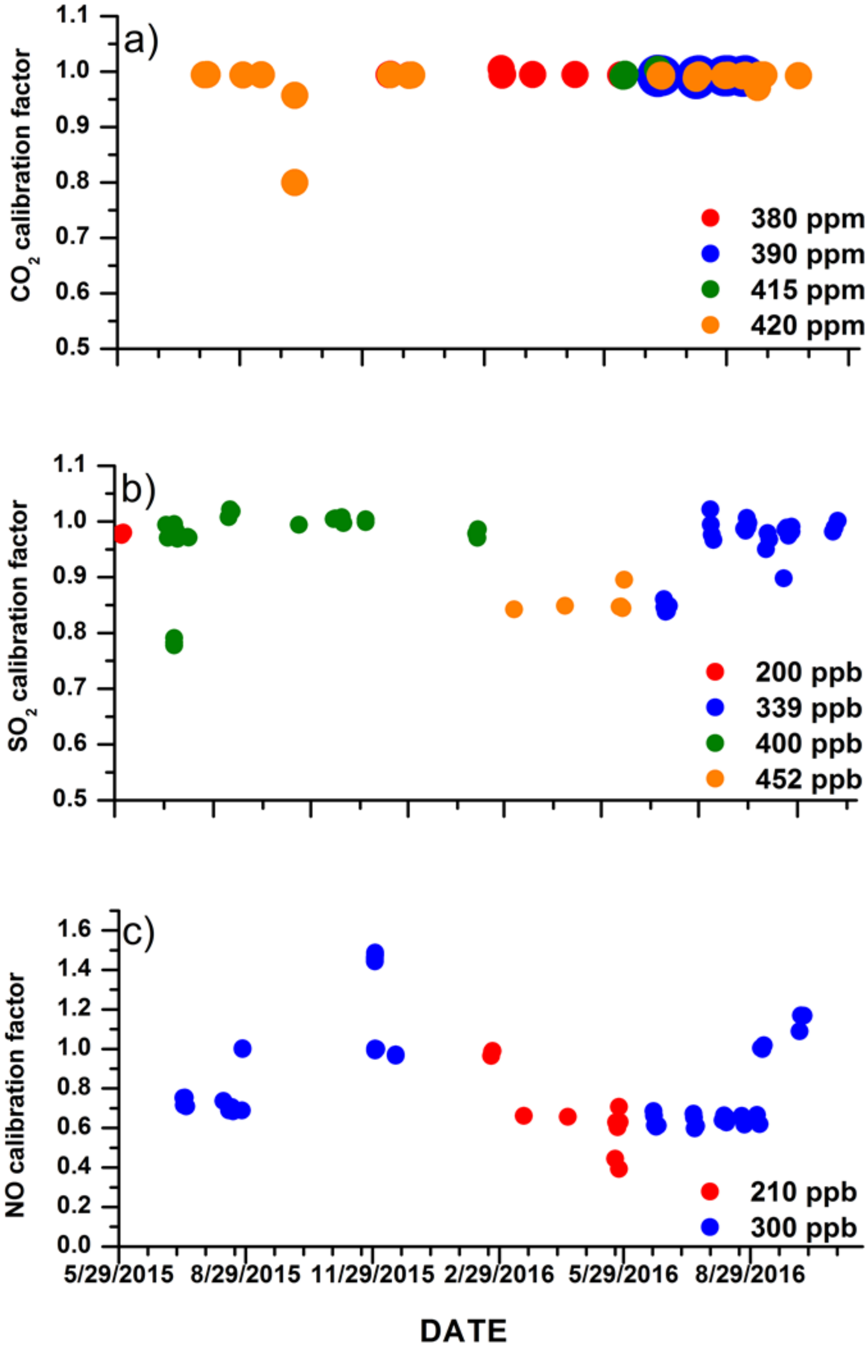


Figure 15. Time series of the sniffer calibration response for the airborne system. (a) CO_2 calibrations. (b) SO_2 calibrations. (c) NO_x calibrations. The different colors correspond to different reference gases.

3.2.3 Quality assessment of data

The airborne data in this project have been evaluated manually with an assessment of the measurement quality based on the quality criteria in Table 6.

Table 6. Quality criteria applied for the airborne measurements. Some of the criteria suggested for future use are also given.

Criteria	Comment	High	Medium	Low
SNIFFER				
Normal operation	Warning flags for the hardware not set, such as high/low temperature, low voltages etc	Required	Required	Depends
Preheating	Preheat instrument 2 h before departure	Required	Required	Required
Calibration	1 h before departure. Check that difference in data correction factors are within 20 % of nominal value; if so change the calibration parameters of the instruments.	Required	Required	Required
ΔCO_2 in plume	Peak height	>4 ppm	2-4 ppm	1-2 ppm
Δt_{CO2} in plume	Time duration in plume	>3 s	>2 s	>1 s
$\Delta CO_{2,plume}/stdev(CO_{2bkg})$	Peak signal above background noise (standard deviation)	4	3	2
ΔSO_2 in plume*	Peak height	>4 ppb	2-4 ppb	1-2 ppb
ΔSO_2/ (0.098%*ΔNOx)	*Interference effect, If interference dominates uncertainty increases*	*NA*	*NA*	*NA*
$\Delta t_{SO2}/\Delta t_{CO2}$	Skewness of plume, compared to CO_2 measurement. In all cases we reduce this effect by using only the time period with CO_2 plus 2 s	<2	2-3	3-5
No of ships with overlapping plumes		1	1	1
Wind speed		3-8 m/s	2-10 m/s	1- 12 m/s
OPTICAL				
$\Delta NO_{2,optical}$		>10 mg/m^2	>8 mg/m^2	>7 mg/m^2
SNR SO2	(ΔSO_2/stdev(SO_2_baseline))	3	2	2

4 Results

In this section we describe the fixed measurements at the Great Belt Bridge and airborne ones from the Navajo Piper aircraft and we provide a discussion of the measurements uncertainty.

4.1 Fixed site measurements at Great Belt Bridge

The main time period of this project corresponds to June 15, 2015 to January 31, 2017. However, since the sniffer measurements at the Great Belt Bridge were continued also after 2016, the data set here is further extended to May 31, 2017 in some of the analyses. We have also excluded the period June 2015 to May 2016 from the statistical analysis since the sniffer then had reduced sensitivity, as explained in section 4.3. During the period June 2015 to January 2017 the system was in operation during 480 days out of 590 days. In addition, local weather conditions only made it possible to measure ship emission plumes during 60 % of the reported time span. The obtained data set, divided into 3 qualities, corresponds to 11161 ship plumes. In the upper panel of Figure 16 is illustrated the time periods when the wind was appropriate for measurements and when the instruments were working in a sufficient way. In the lower panel of Figure 16 the FSC data is plotted with color coded measurement quality. The statistics shows that 56 % of the ship measurements had high or medium quality, Figure 17.

In Figure 18 the frequency distribution of all FSC measurements at the Great Belt Bridge between June 2016 to May 2017 is shown, corresponding to 3675 individual ship measurements of good or medium quality. Here a ship is counted twice if the measurements are conducted on separate days. The reason for not including the first period in this analysis, i.e. June 2015 to May 2016, is because the gas inlet caused measurements artifacts which reduced the sensitivity of the instrument during this period, see section 4.3. The measured FSC data has a median value of 0.025 % m/m. By analyzing the variability of 30 different individual ships that were measured on more than 9 occasions we estimate that the random noise (precision) of the measurements follows a Gaussian distribution with a width corresponding to a standard deviation of 0.04 % m/m in FSC units. Here it is assumed that the FSC was constant for each individual ship and since the sulfur content of fuel deliveries may vary there is no guarantee that the same ship will have the exact same sulfur emission each time it passes the sniffer. The measured variability is for this reason *our best upper estimate of the real precision* of the sniffer measurements. In Figure 18 the random noise distribution, centered at the measured median FSC value of 0.025 % m/m is shown. It can be seen that the experimental data follows this curve rather well supporting that our estimate of the random noise is reasonable. The median FSC value of the sniffer measurements, 0.025 % has been be compared to 756 on board measurements by the Swedish and Danish port state control authorities for the same time period showing that the sniffer measurements appears to have a negative bias of 0.055 % in FSC units. From the derived bias and precision above is estimated that the threshold for non-compliance is 0.14 % m/m in the Great Belt Bridge sniffer data set. This is further explained in section 4.3.

In Figure 19, a histogram is shown of the the number of ships with different FSC levels for the Great Belt Bridge sniffer data corresponding to measurements between June 2016 to May 2017. The data for the high FSC ships is highlighted. The data for medium and high quality is indicated and all negative data points were assigned a FSC value of 0.00 %. In Figure 20 the fraction of ships below a certain FSC level are shown for the period June 2016 to May 2017. The fraction below the compliance threshold of 0.14 % m/m is 95.5 %. Hence 4.5 % of the ships are running on non-compliant fuel with a confidence limit of 95 %. Note that the threshold is corrected for the measurement bias (biased threshold) as discussed in section 4.3.1. The ships using FSC above 0.5 % m/m here corresponds to 1 % of the total fleet. As discussed in section 5.1 the compliance appeared to be lower during 2016 than 2015. During 2015 and 2016 the compliance rate from onboard sampling by port state controls authorities was 95 % in both Denmark (*pers comm Dorte Kubel, Danish EPA*) and Sweden (*pers comm Caroline Petrini, Swedish transport agency*).

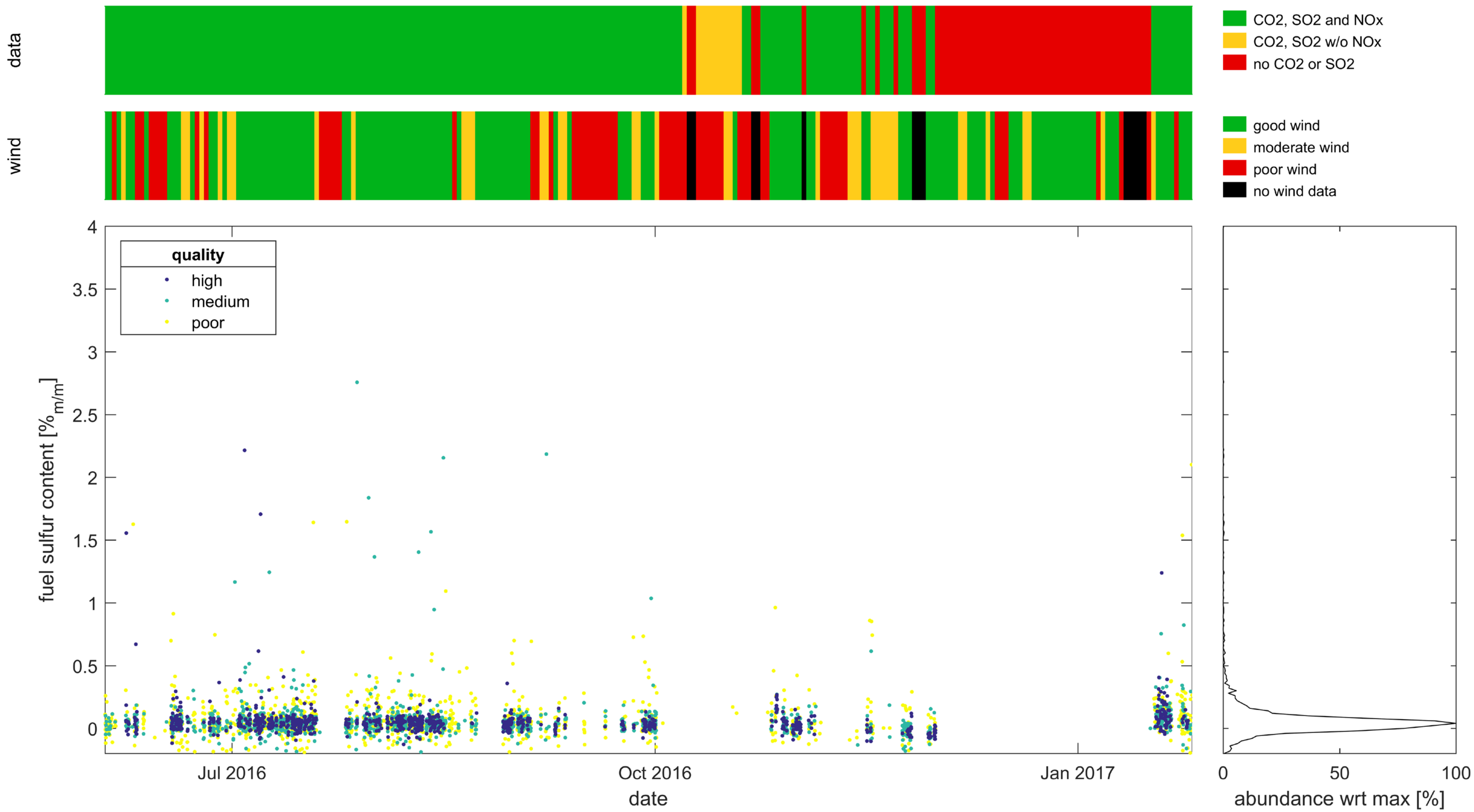


Figure 16. Measurement statistics (4367 ship plumes) at the Great Belt between June, 2016 to January, 2017. (a) measurements rate and wind variability through time. (b) FSC distribuition over time with coding of different qualities. The negatives FSC values correspond to NOx compensation when sulfur values where below the detection limit. In the bottom right is shown frrequency distribution of the measurements versus FSC.

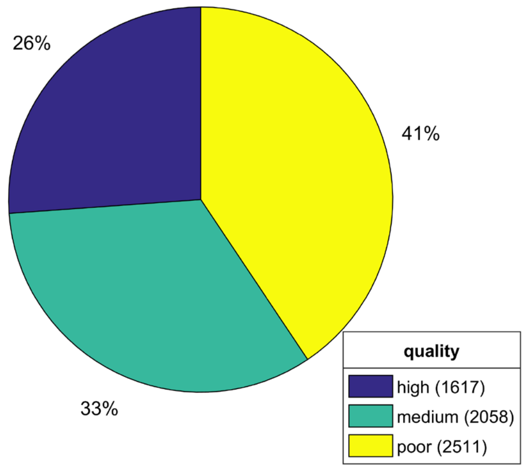


Figure 17. Statistical distribution of measurement quality for the sniffer data acquired at the Great Belt Bridge and number of ships for the period June 2016 to May 2017.

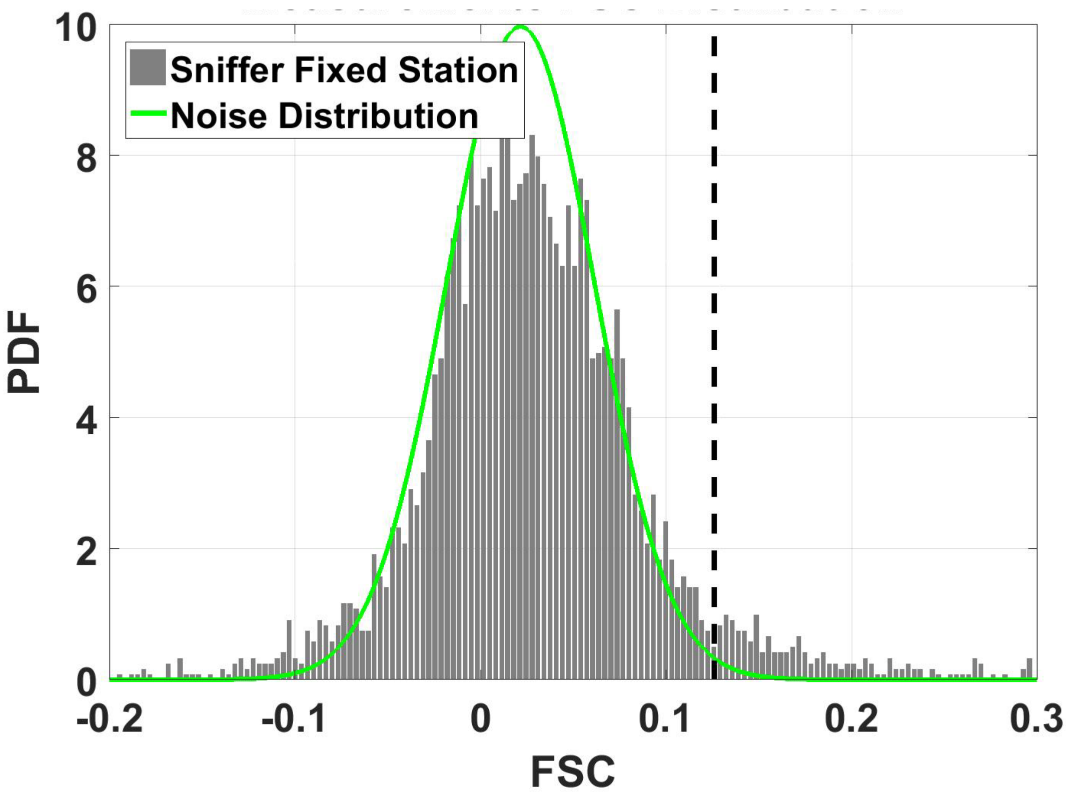


Figure 18. Statistical distribution (probability density function) of the FSC corresponding to 3675 individual ships measured with sniffer at the Great Belt Bridge. The data covers the period June 2016 to May 2017. The green curve corresponds to the random noise distribution (precision) of the measurements obtained from multiple measurements of single ships. The dotted line is the estimated non-compliance limit for which the instrument errors (precision and bias) have been accounted for.

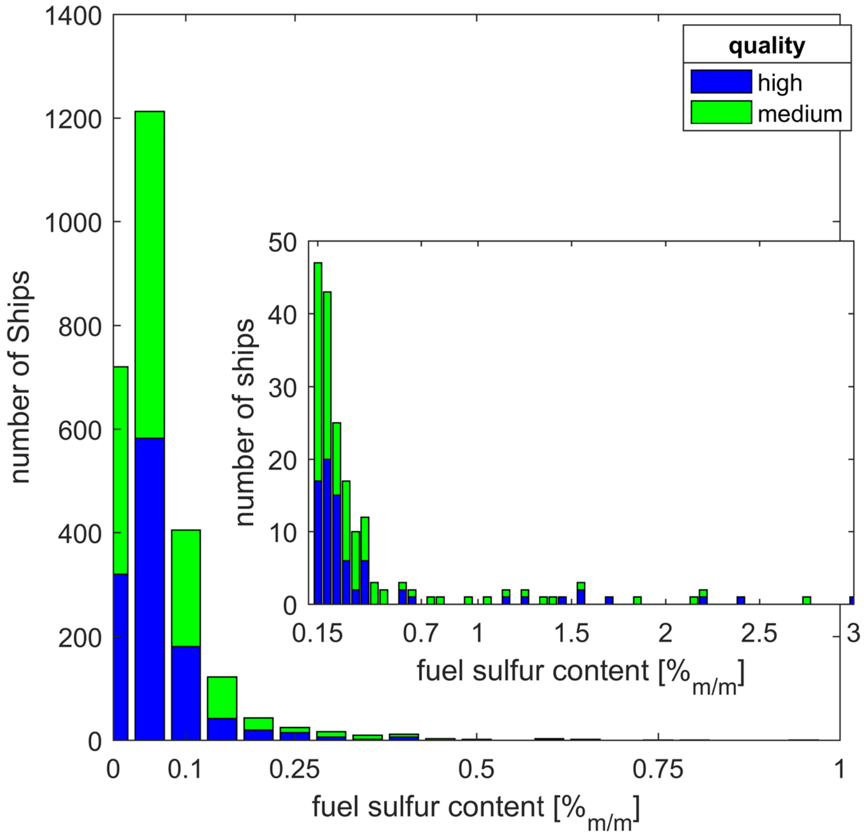


Figure 19. Histogram of fuel sulfur content shown for different measurement qualities. In the inset the distribution of FSC above 0.15 % m/m is highlighted. The data correspond to 3675 ships measured between June 2016 to May 2017.

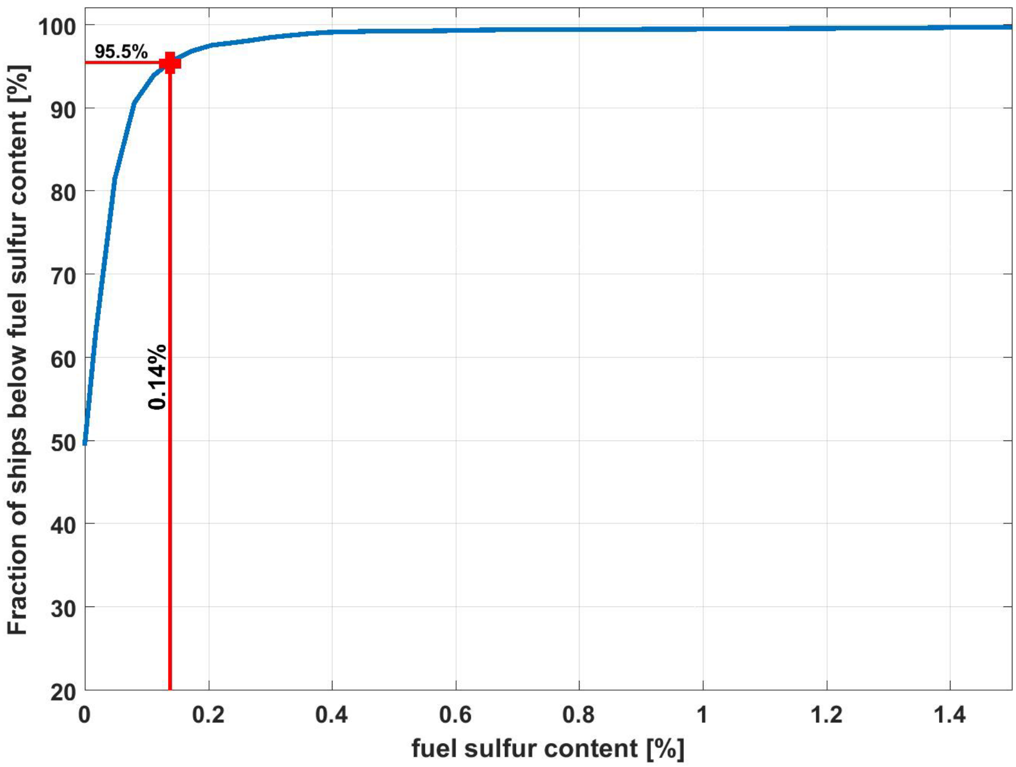


Figure 20. The fraction of ships that were measured below a certain FSC level at the Great Belt Bridge for the period June 2016 to May 2017. In addition the biased compliance limit threshold is shown, indicating a compliance level of 95 %. Here we have changed the threshold to account for the measurement bias.

4.2 Airborne Measurements

Airborne surveillance of individual ships on Danish waters was carried out in the period July 2015 to October 2016 using the Navajo Piper aircraft stationed in Roskilde (Figure 10) equipped with both sniffer and optical sensors. The measurement statistics are shown in Figure 21, with actual data with quality flag and frequency distributions in the bottom. In total, 245.5 flight hours were carried out and they were distributed over 58 airborne missions with duration of about 4 hours each, see appendix I. The airborne dataset constitutes a total of 947 sniffer measurements and 741 optical measurements of ship plumes.

The spatial distribution of the airborne data is shown in Figure 22 corresponding to measurements of individual ships using both sniffer and optical sensors. The obtained FSC values are color coded: green (<0.16 % m/m), yellow (0.16 - 0.3 % m/m) and red (>0.3 % m/m) and the letters A - D correspond to 4 different areas that were monitored. Measurements marked in yellow and red are above the compliance threshold, as discussed below. As can be seen, there is no apparent pattern in the geographical location of these measurements.

In Figure 23, the number of measured ships per hour is shown for the different flight missions in 2016. The average number of ships measured per hour combining the sniffer and optical sensor was 6 ships per hour through varying climatic conditions and seasons, with about 7 ships per hour in good conditions. Here the most important parameter was the density of ships. In the case of only sniffer measurements with high or medium quality 3.5 ships per hour were measured. Since all flight measurements were started at Roskilde airfield the measurements were less effective in area D and C compared to measuring area A and B, since the same ships were passed twice in the former one. The measurements were less efficient at strong wind speeds but also in weak wind speeds it was sometimes difficult to measure in an effective manner, since the air then got rather polluted because of old remaining ship plumes.

It should be pointed at that the main objective was not to measure as many ships as possible but rather to “produce surveillance” in an operational manner at different geographical areas and at different times of the year. The measurements were therefore sometimes carried out at unfavorable meteorological conditions. In order to optimize further measurement campaigns, it would be advisable to constrain the measurements to wind speeds below 8 m/s and fair weather and use different airports as starting point for the surveillance, to eliminate transit time.

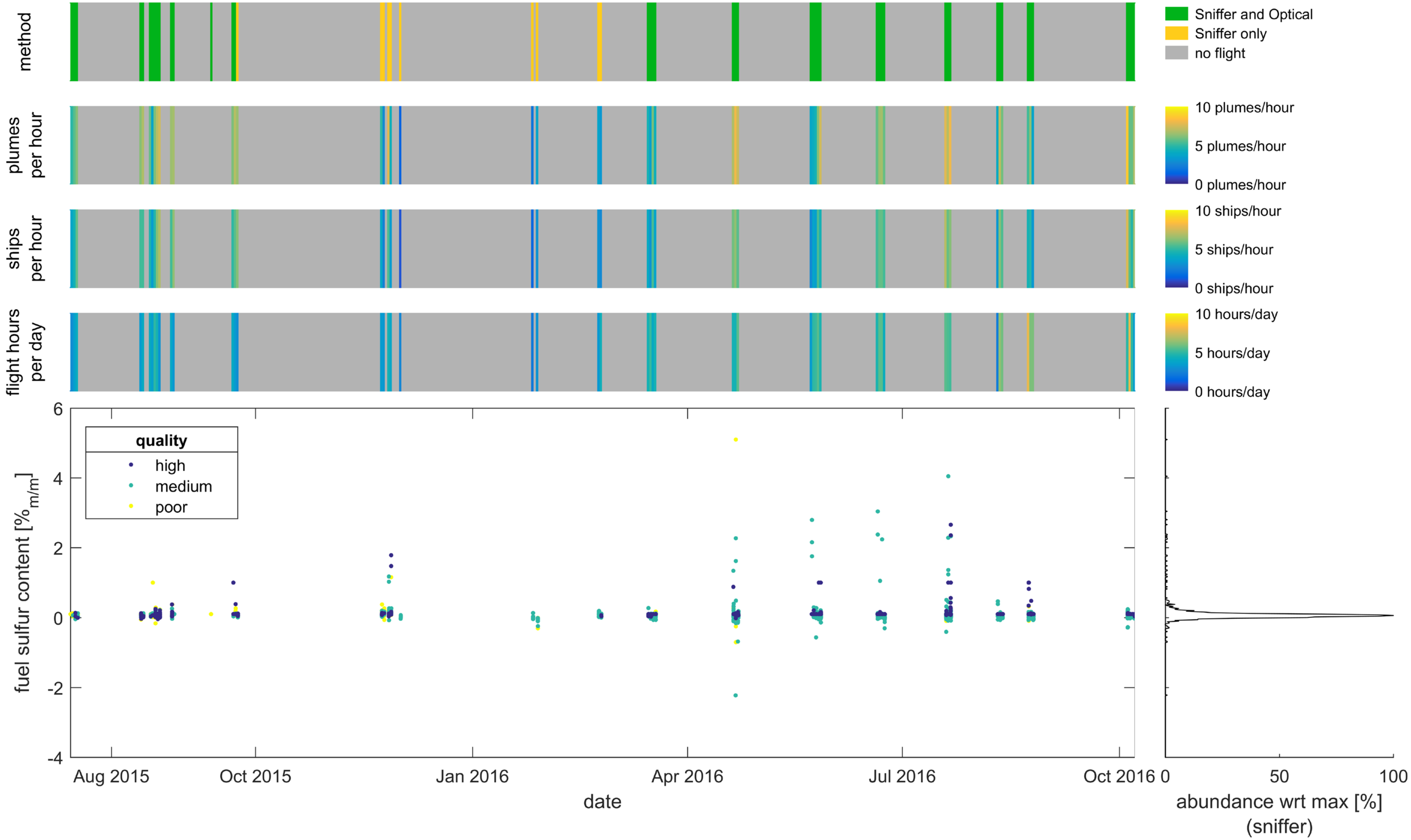


Figure 21. Statistics of the airborne measurements between July 14, 2015 and October 7, 2016. (a) Variability of the measurements rate through time. (b) FSC distribiution over time

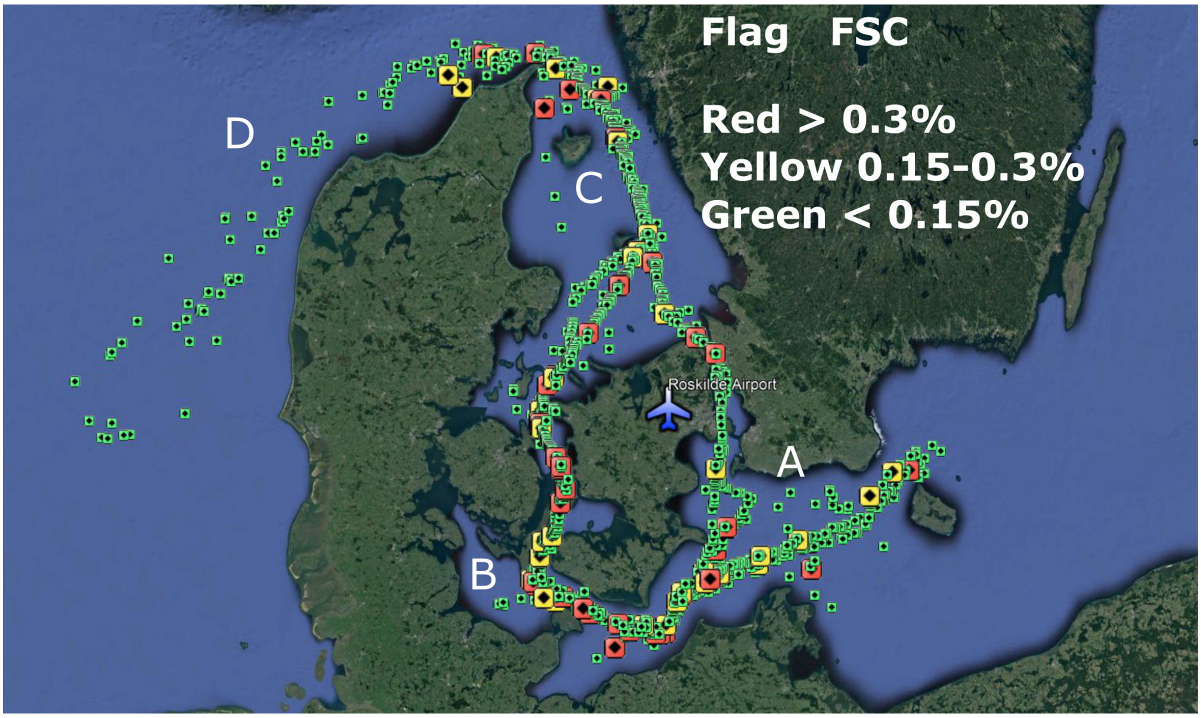


Figure 22. Spatial distribution of the ships measured around the Danish coast during the period July 2015 to October 2016. The data correspond to measurements of individual ships using both the sniffer and optical sensor. The obtained FSC values are color coded: green (<0.16 % m/m), yellow (0.16-0.3 % m/m) and red (>0.3 % m/m). The letters A - D correspond to the 4 different areas that were monitored.

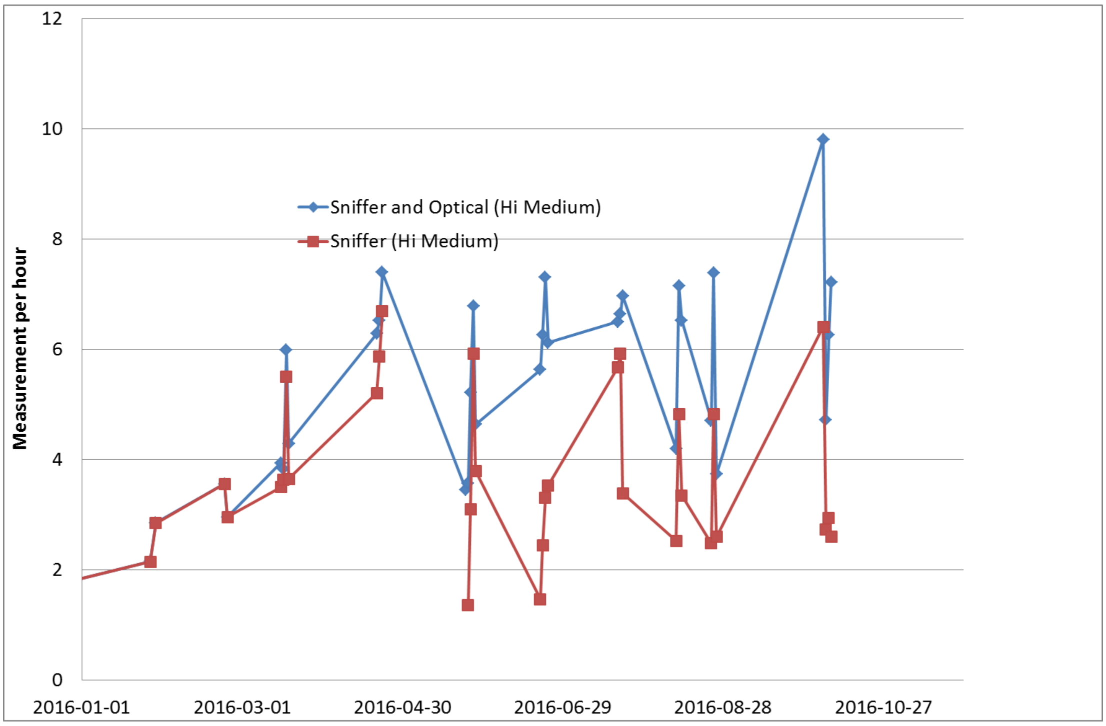


Figure 23. The number of ship measurements per hour during 2016 using both optical and sniffer measurements.

Figure 24 shows the statistical distribution (probability density function) of the FSC corresponding to 820 individual ships measured with sniffer from the aircraft. Note that a ship is counted twice if the measurements are on separate days. The data covers the period June 2015 to October 2017 and the values for an individual ship are estimated by an average of all measurements during the same day. However, when averaging the individual measurements we have assigned different weights for the measurements qualities high, medium and poor, respectively, corresponding to 100%, 30 % and 10 %.

The green curve is an upper estimate of the random noise distribution (precision) of the measurements. This was obtained from calculating the standard deviation of the FSC data centered at the median value, between -0.1 % to 0.15 %, and corresponding to 0.049 %. The FSC data has a median value of 0.035 %. The dotted line in Figure 24 corresponds to the estimated compliance limit, 0.16 %, for which the instrument errors (precision and bias) have been accounted for, see section 4.3.

In Figure 25 a histogram is shown of the number of ships with different FSC levels for airborne sniffer data corresponding to 2015 and 2016. The data for the high FSC ships is highlighted. The data for medium and high quality is indicated and all negative data points were assigned a FSC value of 0.00 %. The reason for this is that negative data points are caused by noise.

In Figure 26 the fraction of ships below a certain FSC level are shown for the same data as in Figure 24, i.e., for 2015 and 2016. The fraction below the compliance threshold of 0.16 % m/m is 94 %. Hence 6 % of the ships are running on non-compliant fuel with a confidence limit of 95 %. Approximately half of the non-compliant ships showed gross non-compliance using FSC higher than 0.5 % m/m . However, as discussed in section 5.1 the compliance rate was significantly lower during 2016.

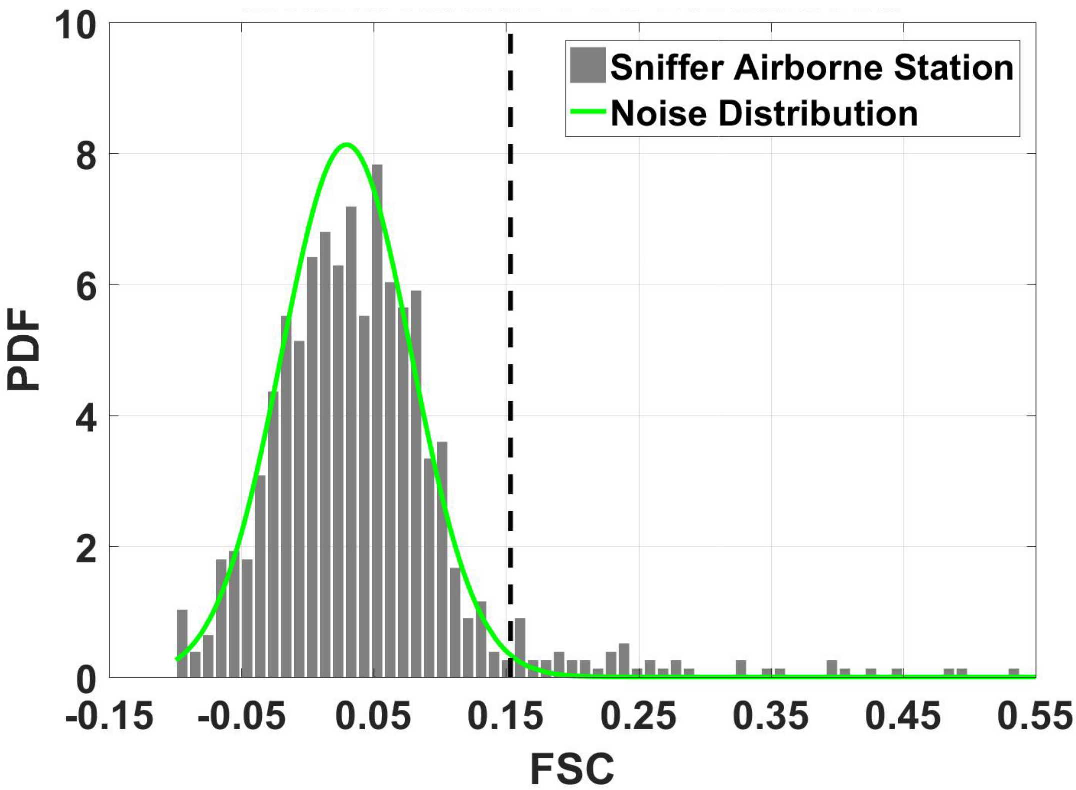


Figure 24. Statistical distribution (probability density function) of the FSC in 820 individual ships measured with sniffer from aircraft. The data covers the period June 2015 to October 2017. The green curve corresponds to the random noise distribution (precision) of the measurement data below 0.16%. The dotted line is the estimated non-compliance limit for which the instrument errors (precision and bias) have been accounted for.

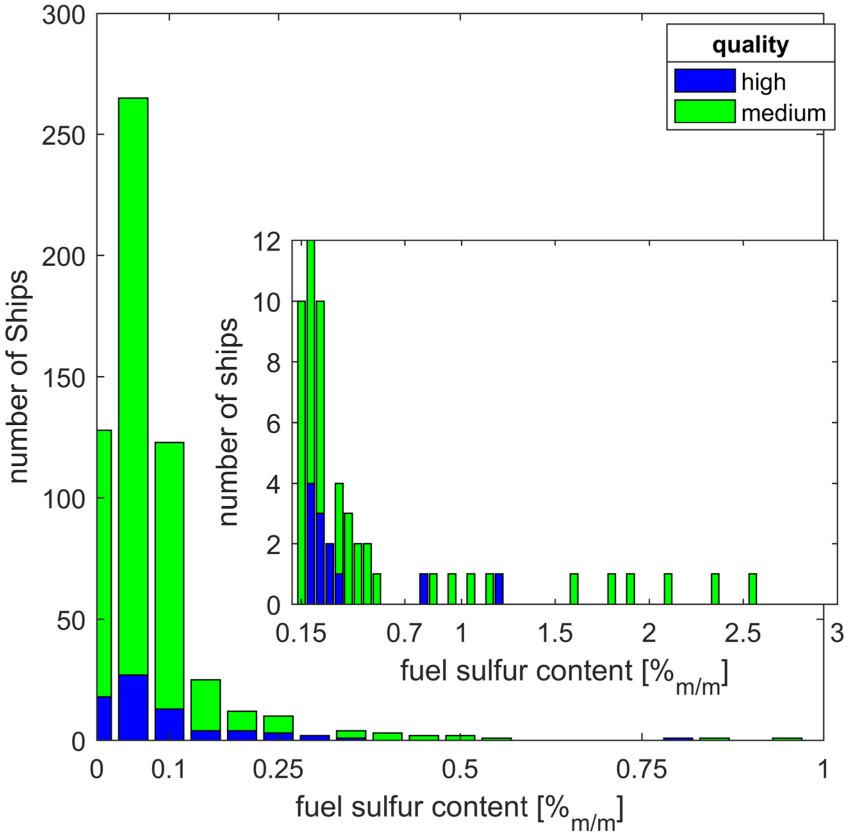


Figure 25. A histogram of the number of ships with different FSC levels obtained from airborne sniffer data measured in 2015 and 2016. The total number of individual ships correspond to 820.

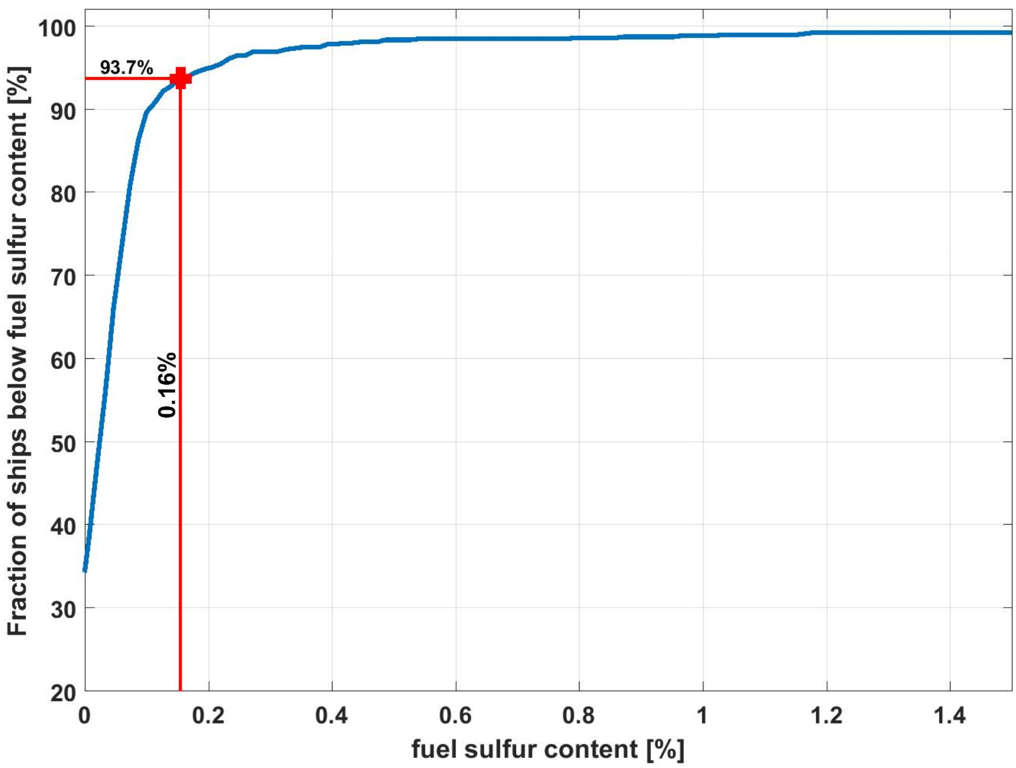


Figure 26. The fraction of ships that have FSC level below a certain level for data corresponding to 820 individual ships measured from aircraft during June 2015 to October 2016. In addition the compliance limit threshold is shown , indicating a compliance level of 94 % for the full period.

4.3 Assessment of the uncertainty of the calculated equivalent sulfur

This section describes the aspects taken into account regarding the assessment of the uncertainty and the estimation of the non-compliance threshold for the FSC values obtained using the sniffer method.

4.3.1 Estimation of precision, accuracy and compliance thresholds

The *precision* of the measurements have been estimated either from multiple measurements of the same ships (fixed station) or from the variability of the data close to the median value (airborne), as partly discussed in the previous sections.

For instance, for the measurements at the Great Belt Bridge multiple observations (> 9) of 30 individual ships measured during 2015 and 2016 were used. From the square root of the sum of the variances of individual ships we obtained an overall precision (1σ) of 0.04 % in FSC units. For the aircraft it was not possible to estimate the measurement precision using the multiple measurements approach. Instead a Gaussian distribution function was fitted to the data centered on the measured median value, i.e. using FSC data only in the range -0.1 % to 0.15 %. In this manner was obtained a precision value corresponding to 0.049 % (1σ). This value corresponds to the scatter in the measurements of low FSC ships and it would have been an accurate estimate of the precision if all ships were using the same FSC. Since this is not the case the value corresponds to an upper estimate of the precision.

The airborne instrument was running without a kicker which causes sensitivity to vapors/particles from lubrication oil, as explained in section 2.3 and 2.4. However, this potential measurement artifact is not evident in the precision value for the airborne measurements which is considerably better than similar measurements at the Göteborg ship channel showing a precision of only 0.1 % m/m in FSC for non-kicker measurements (Mellqvist et al., 2017b). The reason is probably that ships which operate at steady state conditions in the open sea, as is the case for the airborne measurements, emit considerably less lubrication oil particles than the ones maneuvering close to the harbor.

The *accuracy* of the sniffer measurements have been assessed by comparison to on board sampling by port state control authorities. The Swedish port state authority (*pers. comm.. Caroline Petrini, Swedish transport Agency*) measured 440 vessels in 2015 and 178 vessels in 2016 with the same median value on both years (0.08 % m/m). The same average value was obtained from 316 fuel samples in Danish ports in 2015 and 2016 (*pers. comm. Dorte Kubel, Danish EPA*). It is rather likely that the median FSC of the ships passing the Great Belt Bridge and around the waters of Denmark is the same, or higher, as the port state control data and we have therefore adjusted the threshold for compliance accordingly.

For instance for the sniffer data measured at the Great Belt Bridge there is a negative bias of 0.043 % m/m in FSC units for the period June 2016 to January 2017, when compared to port state control data. Ships running with an FSC value of 0.1 % m/m will hence be measured as having a FSC of 0.057 % m/m, on average. However, since the measurement have random noise associated with them corresponding to a precision with standard deviation 0.04 %, the data will be spread out according to a Gaussian distribution, as shown in Figure 18. Most of the data (95 %) will be within 2 standard deviations from the 0.057 % m/m value; this gives an upper value of 0.137 % m/m in FSC and this is the compliance threshold used in our evaluation. Individual ships with FSC measured above this limit are considered to use non-compliant fuel with 95 % confidence limit. The general threshold for compliance can be described according to Eq 3,

$$Compliance\ threshold_{biased} = 0.1\ \% - bias + \ 2\ \sigma \qquad \text{Eq. 3}$$

, where the *bias* corresponds to the difference between the median value of port state control data and the median value of sniffer measurements, σ corresponds to the precision obtained as the standard deviation of multiple measurements and 0.1 % is the SECA limit for FSC. We can not explain the reason for the negative bias and potentially it is caused by tubing losses for low levels of SO_2. One could also speculate that a higher proportion of the sulfur could be in particulate form at low levels than at high.

Note that the compliance threshold is modified to account for the bias in our data, so it can be used to calculate compliance levels. It is however, not the threshold for the real data, since in this case one should use the un-biased threshold. For instance, in the case of the Great Belt Bridge the real threshold, at 95 % confidence limit is 0.18 % m/m. This means that it is not possible to detect non-compliant ships using a FSC in the range 0.1-0.18 % m/m.

When performing this analysis it has been observed that the median value of the sniffer measurements depended on the time periods, even though the port state data were the same. For instance the median of the measured FSC values for the period June 2016 to January 2017 was 0.038 % m/m while for the longer period June 2016 to May 2017 the value was 0.025 % m/m. This hence impacts the bias compliance threshold, so for the longer period it would hence be 0.125 %. Instead of a compliance limit of 95.5 % the value then changes to 94.7 %. We have not taken this effect into account and instead chosen one median value for each measurement system for the full period. This will cause some additional uncertainty in the analysis of compliance limits.

Due to the complexity of the measurements it is difficult to assess their accuracy from theoretical estimations and the best approach is to compare to other measurements. In 2008 we did such a comparison for high FSC ships as described in section 2.4, showing an overall estimated relative uncertainty for SO_2 of 23 % with a relative precision of 0.19 % at the 1 % m/m FSC level for the airborne measurements.

In Figure 27 is shown the FSC of a scrubber ship obtained by the sniffer at the Great Belt Bridge. In addition is shown the corresponding data from on board sensors on the ship. We have also added a curve in which the sniffer FSC data have been corrected for the overall bias value of 0.055 % m/m, as explained in section 4.1. The difference between the sniffer and the onboard FSC data is - 0.02 ± 0.023 % m/m. For the bias corrected data the corresponding difference is instead 0.03 ± 0.023 % m/m. The obtained differences are smaller than the estimated errors.

One potential additional artifact in the airborne measurements is the fact that the contact with the ship gas plume is very short when flying, i.e. usually a few seconds. To evaluate this we have made a "puff test" in which a calibration gas with a high concentration mixture of SO_2 (203.9 ppm) and CO_2 (4.293 %) is injected in front of the measurement probe of the aircraft. The SO_2 to CO_2 ratio in this gas corresponds to a 1.1 % m/m FSC ship according to

Eq. 1. In Figure 28 an example of one such puff measurement is shown with the measured concentrations of SO_2 and CO_2 shown versus time. The corresponding results for several experiments in which the measured ratios of SO_2 and CO_2 have been converted to FSC according to Eq. 1, are shown in Figure 29. The FSC obtained from the plume measurements corresponds to 1.01± 0.13 % m/m; hence there is a negative bias in the estimated FSC of -0.1 % m/m (corresponding to the accuracy) and a spread of the data corresponding to a precision of 0.13% m/m. Both of these values are consistent with previous estimates as descried in section 2.4. In Table 7 the overall estimated uncertainty for the measurements is summarized, based on the discussion above and the one in section 2.4. Some of the reasons for the uncertainties are discussed in section 2.3, Table 2.

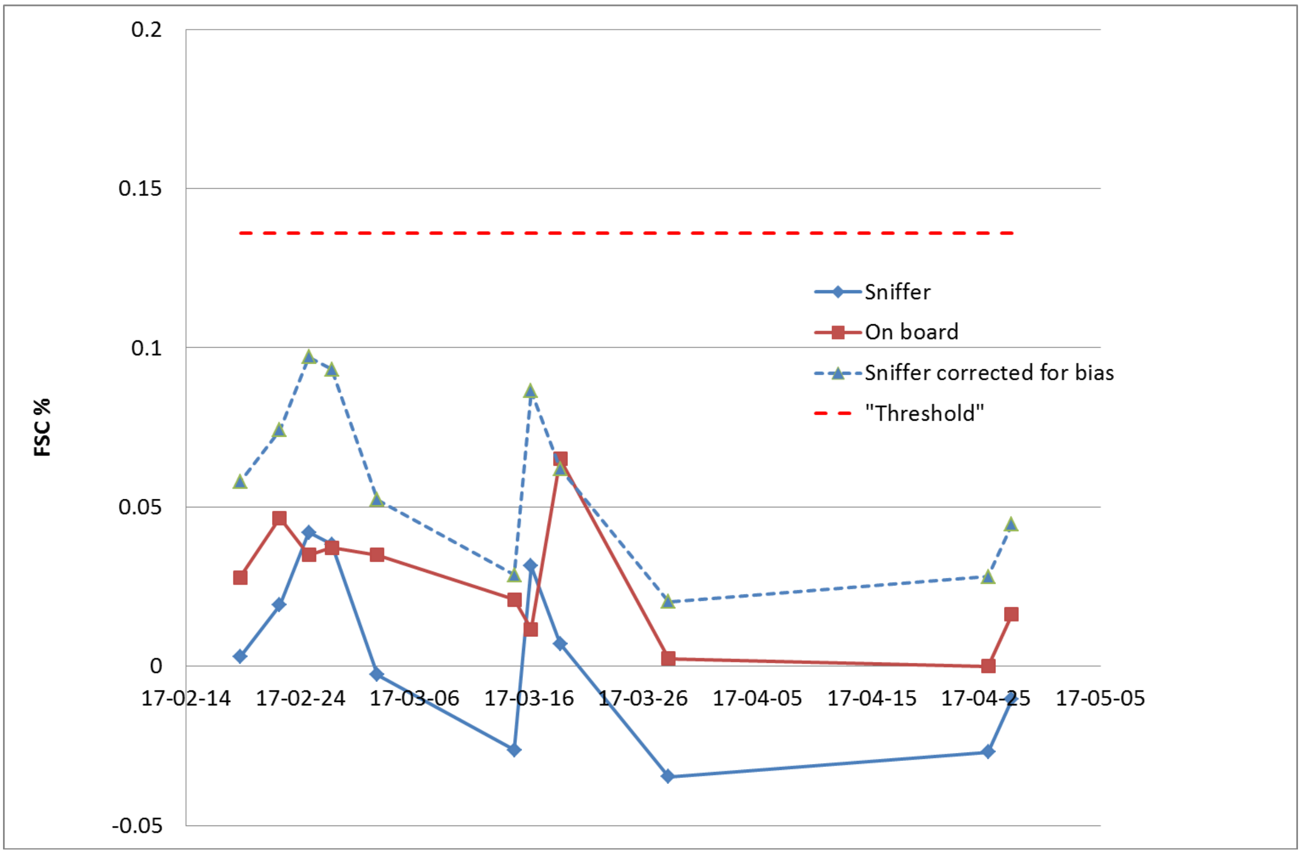


Figure 27. Comparison of the sniffer instrument at Great Belt Bridge (Mellqvist, 2017) with on board sensors on a ship equipped with scrubber. In addition, the measured sniffer FSC data have been corrected upwards by 0.055 % m/m so that the median values for all ships correspond to on board measurements by the port state control authorities.

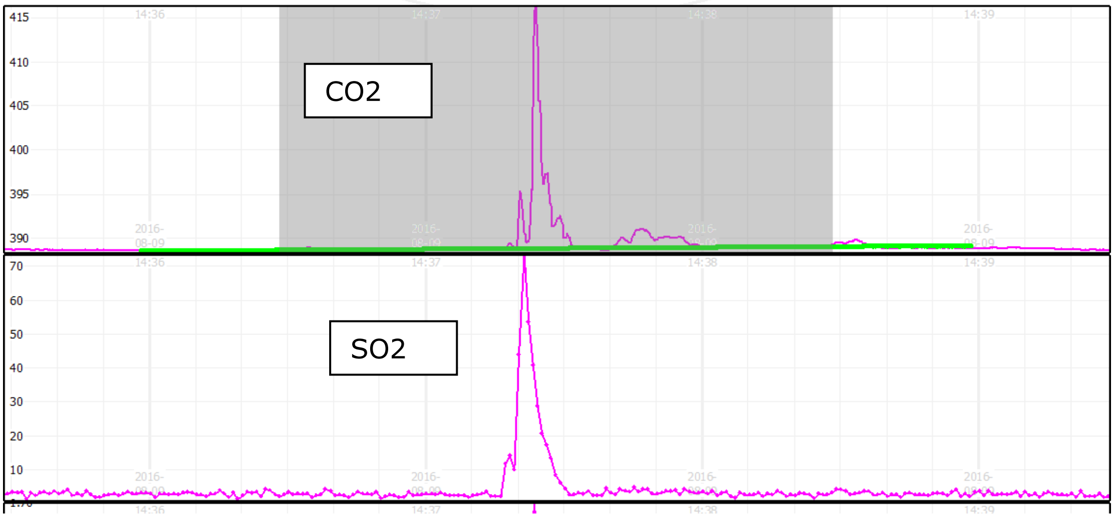


Figure 28. A quality assurance test in which a short pulse (10s) of premixed SO_2 and CO_2 gas was blown across the air-plane inlet and analyzed by the sniffer systems in the aircraft. Here is shown the concentration of CO_2 (top) in ppm and SO_2 (middle) in ppb versus time.

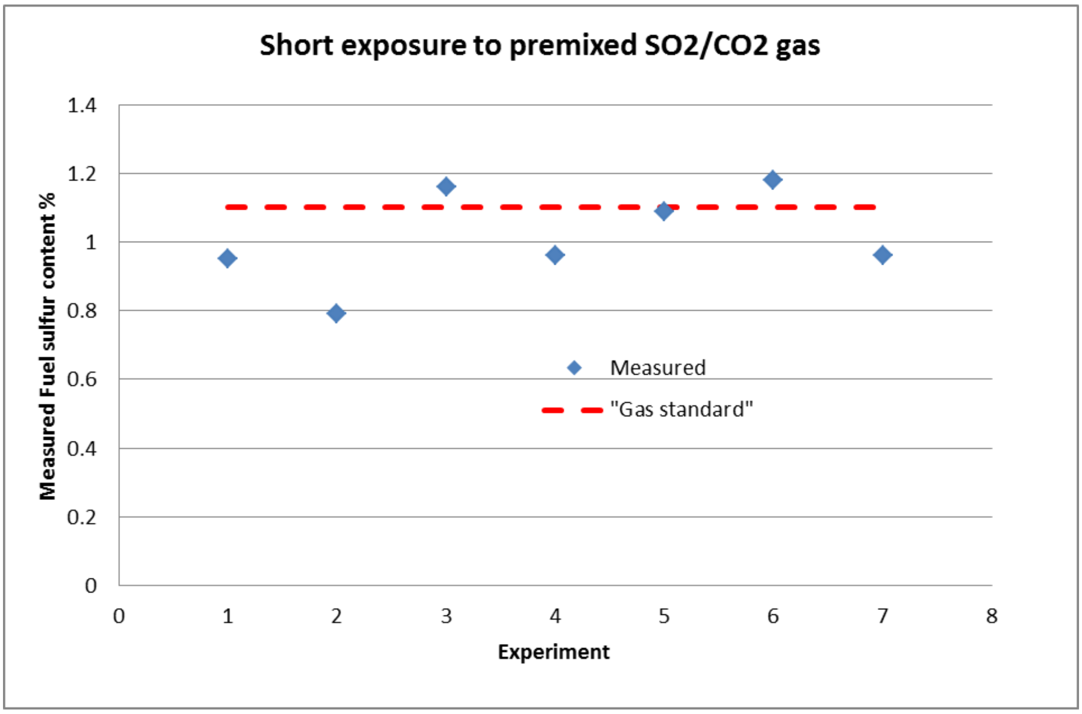


Figure 29. A quality assurance test in which short pulses of premixed SO_2 and CO_2 gas was blown across the airplane inlet and analyzed by the sniffer systems in the aircraft. The general accuracy of the sniffer measurements was 10 % (0.1 % m/m) with a precision of 13% (0.13% m/m).

Table 7. Estimated overall uncertainty for the sniffer FSC measurements in this study. All values correspond to the absolute FSC unit.

	0.1% m/m **Fixed**	**0.1% m/m** **Airborne**	**1 % m/m** **Fixed/airborne**
Random uncertainty abs FSC unit	±0.04% m/m	±0.049% m/m	±0.19 % m/m (1)
Systematic bias	-0.04%-0.055% m/m	-0.045%	-0.1 % m/m (2)
Unbiased Threshold(2) for compliance limit (95 % confidence limit)	0.18% m/m	0.2% m/m	

(1) Beecken 2014a and other studies, see section 2,
(2) Balzani 2014

4.3.2 Measurement artifacts fixed station

As described in section 3.1.3 a metallic sampler was used as inlet between June 2015 to middle of May 2016 to prevent water droplets and salt to enter the sniffer tube system at the Great Belt. Suspicion was however raised that there was a problem with the inlet after comparing airborne and fixed measurements. A test in which premixed SO_2/CO_2 gas was puffed in front of the inlet was carried out, Figure 30, showing that the probe reduced sulfur by almost 50 %. For this reason the inlet was replaced as described in section 3.1.3.

To assess the effect of the gas inlet on the sulfur measurements we have compared the FSC results for 5 high FSC vessels (> 1 %) that were measured by sniffer both at the Great Belt Bridge and from the aircraft, close in time and in the vicinity of the bridge. The results showed that 70 – 90 % of the sulfur was lost when using the metallic inlet probe. However, still 3 out 5 high sulfur ships could be detected from the fixed station as using non-compliant fuel with 95 % confidence limit and 4 out 5 with 67 % confidence limit. During this period, i.e. Oct 2015 to May 2016, 3 % out of 2537 ship measurements were higher than the compliance threshold. It is uncertain whether ships running on moderately high FSC (0.5% or lower) could be observed during the first period. When comparing the average FSC of multiple measurements of 9 ships running with low sulfur, a difference of almost 100% is seen when comparing the first period against the second one, indicating that the sulfur for ships using low FSC (0.08%) was completely lost in the aluminum inlet system. Another indication of sulfur loss is the fact that the median FSC value is significantly lower for the first measurement period (June 2015 to May 2016), i.e. 0 % FSC, compared to the second one (June 2016 to May 2017), i.e. 0.025 % FSC. To summarize, the measurement system was much less sensitive and showed systematically lower values in the first year of the measurements; but in most cases the high sulfur ships were still flagged as using non-compliant FSC.

The second artifact with the fixed station was the fact that the instrument was running without a kicker during the period, June to October 2015. During this period the median FSC value was 0.03 % m/m. However 16 % out of 2265 measurements were higher than the compliance threshold of 0.14 %, probably mostly due to this artifact. As mentioned before, the same effect was not visible in the statistics of the airborne data, even though the latter measurements were carried out without a kicker.

Figure 30. Testing the gas inlet for short time exposure using a premixed SO_2 and CO_2 gas corresponding to a 1 % vessel.

5 Discussion

In this section we discuss how well the ships comply with the FSC limits of the EU sulfur directive, and whether there are systematic patterns in this behavior.

5.1 Comparison between fixed and airborne station

One objective in this study was to investigate whether the measurements at the fixed station were representative for the emissions on open sea. We have therefore compared the time period for which good quality measurements from both the airborne and fixed station at the Great Belt Bridge were overlapping, corresponding to June to October 2016. In Figure 31 the fraction of ships that were measured below a certain FSC level are shown for the aircraft and fixed site, with their respective threshold. The FSC compliance rates were 92 % for the aircraft and 95 % for the fixed station. Here 2 % and 1 % of the total ships, respectively, corresponded to gross non compliers (> 0.5 % FSC) in the airborne and fixed station. The difference between the two data sets, and higher values from the air could possibly indicate that the ships have switched over to low/high FSC fuel too early/soon. There could also be a contribution from the measurement interference with organic vapors causing somewhat higher values in the aircraft data, although this is not evident in the error characterization of the airborne data as discussed in section 2.3 and section 4.2. For the aircraft data the compliance levels during the second half of 2016 were lower, i.e. 92 %, than for the full monitoring period, June 2015 to October 2016, corresponding to 95 % as shown in Figure 26.

For the fixed station the compliance level measured at the Great Belt Bridge for the second half of 2016 was 94.6 % and this was considerably lower than the first half of 2017, corresponding to 97.4 %., see Table 8. Again the numbers above can be compared to onboard sampling by port state control during 2015 and 2016, showing compliance rates of 95 % in both Denmark (*pers comm. Dorte Kubel, Danish EPA*) and Sweden (*pers comm. Caroline Petrini, Swedish transport agency*).

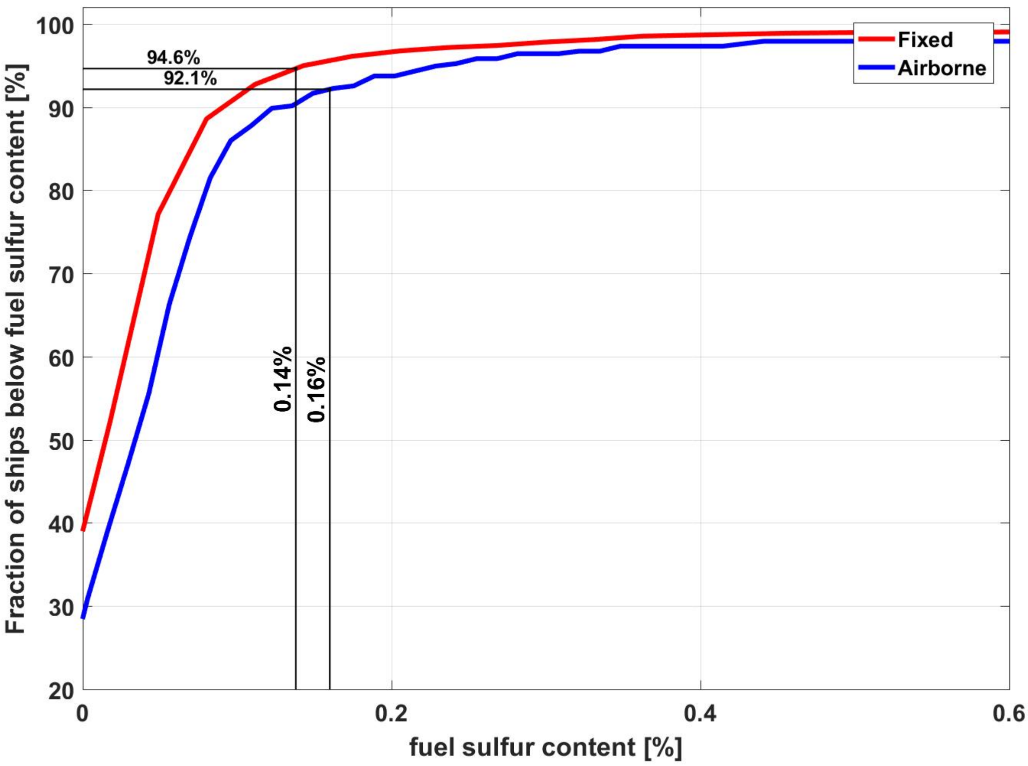


Figure 31. The fraction of ships that were below a certain FSC level for data corresponding to 343 individual ships measured from aircraft and 1691 ships measured from the Great Belt Bridge during June to October 2016. The median FSC value for the fixed station is 0.04 % m/m and for the airborne 0.035 % m/m.

Table 8. Comparison of measured FSC statistics in ships from the Great Belt Bridge between two time periods.

Statistical parameters	Period 1, 2016 (June 1- Oct 30)	Period 2, 2017 (Jan 25 -May 31)	Difference in FSC units m/m	Relative difference
No of ships	1691	1663		
Mean	0.06 %	0.02 %	-0.04 %	-67 %
10th percentile	-0.02 %	-0.04 %	-0.02 %	100 %
25th percentile	0.006 %	-0.013 %	-0.02 %	-317 %
50th percentile	0.04 %	0.013 %	-0.03 %	-68 %
75th percentile	0.07 %	0.043 %	-0.03 %	-39 %
90th percentile	0.11 %	0.078 %	-0.03 %	-29 %
Compliance level (threshold 0.14%)	94.6 %	97.4 %	2.80 %	3 %

Figure 32 shows the temporal distribution between Oct 2015 and May 2017. From the graph, it appears that there was more non-compliance cases during the summer period of 2016. Part of this effect is due to a reduced sensitivity of the sniffer instrument before May 23 2016.

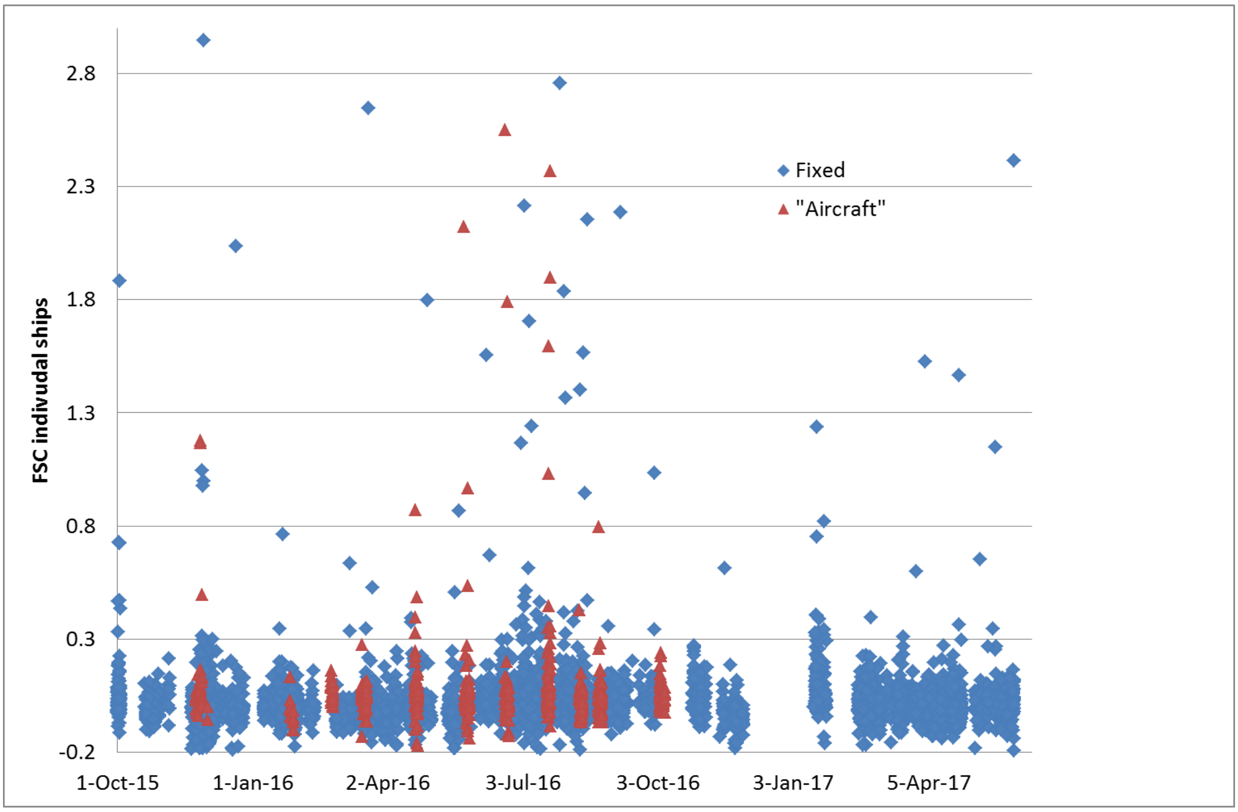


Figure 32. Variability of FSC and SO_2/NO_2 ratios measured by the fixed station and airborne measurements. During this period, the fixed measurements were regularly accompanied by airborne measurement campaigns.

5.2 Comparison to port state control

In Figure 33 is shown a comparison of FSC data for individual ships obtained from on board inspections by Swedish port state against sniffer results. The data shows individual ships that have been checked both by port state control during 2015 and 2016 and measured with sniffer within the same year from Great Belt Bridge, Göteborg and airborne measurements around Denmark (the latter as part of the Compmon project). Red values correspond to ships which have been found to run on non-compliant fuel.

For 7 % of the ships in this data set, the on board sampling showed that the ships were using non-compliant fuel (higher than 0.11 %) and in 31 % of these cases the sniffer indicated that these particular ships was non-compliant with EU sulfur directive. This indicates that if the port state control was done based on guidance from the sniffer measurements the probability of finding non-compliant ships would be 31 %. This corresponds to an increase of a factor of 4 from the 7 % probability of the port state control, based on a choosing ships in a random manner.

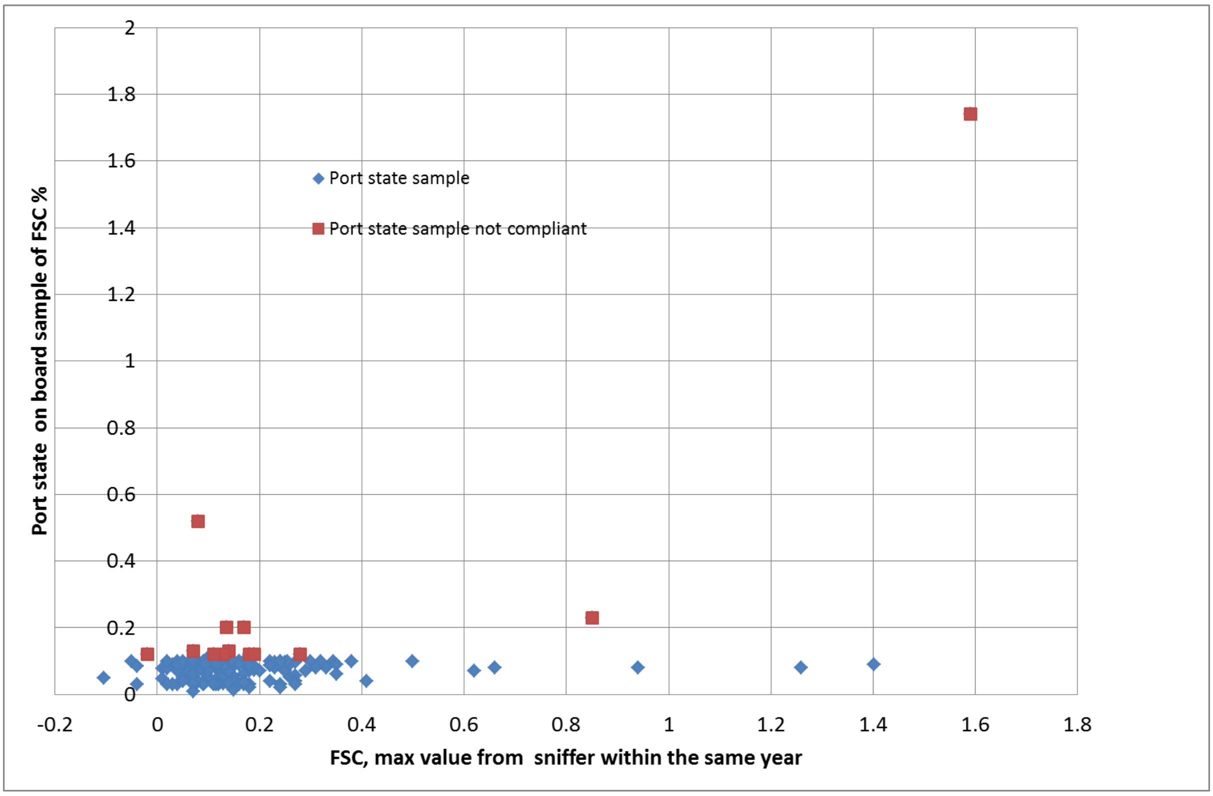


Figure 33. Comparison of ships that have been measured by Swedish port state during 2015 and 2016 and sniffer measurements in this project from the fixed and airborne measurements.

5.3 Observed cases of non-compliance

During the period October 2015 to May 2017, 241 individual ships were measured at the Great Belt Bridge to use non-compliant FSC. Note that in this report a ship is counted twice if the measurements are done on separate days, but in this case we only count each ship once. The above period is used here since then the fixed instrument had no interference from organic vapors, although it was less sensitive to sulfur during the first period. The high quality measurements of non-compliant ships have been flagged in the EU database THETIS-EU by the Danish EPA and this information is used by the port state control authorities when making decisions on board inspections. The majority (88 %) of the non-complying ships were only detected high once. This supports the conclusions in the next section about differences in FSC for ships that pass the Danish area frequently and occasionally, respectively.

A few shipping companies appears to be overrepresented in the non-compliance statistics. For instance, for one company more than 38 individual vessels have been measured on 116 occasions at the Great Belt Bridge, showing an overall compliance rate of 89 %, to be compared to the 95 % measured on average. For the aircraft data, 11 ships have been measured from the same company with a compliance rate of 73 %. Another company, showed 83 % compliance rate at the Great Belt Bridge (4 vessels measured 12 times) and 66 % from aircraft but for only 2 vessels but measured only on 3 occasions. There are some uncertainties here due to the relatively few number of ship measurements.

Several of the ships operating in the SECA are equipped with scrubbers which remove the SO_2 from the flue gas. The ships are in this manner able to operate on high sulfur fuel. The absolute majority of the scrubber ships are certified according to what is called scheme B, requiring a certified monitoring system on board that continuously documents that the emissions stay within the requirements of the EU sulfur directive. The apparent FSC is actually calculated in the same manner as in Eq.1 based on the ratio between SO_2 and CO_2 in the flue gas and the limit for the apparent is the same. i.e. 0.1 % m/m. However, there are a number of examples in our dataset of scrubber ships that have been measured above the compliance thresholds. One example here is a ship, flagged outside the SECA, which was measured, both from the fixed station and the aircraft, to run with apparent FSC which was higher than the compliance limit. The ship was later checked by Swedish port state control authority and there was only one scrubber equipped on one of the 4 engines even though the ship was running on high sulfur fuel. Another example is a ship equipped with scrubbers that passes the Great Belt frequently. In Figure 34 FSC measurements of this ship are shown together with measurements from a sister ship also equipped with similar scrubbers. Some of the on board sensors were compared to our measurements in Figure 27. It can be seen in the figure that the apparent FSC for ship (A) became high in the summer of 2016. This was discussed with the ship operator and it turned out one of 4 scrubbers was not in operation due to a technical problem. A third example is a ship equipped with scrubber that operates according to a regular schedule inside SECA. This was encountered on three separate missions from the aircraft, spread over a month, running with apparent FSC which was considerably higher than the limit.

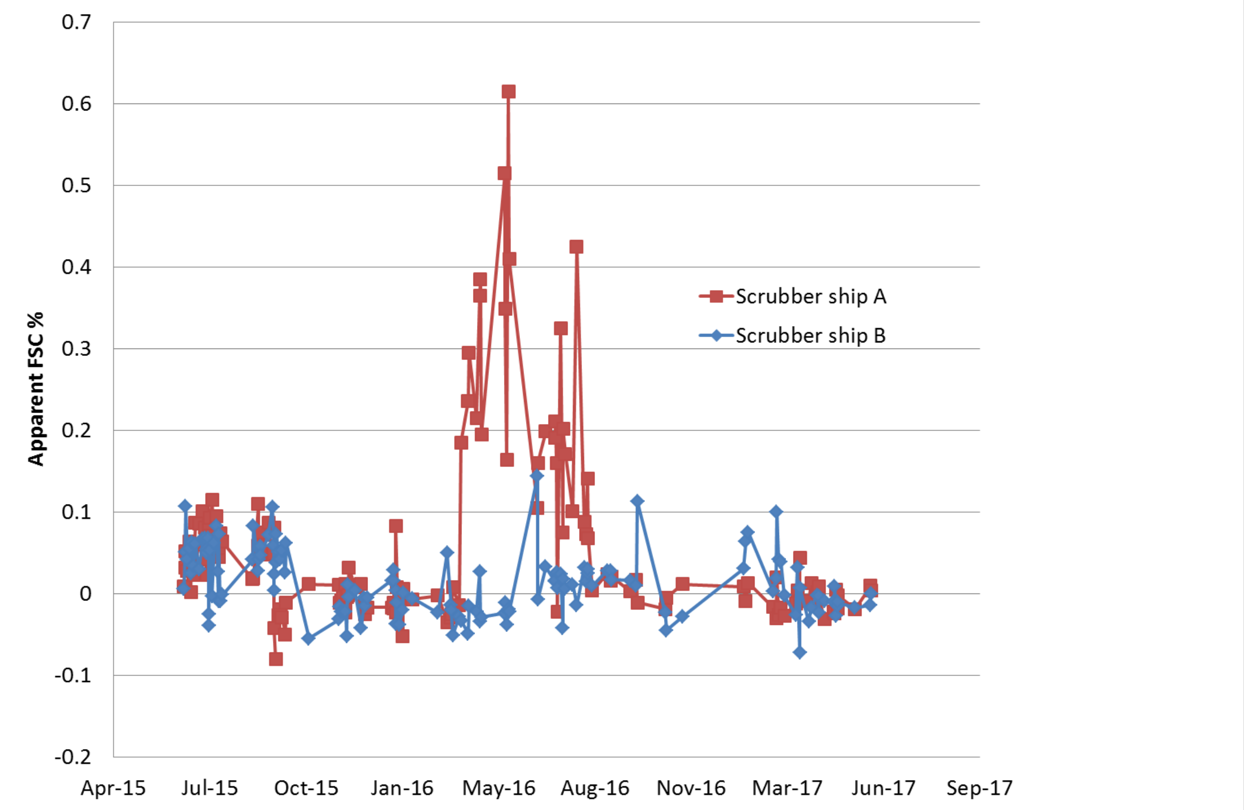


Figure 34. Apparent FSC at the Great Belt Bridge measured from two similar ships equipped with scrubbers.

5.4 Overview of FSC on navigation patterns

By analyzing AIS information it is possible to determine if a ship navigates frequently or occasionally in the area around Denmark fixed station. We have studied the median values of 37 ships that were measured to be non-compliant at the Great Belt Bridge or from the aircraft. It was found that 45 % of these were only occasionally in the area while the others operated in the Danish area at least a few times a year. Figure 35 shows the results of the FSC average when the sample is divided between the two previously outlined groups. The groups labelled as occasional have 60 % higher emissions on average. Similar differences apply also for the median and 25^{th} and 75^{th} percentiles.

Most of the ships classified as occasional were identified during the second half of 2016 (July-November 2016) and they are typically ships operating over long distances. The ships classified as "frequent" are ferries and cruisers with very clear and defined routes and, vessels working within the SECA.

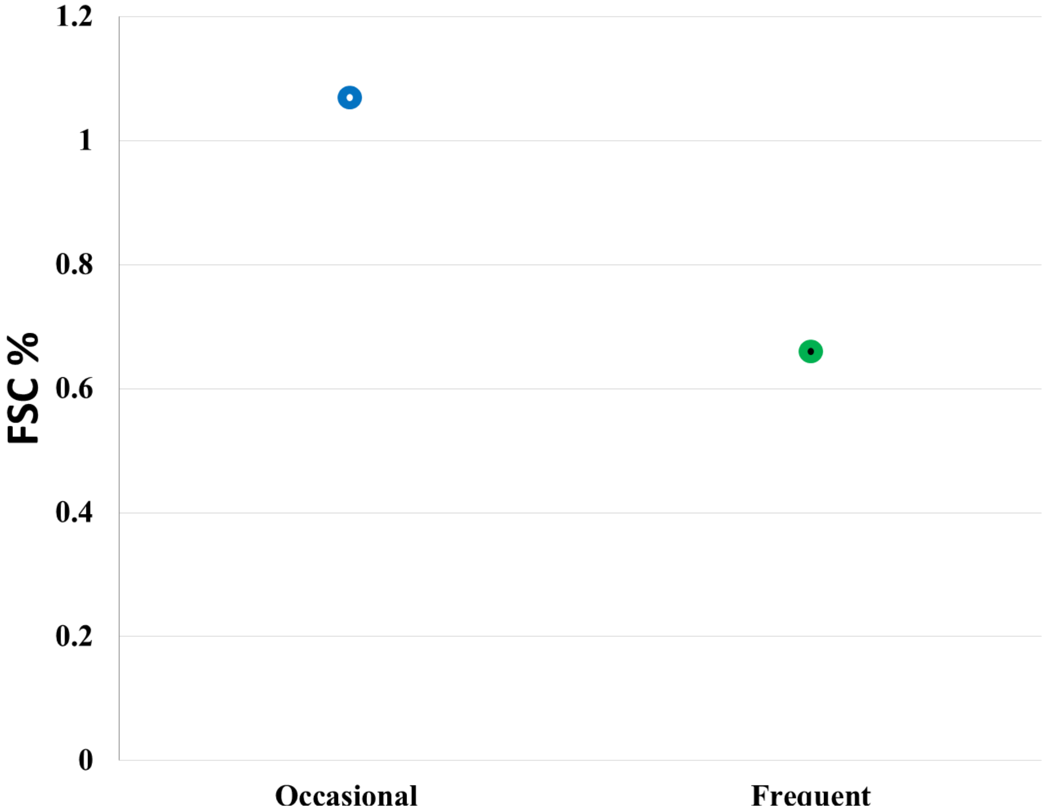


Figure 35. The average FSC for ships measured above FSC 0.16% divided into occasional and frequent ships, depending how often they are in the Danish area.

6 Conclusion and outlook

Operational

Based on the airborne measurements it is feasible to carry out 5 - 10 ship measurements per hour depending on the ship traffic and objective of the measurements. The precision in measured FSC for the sniffer system is ±0.05 % m/m with a small negative bias, -0.05 % m/m, possibly due to sampling losses. There is a potential measurement artifact due to organic vapors but in our statistical analysis it can't be detected when measuring from air, only from the fixed stations.

The optical sensor is a good tool for increasing the amount of ships measured in combination with easier flight operation and therefore better safety. Measurements performed elsewhere indicates that ships can be classified either as low (0.1 % m/m) or high FSC (1 % m/m) ships with 80 - 90 % probability. We managed to measure 813 ships with the optical sensor to be compared to 914 with the sniffer. The comparisons is however not straightforward since the optical and sniffer instruments were operated independently and did not always measure on the same ships.

From the fixed station approx. 4000 measurements per year with high or medium quality can be carried out. The precision in the measured FSC is ± 0.04 % m/m with a small negative bias (-0.05 % m/m), possibly due to sampling losses. Hence ships with FSC above 0.18 % m/m can be controlled with 95 % confidence interval. If the SO_2 sniffer instrument would be replaced by a commercially available, laser based instrument the uncertainty would likely decrease by a factor of 2 to 3. However this system would almost double the hardware price of the sniffer system.

Compliance levels and trends

The compliance level during the period and platform varies between 92 - 97 %. Only 1-2 % of ships were in gross non-compliance with the EU sulfur directive having FSC values above 0.5 % m/m. There are differences over time, with the highest values in the summer of 2016. The compliance level is close to the values (95 %) measured by the port state control authorities in Sweden and Denmark 2015 and 2016. When comparing ships measured port state control authorities and the ones in this project it can be deduced that the efficiency of finding non-compliant vessels could be increased by at least a factor of 4 if the port state controls were guided by measurements.

Most of the non-compliant ships (90 %) have been measured high only once. But there are cases with individual ships and ship operators that are more abundant in the non-compliance statistics.

The non-compliant ships that are seldom in the Danish area have higher emissions of SO_2 than the non-compliant ones that operate their more frequently. Two ships equipped with scrubbers appear to have had operational problems over some time, at least one of them due to malfunctioning sensors. A ship flagged outside SECA was running on high sulfur fuel oil during a scrubber trial even though only one out of four scrubbers was in operation.

7 Acknowledgement

The project was carried out for the Danish Environmental Protection Agency corresponding to the tender: 2015/S 004-004391. We thank Tue Friis Hansen at Aircraft Aps for providing the aircraft measurements and general support. We also acknowledge the Swedish funding agency Vinnova (IGPS project) and the EU project CompMon for funding the development of the measurement systems and software and providing some additional data to this project. Caroline Petrini (Swedish transport agency) and Dorte Kubel (DEPA) are acknowledged for providing ports state control data.

8 References

Alfoldy, B., J. Balzani, J. and F. Lagler.: FINAL REPORT ON REMOTE SENSING OF SHIPS' EMISSIONS OF SULFUR DIOXIDE, 21.06.2011, 2011, http://ec.europa.eu/environment/air/transport/pdf/ships/Final-report.pdf

Alfoldy, B., et al.: Measurements of air pollution emission factors for marine transportation, Atmos. Meas. Tech., 6, 1777-1791, 2013.

Balzani-Lööv, J. M., et al.: Field test of available methods to measure remotely SOx and NOx emissions from ships, Atmos. Meas. Tech., 7, 2597-2613, 2014, www.atmos-meas-tech.net/7/2597/2014/ doi:10.5194/amt-7-2597-2014, 2014.

Beecken, J., Mellqvist, J., Salo, K., Ekholm, J., and Jalkanen, J.-P.: Airborne emission measurements of SO_2, NOx and particles from individual ships using sniffer technique, Atmos. Meas. Tech., 7, 1957–1968, 2014a.

Beecken J. et al.: Emission Factors of SO_2, NOx and Particles from Ships in Neva Bay from Ground-Based and Helicopter-Borne Measurements and AIS-Based Model, Atmos. Chem. Phys. Discuss., 14, 25931-25965, 2014b

Beecken, J.: Remote Measurements of Gas and Particulate Matter Emissions from Individual Ships, PhD dissertation, Earth and Space Sciences, Chalmers University of technology, ISBN 978-91-7597-141-4, ISSN 0346-718X, 2015

Berg, N.: Remote Measurement of Ship Emissions, Technical report 43 L, Earth and Space Sciences, Chalmers University of Technology, 2011

Berg, N., Mellqvist, J. et al.: Ship emissions of SO_2 and NO_2: DOAS measurements from airborne platforms, Atmos. Meas. Tech., 5, 1–14, 2012.

Eichler, P., et al., A novel inlet system for online chemical analysis of semi-volatile

submicron particulate matter, Atmos. Meas. Tech., 8, 1353–1360, 2015

Eichler et al., Lubricating Oil as a Major Constituent of Ship Exhaust Particles, Environ. Sci. Technol. Lett., 2017, 4 (2), pp 54–58

Jalkanen, J.-P. Et al.; A modeling system for the exhaust emissions of marine traffic and its application in the Baltic Sea area. Atmospheric Chemistry and Physics (9), 9209-9223, 2009

Mellqvist, J and Berg N., Final report to Vinnova: IDENTIFICATION OF GROSS POLLUTING SHIPS RG Report (Göteborg) No. 4, ISSN 1653 333X, Chalmers University of Technology, 2010 (http://publications.lib.chalmers.se/publication/214636)

Mellqvist, J., Ekholm, J., Salo K. and Beecken J., Final report to Vinnova: IDENTIFICATION OF GROSS POLLUTING SHIPS TO PROMOTE A LEVEL PLAYING FIELD

WITHIN THE SHIPPING SECTOR, RG Report (Göteborg) No. 11, Chalmers University of Technology, 2014 http://publications.lib.chalmers.se/publication/214636

Mellqvist, J., Beecken, Conde, V, and J. Ekholm, CompMon report: Certification of an aircraft and airborne surveillance of fuel sulfur content in ships at the SECA border, Chalmers University of Technology, 2017a, (https://compmon.eu/)

Mellqvist, J., Beecken, Conde, V, and J Ekholm, CompMon report: Fixed remote surveillance of fuel sulfur content in ships from fixed sites in the Göteborg ship channel and at Öresund bridge, Chalmers University of Technology, 2017b, (https://compmon.eu/)

Mellqvist, J., Beecken, and J Ekholm, Remote Quantification of Stack Emissions from Marine Vessels in the South Coast Air Basin, Report to South Coast Air Quality Management District, 2017c, (http://www.aqmd.gov/fenceline-monitoring/project-3)

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

Moldanova, J. et al.: Characterization of particulate matter and gaseous emissions froma large ship diesel engine, Atmospheric Environment 43, 2632–2641, 2009

Petzold, A. et al.: Experimental studies on particle emissions from cruising ship, their characteristic properties, transformation and atmospheric lifetime in the marine boundary layer, Atmos. Chem. Phys., 8, 2387–2403, 2008.

Appendix I.

Number of measured plumes and ships from flight.

date	plumes		ships		flight hours
	total	per hour	total	per hour	
2015-07-14	26	9.2	10	3.5	2:50
2015-07-15	29	9.7	12	4	3:00
2015-07-16	36	9.8	17	4.6	3:40
2015-07-17	57	13.4	25	5.9	4:15
2015-08-13	46	13.1	20	5.7	3:30
2015-08-14	67	16.5	22	5.4	4:04
2015-08-17	50	12.1	20	4.8	4:08
2015-08-18	23	6.3	13	3.5	3:40
2015-08-19	53	10.6	26	5.2	5:01
2015-08-20	81	17.5	33	7.1	4:37
2015-08-21	50	15.8	22	6.9	3:10
2015-08-26	62	15.4	25	6.2	4:02
2015-08-27	23	6.7	22	6.4	3:25
2015-09-21	41	9.8	24	5.7	4:11
2015-09-22	38	10.8	22	6.2	3:32
2015-09-23	20	6.6	19	6.3	3:02
2015-11-23	14	4	13	3.7	3:31
2015-11-24	10	2.9	9	2.6	3:25
2015-11-26	30	6.6	25	5.5	4:34
2015-11-27	15	3.7	14	3.5	4:01
2015-12-01	4	1.5	3	1.1	2:43

2016-01-26	5	2.1	4	1.7	2:20
2016-01-28	12	3.8	11	3.5	3:10
2016-02-23	12	3.5	11	3.3	3:23
2016-02-24	15	3.4	13	3	4:24
2016-03-15	23	5	19	4.1	4:35
2016-03-16	22	4.2	18	3.4	5:14
2016-03-17	24	5.7	23	5.5	4:11
2016-03-18	26	5.6	19	4.1	4:40
2016-04-20	34	7.4	29	6.3	4:37
2016-04-21	36	7.8	27	5.9	4:36
2016-04-22	42	7.4	32	5.6	5:41
2016-05-23	23	6.6	15	4.3	3:29
2016-05-24	28	5.4	20	3.9	5:09
2016-05-25	30	5.8	19	3.7	5:11
2016-05-26	46	8	34	5.9	5:45
2016-05-27	37	7.8	23	4.8	4:45
2016-06-20	27	5.6	26	5.4	4:48
2016-06-21	47	8.2	34	5.9	5:45
2016-06-22	56	9.7	36	6.3	5:45
2016-06-23	35	8.2	24	5.6	4:15
2016-07-19	69	12.2	41	7.3	5:39
2016-07-20	49	8.8	33	5.9	5:35
2016-07-21	52	9.8	35	6.6	5:19
2016-08-10	11	4.6	8	3.4	2:23

2016-08-11	72	11.2	41	6.4	6:26
2016-08-12	64	10.7	35	5.8	5:59
2016-08-23	54	6.7	37	4.6	8:05
2016-08-24	55	8.8	31	5	6:14
2016-08-25	34	5.5	21	3.4	6:10
2016-10-04	52	10.4	35	7	5:00
2016-10-05	62	7.7	46	5.7	8:04
2016-10-06	41	8	28	5.5	5:07
2016-10-07	33	9.5	25	7.2	3:28