Answers to Reviewer 1:

General comment:
This paper describes a large set of ship emission measurements carried out downwind of by passing ships in the Elbe River. The method has been developed and used within the scientific community during the last 10 years. The paper is well written. It has focus on compliance monitoring and considering that the shipping sector has been forced to decrease their emission by an order of magnitude from January 1st 2015, by running 0.1% sulfur fuel content instead of 1 % on northern European waters, these data are very interesting, since they correspond to one of the first data sets. For the reason above I believe the paper should be published. The paper should however be improved in the technical discussion regarding the quality etc.
We thank the reviewer for the general positive comments and the recommendation. Below, we reply point-by-point to the specific comments. As far as possible, we have considered the suggestions in the revised manuscript.

Specific comments:
Row 28, p 11033: The author mentions MGO but not the fuel many ships seems to be using, i.e. desulfurized heavy fuel oil.
We have changed the manuscript to include different kinds of fuels besides MGO.

Row 7, p 11036: We have some own experience on similar measurements showing that 1 minute resolution is somewhat slow and that ship plumes drifts by rather quickly. It would be good if the author can discuss /elaborate on this issue further and include in the uncertainty discussion.
The width of our peaks is typically between one and five minutes, even if we look at the CO2 data in one second resolution. This agrees well with an example given in Beecken et al., 2014b, Figure 1, where the ground based measured plume has a width of about 2 minutes. Peaks that are too narrow will become too small when averaged to 1 minute and are thus excluded from our analysis. Increasing the resolution to seconds would probably improve the ability to detect smaller peaks as well as to distinguish between plumes of ships that are close together but not increase the accuracy of our measurements.

Row 13, p 11036: The SO2 fluorescence technique has cross sensitivity to NO. Has this been assessed and corrected for?
Yes, there is a NO cross sensitivity for our SO2 device which has been assessed via calibration with six different NO concentrations ranging between 100 ppb and 450 ppb. It showed that 0.8 % of NO is measured by our SO2 instrument as a SO2 signal. With our measurements of NO parallel to SO2, we can estimate this cross sensitivity for each measured plume individually and correct the SO2 data.
Since we have underestimated this effect in our analysis of the January 2015 data set, we have reanalyzed the data with NO correction and changed Figure 4 accordingly. This improves the quality of our data set but does not change the message of our manuscript. We thank the reviewer for his suggestion.
We have changed the manuscript in section 2 Instrumentation to: "*There is a NO cross sensitivity for SO2 which gives for 0.8 % of the NO signal an SO2 signal. We have determined this value of 0.8 % via a set of 6 calibration measurements of different NO concentration between 100 and 470 ppb.*"
Also in section 3 Data analysis: "*The peak area value of the SO2 peaks is corrected with 0.8 % of the peak area value of the NO peaks to account for the cross sensitivity*."
We have added the Institute for Hygiene and Environment to the acknowledgements, who help us with the additional calibration.

Row 3, p 11037: The LICOR instrument is a rather nonlinear instrument. The spa calibration carried out was rather crude from 306 to 990 ppm corresponding to a too large interval in my mind, since the instrument is nonlinear in this interval. The author should elaborate on this issue in the uncertainty discussion

According to the specifications of our LICOR 840A instrument, its measurement range is from 0 to 3000 ppm and company calibration shows that it is linear within this range. We have chosen our calibration gases to lie within the range and have a low and a high value. Measurements of both calibration gases show within the uncertainty of the gas concentration only an offset, which is corrected for.

Row 3 to row 16, p 11039: The problem of obtaining good CO2 baseline values in a measurements site inland with long averaging time should be discussed. Also the nonlinearity of the CO2 sensor (see above).

The baseline is determined via a succession of three centered running means where peaks are excluded after each run and data is interpolated for the excluded peak. This gives a good baseline. Periods with very variable background signals where the baseline determination is not acceptable are excluded from our analysis. We have added to the manuscript: "*Background signals for each gas are determined via a customized running mean filter. Only for those events for which there was a significant CO2 peak and a clearly determinable background, are the SO2 signals analysed.*"

Row 3 to row 16, p 11039: Since ships today run on low sulfur fuel content the precision in the SO2 measurement, and the baseline assessment etc, is more difficult since there is very little signal, especially for small ships. This should increase the SO2 uncertainty to higher than 15-30% which is the typical uncertainty found in other studies for instance by Balzani et al. Also the threshold of 0.15% is questionable in my mind for the same reason.

We have a quite strict quality control for the peaks we are analyzing to prevent the increase of uncertainty in our analysis, which is a compromise between the number of ships we can analyse and the quality of our data. For 10 % of our analysed ships we have a detection limit for the sulfur content of 0.05 % due to low CO2 values. For the rest of our data, we are confident in our uncertainty of up to 30 % and better when the peak concentrations are higher.

Row: 27 p 110039; I believe the referred paper (Beecken 2014a) is about airborne measurements on the open sea while here the measurements is an inland measurements. The authors should discuss this.

Yes, the data we referred to is measured by in-situ instruments from airplane. Although the conditions are different and measurements are more difficult, we still think the data is comparable. We have commented this in the manuscript: "*This is better than previously published compliance rates of 85 % of 174 ship plumes (Beecken et al, 2014a), although it should be noted that this study did not describe the uncertainty considerations and was measured by airplane on the open sea. The latter may imply that compliance might not be so high when no direct control is possible. Compliance rates at other locations for land based measurements show values of 90 % of 255 ship plumes and 97 % of 211 ship plumes (Beecken et al, 2014b).*"

Row 12, p 12: These data are presumably the first published in ACP for 2015 data but to my knowledge there are phd dissertations including similar results.

Yes, there is a PhD dissertation from Jörg Beecken in 2015, in which first measurements of January 2015 are presented. We have referenced this phd thesis and changed the phrasing in our manuscript: "*There are preliminary results for first SFC measurements in January 2015 presented in Beecken, 2015, which are comparable with our measurements, although with slightly higher uncertainty and lower compliance rates.*"

Row 17, p 11041: Even though the oil price is so low that MGO now has the same price as HFO last year the difference between the two still remains.

That is true, but the low prices might still help with the acceptance of the new regulations. We have rephrased this sentence: "*It should be noted that the global oil price and thus MGO costs for the needed sulphur quality in January 2015 was the lowest since 2009, which could have a positive influence on the acceptance of the new regulation.*"

Figure v2. Is the data NO (as written in labels and text) or NOx. Why not NOx?
We have shown in Figure 2 the comparison between NO data in 2014 and January 2015 since NO is a parameter which we are using in our analysis. We could use NOx data as well but it would show similar behavior.

Technical Corrections:
The paper is well written in most places.
Thank you!

Row 15. P 110033: insert which between and which are basically Row 9 p 11034: change to "and, when suspicion is raised, take… "
Changed as suggested.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 11031, 2015.

Answers to Reviewer 2:

Page 11035, Line 9: abbreviation of ft is not clear.
We have changed this to "*foot*".

Page 11036, line 10: What is mean by 'certified instruments'?
The instruments are certified according to directives of the European Union for air quality measurements: EN 14212 for SO2, EN14211 for NOx, and EN14625 for O3. We have added this to the manuscript.

Page 11036, line 23: unclear what is meant by: difference of total NO. Should it be NOx and NO without conversion?
Yes, it should be NOx and we have changed this in the manuscript.

Page 11037, line 15 – 22: Why the AIS data is not used to identify all peaks?
We could associate every NO peak to an individual ship with the AIS data before analysing the CO2 and SO2 signal, but we would have to exclude those peaks without clear signals later anyway and would not gain any additional information.

Page 11037, line 22-25: It seems difficult to calculate the peak area since the response times of SO2 analyser and the CO2 analyser are so different and the data collection frequency is 1 min-1 i.e. the response time of the SO2-analyzer, frequency of the data collection and the duration of the peak concentrations are of the same order of magnitude. Please explain in more detail how the peak areas are calculated?
We calculate the peak area for each gas individually. After subtracting the background for the gas, we identify the peak maximum, and the start and end points of the peak. All values from start to end are added up to be the peak area. Using 1 minute data collection frequency is a compromise between better accuracy of our instruments with their longer response times (especially SO2) and easier data analysis on the one hand and better resolution of the peaks and better assignment of ships to peaks close together on the other hand.

Page 11037, line 23: How to define the background with respect of the peaks?
Please see the answer to reviewer 1. We have changed the manuscript to: "Background signals for each gas are determined via a customized running mean filter. Only for those events for which there was a significant CO2 peak and a clearly determinable background, are the SO2 signals analysed."

Page 11039, line 4: There are uncertainty sources that have different contribution for different analysers: moisture, cross interferences by other chemical species. How, e.g. the contribution of NO on SO2 measurements has been taken into account?
We do have cross sensitivity with NO and we have reanalysed our data to include this effect. Please see the answer to reviewer 1.
We have changed the explanation of peak analysis to: "*Background signals for each gas are determined via a customized running mean filter. Only for those events with a significant CO2 peak and a clearly determinable background, SO2 signals are analysed. For all peaks the individual peak area above the background concentration is determined. This accounts for the difference in peak width for each gas due to different time resolutions of the respective instruments. The peak area value of the SO2 peaks is corrected with 0.8 % of the peak area value of the NO peaks to account for the cross sensitivity.*"

Page 11039, line 16: How to end up the uncertainty of measurements of 15 %. This may be too low estimation especially at the range of 10 ppb.

The uncertainty of 15 % is the best case scenario where the peaks are large compared to the background noise and a well-mixed plume is assumed with a conversion efficiency of 1 % from sulphur to SO2. Our measurements lie well within the range of 15 to 30 percent uncertainty especially for sulphur contents around 0.1% and higher.

Page 11040 line 10. Rounding the 0.1 % limit up to 0.15 % may be speculation and cause a 50 % increase from the limit. Please explain is this assumption in consistent with the legislation?
This is just a suggestion on how to handle information about sulphur content obtained by in situ instruments such as our system and give a value to define what is compliant with the regulations or not. We have chosen this value of 0,15 % which exceeds our uncertainty, to be sure that we do not accuse ships of a regulation violation which was caused by the uncertainty of our measurement, and also such that other groups with potentially higher uncertainty of their measurement systems can still compare their results with ours.

Page 11041 line 14-15. The reader gets very easily the idea that 95.4 % of ships follows the rules for sulphur content of ship engine fuel while you were able to measure 10 to 40 % of the whole ships? Please clear the sentence. Does this method apply if the ship is using different fuel (LNG), use heavy oil but scrubbing technique? Please discuss this issue.
We have rephrased the sentence to be clearer about the number of compliant ships in accordance to the number of measured ships: "*Our data shows that the vast majority (95.4 %) of all the ships we have measured are indeed complying with the new regulation of 0.1 % sulphur fuel content.*"
Since we do not know about the fuels the ships are using or if they use scrubber techniques, we can only estimate a potential sulphur content assuming the ships burn fuel with that sulphur content. Therefore, this method could be used to estimate whether different techniques such as LNG or scrubber technique have comparable results of SO2 reduction to the use of low sulphur fuels. However there are still few ships who use alternative methods and this issue would need further research.

Monitoring compliance with sulphur content regulations of shipping fuel by in-situ measurements of ship emissions

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Abstract

In 1997 the International Maritime Organisation (IMO) adopted MARPOL Annex VI to prevent air pollution by shipping emissions. It regulates, among other issues, the sulphur content in shipping fuels, which is transformed into the air pollutant sulphur dioxide (SO_2) during combustion. Within designated Sulphur Emission Control Areas (SECA), the sulphur content was limited to 1 %, and on January 1st, 2015, this limit was further reduced to 0.1 %. Here we present the setup and measurement results of a permanent ship emission monitoring site near Hamburg harbour in the North Sea SECA. Trace gas measurements are conducted with in-situ instruments and a data set from September 2014 to January 2015 is presented. By combining measurements of carbon dioxide (CO_2) and SO_2 with ship position data, it is possible to deduce the sulphur fuel content of individual ships passing the measurement station, and thus, facilitating monitoring compliance of ships with the IMO regulations. While compliance is almost 100 % for the 2014 data, it decreases only very little in 2015 to 95.4 % despite the much stricter limit. We analysed more than 1400 ship plumes in total and for months with favourable conditions up to 40 % of all ships entering and leaving Hamburg harbour could be checked for their sulphur fuel content.

1 Introduction

Shipping is a major part of the global transportation sector and its importance is still growing. According to the United Nations Conference on Trade and Development's Review of

Maritime Transport, in 2013 a total of 9.6 billion tons were transported via ships. This corresponds to a growth rate of this sector of 3.8 % per year. (UNCTAD, 2014) Despite being the most efficient and least emitting mode of transportation per ton of cargo compared to land based or airborne transport, shipping emissions nevertheless are a considerable fraction of total anthropogenic emissions and have a significant impact on air quality of coastal areas. 70 % of shipping emissions are produced within 400 km off the coasts (Corbett et al., 1999) and can cause severe health and environment problems to these regions (Corbett et al., 2007, Eyring et al., 2010).

The International Maritime Organisation (IMO), an agency of the UN with 171 Member States, has decided on measures to limit the impact of shipping emissions by adopting on the MARPOL Annex VI protocol in 1997. One part of these measures and the one, on which this study focuses, is the reduction of sulphur in ship fuel in order to reduce sulphur dioxide (SO_2) emissions. When oxidised, SO_2 forms small sulphate particles, which have an effect on cloud properties and change their reflectivity and lifetime (Lauer et al., 2007). SO_2 emissions by ships lead to an enhanced sulphate concentration of 10 - 50 % in coastal areas (Matthias et al., 2010), which increases acidification by acid rain (Endresen et. al., 2003). Gaseous SO_2 as well as sulphate particles have health effects on humans, when inhaled. SO_2 is produced during the combustion process by burning sulphur that is contained in the fuel. Ship engines have been developed to be able to burn Heavy Fuel Oils (HFO) that have a very high sulphur content of up to several percent and are basically a waste product of oil refineries and thus very cheap.

The IMO regulations concerning sulphur content came into force in 2005 and were revised in 2008, the revision came into force in 2010. On all oceans worldwide, the allowed sulphur content in HFOs was capped at 4.5 %, and after 2012 this limit was reduced to 3.5 %. In addition, so called “Sulphur Emission Control Areas” (SECA) were established with an even further reduced sulphur limit. One SECA is along the North American Coast, and another one comprises the Baltic Sea and the North Sea up to the Shetland Islands and to the western entrance of the English Channel. Within these SECAs the sulphur limit was initially set to 1.5 %, which was reduced to 1.0 % in 2010 and has now reached its current reduction step in January 2015 with a limit of 0.1 %.

While the 1 % limit could still be met with sulphur reduced HFO, the new regulation forces ships to either use more expensive alternatives such as Marine Gas Oil (MGO), or Ultra low

sulphur HFO, or consider reconstruction to enable the use of alternative fuel such as liquefied natural gas (LNG) or methanol. As an alternative technology, the operation of exhaust gas cleaning systems (scrubbers) is also permitted, as long as it provides the same level of protection against sulphur dioxide emissions as the use of low sulphur ~~MGO~~fuel. These alternative options have been deployed to some ships and first studies have documented their effectiveness and economic efficiency (Reynolds, 2011, Jiang et al., 2014), but they are still under development and not very widespread, and for the vast majority of ships the only option to meet the regulations is to use desulphurised fuel ~~MGO~~.

With the regulations in place, the question remains on how to efficiently verify compliance of the ships. To date, compliance is checked by inspection authorities who enter ships at berth, review fuel log books and fuel quality certificates and, when suspicion was raised, take a fuel sample to be analysed at certified laboratories. With the results of these analyses, it is possible to verify compliance and if needed, take legal actions. However, these controls can check just a minor number of ships. It is also not possible to evaluate the performance and compliance of scrubber technology by sulphur prediction in bunker oil samples which would be problematic if this method becomes more popular and common in future. Another problem is to control ship fuel of ships on the open sea.

For these reasons, several studies have suggested the implementation of air quality measurement systems especially aiming at the surveillance of ship emissions. One simple but efficient method is direct and simultaneous measurements of pollution trace gases with in-situ instruments. These instruments can quite easily be adapted to measurement conditions on airplanes, research vessels and trucks and have been used in a variety of campaigns in recent years (Sinha et al., 2003, Schlager et al., 2006, Agrawal et al., 2009, Williams et al., 2009, Diesch et al., 2013, Balzani Lööv et al. 2014, Beecken et al., 2014b). Based on the experience from those studies, we have established a measurement station near the harbour of Hamburg to monitor ship emissions, to estimate sulphur contents of fuel on board of passing individual ships. Our ship emissions dataset from September 2014 to January 2015 documents the quality of implementation of the MARPOL VI regulation with respect to compliant sulphur content in shipping fuel used in SECAs and follows the recent strong tightening of the regulation on January 1st 2015.

2 Measurement Site and Methods

The measurements reported here were conducted as part of the Mesmart project, a cooperation between the University of Bremen and the German Federal Maritime and Hydrographic Agency.

2.1 Measurement Site

Hamburg harbour is the third largest harbour in Europe and the 14th largest worldwide. In 2014, it had a 20-foot standard container throughput of 9.7 billion containers according to Hamburg port statistics. On average there are 800 calls per month, of which more than half are container vessels, and the other half consists mainly of Reefer vessels, tankers and bulk carriers. The harbour is located at the mouth of the river Elbe about 110 km inland, see Figure 1.

Measurements were conducted next to the river Elbe in the town of Wedel, which is near Hamburg, on the property and with the support of the Waterways and Shipping Office Hamburg. The instruments were set up right at the northern banks of the Elbe, with an approximate line of sight distance to ships leaving and entering Hamburg harbour of 0.3 and 0.5 km respectively. The average main wind direction at this location is with a southerly component, so that most of the time within the measurement period, the exhaust plumes of the ships were blown to the instruments. The area in the main wind direction south of the measurement station and the Elbe River is rural and sparsely populated with no significant sources of air pollution. Thus the location of the monitoring site is optimal for relatively low background concentrations of nitrogen oxides (NO_x) and SO_2.

2.2 Instrumentation

The concentrations of SO_2, NO_x, CO_2, and Ozone (O_3) were measured continuously with individual instruments, which are combined in a temperature stabilised box to ensure stable measurement conditions and at the same time provide a compact and transportable set-up. Data are stored in an integrated data logger with the time resolution of one minute. Despite different time resolutions of the instruments, we used data normalised to one minute, which is sufficient for the analysis of emission events with a duration in the order of several minutes.

NO_x, SO_2 and O_3 were measured with instruments from the Horiba AP-370 series, which are certified instruments according to EU directives (EN14211 for NO_x, EN 14212 for SO_2, and

EN14625 for O_3) used by German authorities for standard air pollution measurements. CO_2 was measured with a Licor 840A analyser. The O_3 measurements were not used for this study and are just mentioned for completeness.

SO_2: The Horiba APSA-370 is based on the UV-fluorescence method, using the excitation of SO_2 molecules by UV light and measuring the fluorescence which is a function of SO_2 concentration. The response time of the instrument is specified to be less than 120 s. Calibration was carried out with a standard gas mixture from Air Liquide with a concentration of 99.7 ppb SO_2 with an accuracy of 5 %. In addition, a daily control was obtained by the measurement of zero gas produced with a scrubber, and span gas from an internal permeation source with 175 ppb SO_2. There is a NO cross sensitivity for SO_2 which gives for 0.8 % of the NO signal an SO_2 signal. We have determined this value of 0.8 % via a set of 6 calibration measurements of different NO concentration between 100 and 470 ppb.

NO_x: The Horiba APNA-370 measures the chemiluminescence of NO molecules reacting with O_3. To obtain information about the NO_2 concentration, the device contains a deoxidation converter to transfer NO_2 molecules to NO. The NO_2 concentration is calculated by the difference of total NO_x, representing NO + NO_2, and NO without conversion. The response time for measurement of both gases is 90 s. The instrument is calibrated with an Air Liquide standard gas mixture with a concentration of 216.0 ppb NO and an accuracy of 5 %. A daily control with scrubber produced zero gas and a NO_2 span gas of 105 ppb is also implemented.

CO_2: The Licor 840A is a non-dispersive infrared gas analyser. It has a response time of 1 s and was calibrated with two Air Liquide standard gas mixtures with 306.6 ppm and 990.0 ppm CO_2 with an accuracy of 2 %.

The trace gas measurements were complemented with measurements of wind, temperature, air pressure and precipitation by a compact weather station (Lufft WS600). With an AIS (Automatic Identification System) receiver the information transmitted by passing ships was collected, which includes identification number, name and type of the ship as well as position, course, and speed.

3 Data analysis

To obtain the sulphur content of ship fuel in use, the enhancement of SO_2 and CO_2 in measurements affected by exhaust gases is measured, and the ratio of these SO_2 and CO_2

peaks is used to calculate the fuel sulphur content. The combination of the trace gas peak time, the wind direction, and the AIS information enables the identification of the peak related ship.

When wind conditions are favourable for measurements, the exhaust plumes of ships passing the instrument leave a distinctive enhancement of the measured component against background concentrations. Since this enhancement is most significant in NO measurements, and NO is an indicator for recent combustion processes, these NO peaks are used to identify the time stamp of a ship emission event. For these time stamps peaks in CO_2 are then identified, which is more complicated because background concentrations are larger and more variable due to the surrounding vegetation. Background signals for each gas are determined via a customized running mean filter. Only for those events for which there was ~~with~~ a significant CO_2 peak and a clearly determinable background, are the SO_2 signals ~~are~~ analysed. For all peaks the individual peak area above the background concentration is determined. This accounts for the difference in peak width for each gas due to different time resolutions of the respective instruments. The peak area value of the SO_2 peaks is corrected with 0.8 % of the peak area value of the NO peaks to account for the cross sensitivity. With the assumption that fuel contains 87±1.5 % carbon (Cooper et al., 2003) and 100 % of the sulphur and the carbon content of the fuel are emitted as SO_2 and CO_2 respectively, the sulphur fuel content (SFC) mass percent can be calculated as follows:

$$SFC[\%] = \frac{S[kg]}{fuel[kg]} = \frac{SO_2[ppm] \cdot A(S)}{CO_2[ppm] \cdot A(C)} \cdot 87[\%] = \frac{SO_2[ppb]}{CO_2[ppm]} \cdot 0.232[\%] \quad (1)$$

where A(S) is the atomic weight of sulphur and A(C) of carbon. Using this formula, it is relatively simple to calculate the sulphur content for each set of peaks. For a discussion about the uncertainties of this formula see section 3.1.

The second part of the data analysis is the attribution of the identified emission events to individual passing ships. Within 30 minutes before each event, which is characterised by the time the emissions arrive at the instruments, the AIS data is analysed for ship positions close to the measurement site. In combination with wind information this yields in most cases the

identification of the individual ships having caused the emission. The time the exhaust plume travels from being emitted to being analysed is about 2 to 10 minutes dependent on wind speed and direction. However, there are events in which there are two or more ships too close to each other, or where no AIS signal was received, such that no single ship can be associated to the signal. These events are excluded from the data set.

3.1 Uncertainties

There are several aspects that influence the accuracy of the calculated values of the sulphur content for each ship. The SFC-formula (1) assumes a 100 % conversion from sulphur to SO_2 during combustion, which is only true for an idealised combustion process. There is a range of uncertainty with respect to the amounts of sulphur oxidised and released as particles. Studies found that there could be an underestimation of the sulphur fuel content between 1 - 19 % from assuming complete conversion (Schlager et al., 2006, Agrawal et al., 2008, Moldanova et al., 2009, Balzani Lööv et al., 2014).

The uncertainty or sum of systematic and random error of our measurements is determined from a combination of the calibration uncertainty and the uncertainty resulting from the signal to noise ratio (SNR). CO_2 values with a SNR of less than five are excluded from the data, which leads to an upper limit uncertainty of 20 %. However, the majority of CO_2 values has an uncertainty of around 10 %. For SO_2 we do not exclude data with a low SNR because these are the zero sulphur content cases. The SNR of SO_2 data for a sulphur content of around 0.1 % is 10 or better, with a decrease for lower sulphur content values. For an SNR below 5 we consider the SO_2 signal as zero. This is only important for the January 2015 data, since the measured SO_2 concentrations in 2015 are much lower than for the 2014 data. This is shown in Figure 2 as a comparison between one week in December 2014 and one week in January 2015 with similar weather conditions. While no reduction in NO values can be observed, there is a large reduction in SO_2 values as expected.

All uncertainties added up with the root of sum of squares method, this gives us an uncertainty range for the sulphur content calculations of 15 – 30 %.

4 Results

Using the method described above we were able to identify 824 ship plumes of 474 individual ships within the months of September, November and December 2014. Unfortunately no data are available in October due to instrumentation problems. This data set is the so called pre-regulation-change set, where the regulatory allowed sulphur fuel content for the ships of is 1.0 %. The January 2015 data set consists of 589 ship plumes of 374 individual ships, which since the 1st of January 2015 have to comply with the new 0.1 % rule. As shown in Figure 3, the difference between these two data sets is remarkably obvious.

In the pre-regulation-change data set, 99.6 % of all ships complied with the 1 % sulphur limit with respect to the measurement uncertainty. This is better than previously published compliance rates ~~at other locations~~ of 85 % of 174 ship plumes (Beecken et al, 2014a), although it should be noted that this study did not describe the uncertainty considerations~~-~~ and was measured by airplane on the open sea. The latter may imply that compliance might not be so high when no direct control is possible. ~~(Beecken et al, 2014a),~~ Compliance rates at other locations for land based measurements show values of 90 % of 255 ship plumes and 97 % of 211 ship plumes (Beecken et al, 2014b). However, a study of Diesch et al., 2013, that describes measurements with a mobile laboratory along the Elbe River near our measurement site, found for 139 ship plumes a compliance of nearly 100 %. This could possibly be credited to the special location of Hamburg harbour where ships have to go up the Elbe for more than 100 km.

In accordance with the practice in use that fuel samples analysed in laboratories are considered as exceeding the 0.1 % sulphur limit in a legally binding way above the value of 0.149 %, we suggest to use a corresponding value of 0.15 % as a limit value for discussing the compliance of the ships in our January 2015 data set. This is in consistence with the formerly stated measurement uncertainties. In Figure 4, a more detailed graph of the January 2015 data is shown. The red line shows the 0.1 % limit with the shaded area indicating a conservative 30 % measurement uncertainty. The blue line indicates the suggested 0.15 % limit for compliance discussion. Of all the ships measured in January 95.4 % were complying with the new regulation. There are preliminary results for first SFC measurements in January 2015 presented in Beecken, 2015, which are comparable with our measurements, although with slightly higher uncertainty and lower compliance rates.

Color-coded in the Figures 3 and 4 are the lengths of the ships in 50 m size steps. Even before the regulation change ships smaller than 100 m did not use fuel with sulphur values higher than 0.2 %, most likely because their engines cannot process such fuels or storage capacity for two different kinds of fuels is not available. After the regulation change, those smaller ships still do not use the fuels that reach up to the allowed 0.1 % limit. If one considers only those ships larger than 100 m that could choose which fuel to use and had to change their way of operation, the compliance drops to 93 %.

The number of ships that can be detected for compliance depends strongly on the wind conditions. Assuming the average number of calls in Hamburg harbour according to Hamburg port statistics of 800 ships per month means that 1600 emission events happen at our measurement station of ships on their way in and out of the harbour. For months with good wind conditions like December 2014 and January 2015, we can detect about 30-40 % of those events, for month with unfavourable wind conditions like November 2014, this value drops to less than 10 %.

5 Conclusions

In this study, we have used the method of in-situ measurements of trace gases to implement a system to monitor compliance of ships with sulphur fuel content regulations. This has been discussed and suggested before (Balzani Lööv et al., 2014). Here we present a suitable location for permanent stationary measurements near Hamburg harbour, one of the largest harbours in Europe, and demonstrate a measurement approach that successfully characterises emissions from passing ships. We describe the method used to identify ship emission events and the corresponding ships and present a large data set on fuel usage of ships of altogether 1413 analysed ship plumes. This includes the one of the first data sets, after the most recent regulation change in the North Sea SECA, where fuel sulphur content limits were reduced from 1 % to 0.1 % on January 1st, 2015.

Our data shows that- the vast majority (95.4 %) of all the ships we have measured are indeed complying with the new regulation of 0.1 % sulphur fuel content. Compliance has dropped slightly compared to the value of more than 99 % observed for the 1 % sulphur limit in fall 2014. It should be noted that the global oil price and thus MGO costs for the needed sulphur quality in January 2015 was the lowest since 2009, ~~but we hope that not only economic issues~~

~~influence the decision whether to comply with environmental legislation or not~~which could have a positive influence on the acceptance of the new regulation.

With the described method it is possible to easily and reliably identify those ships that do not comply. It is possible to check 10 - 40 % of all ships entering and leaving the harbour, depending on wind conditions. This should be interesting to government agencies in charge of the control of the SECAs.

Acknowledgements

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References

Agrawal, H., Welch, W. A., Miller, J. W., and Cocker, D. R.: Emission measurements from a crude oil tanker at sea, Environ. Sci. Technol., 42, 7098–7103, 2008.

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

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

Beecken, J., Mellqvist, J., Salo, K., Ekholm, J., Jalkanen, J. P., Johansson, L., Litvinenko, V., Volodin, K., and Frank-Kamenetsky, D. A.: Emission factors of SO_2, NO_x and particles from ships in Neva Bay from ground-based and helicopter-borne measurements and AIS-based modeling, Atmos. Chem. Phys. Discuss., 14, 25931 – 25965, 2014b.

Beecken, J.: Remote Measurements of Gas and Particulate Matter Emissions from Individual Ships, PhD thesis, Chalmers University of Technology, 2015.

Cooper, D. A.: Exhaust emissions from ships at berth, Atmos. Environ. 37, 3817–3830, 2003.

Corbett, J. J., Fischbeck, P. S., and Pandis, S. N.: Global nitrogen and sulphur inventories for oceangoing ships, Journal of Geophysical Research: Atmospheres, 104, D3, 3457-3470, 1999.

Corbett, J. J., Winebrake, J. J., Green, E. H., Kasibhatla, P., Eyring, V., and Lauer, A.: Mortality from Ship Emissions: A Global Assessment, Environ. Sci. Technol., 41, 8512 – 8518, 2007.

Diesch, J.-M., Drewnick, F., Klimach, T., and Borrman, S.: Investigation of gaseous and particulate emissions from various marine vessel types measured on the banks of the Elbe in Northern Germany, Atmos. Chem. Phys., 13, 3603-3618, 2013.

Endresen, Ø., Sørgård, E., Sundet, J. K., Dalsøren, S. B., Isaksen, I. S. A., Berglen, T. F., and Gravir, G.: Emissions from International Sea Transportation and Environmental Impact, Journal of Geophysical Research: Atmospheres, 108, D17, 4560, doi:10.1029/2002JD002898, 2003.

Eyring, V., Isaksen, I. S. A., Berntsen, T., Collins, W. J., Corbett, J. J., Endresen, Ø., Grainger, R. G., Moldanova, J., Schlager, H., and Stevenson, D. S.: Transport Impacts on Atmosphere and Climate: Shipping, Atmos. Environ., 44, 4735 – 4771, doi:10.1016/j.atmosenv.2009.04.059, 2010.

Jiang, L., Kronbak, J., and Christensen, L. P.: The costs and benefits of sulphur reduction measures: Sulphur scrubbers versus marine gas oil, Transportation Research Part D, 28, 19-27, 2014

Lauer, A., Eyring, V., Hendricks, J., Jöckel, P., and Lohmann, U.: Global Model Simulations of the Impact of Ocean-Going Ships on Aerosol, Clouds, and Radiation Budget, Atmos. Chem. Phys., 7, 5061-5079, 2007.

Matthias, V., Bewersdorff, I., Aulinger, A., and Quante, M.: The contribution of ship emissions to air pollution in the North Sea regions, Environmental Pollution, 158, 2241 – 2250, 2010

Moldanova, J., Fridell, E., Popovicheva, O., Demirdjian, B., Tishkova, V., Faccinetto, A., and Focsa, C.: Characterisation of particulate matter and gaseous emissions from a large ship diesel engine, Atmos. Environ., 43, 2632-2641, 2009.

Reynolds, K. J.: Exhaust gas cleaning systems selection guide. Ship operations cooperative program. The Glosten Associates, USA, 2011.

Schlager, H., Baumann, R., Lichtenstern, M., Petzold, A., Arnold, F., Speidel, M., Gurk, C., and Fischer, H.: Aircraft-based Trace Gas Measurements in a Primary European Ship Corridor, proceedings TAC-Conference, 83-88, 2006.

UNCTAD RMT, United Nations Conference on Trade and Development, Review of Maritime Transport, 2014.

Figure 1: Location of the measurement station on the northern bank of the river Elbe, near Hamburg harbour. On the right: picture of instrument box. Map source: OpenStreetMap.

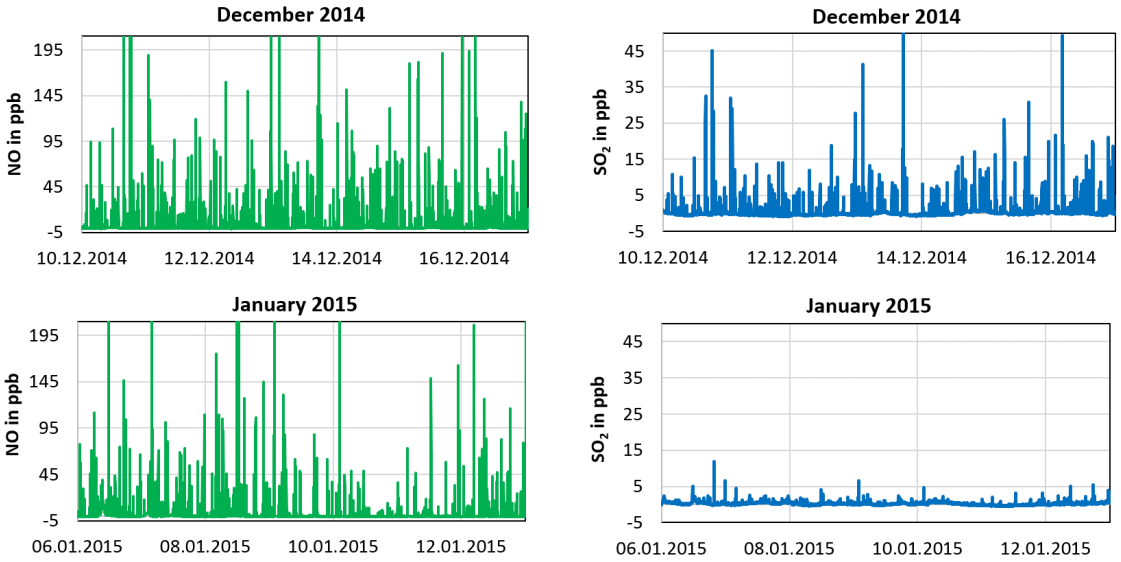
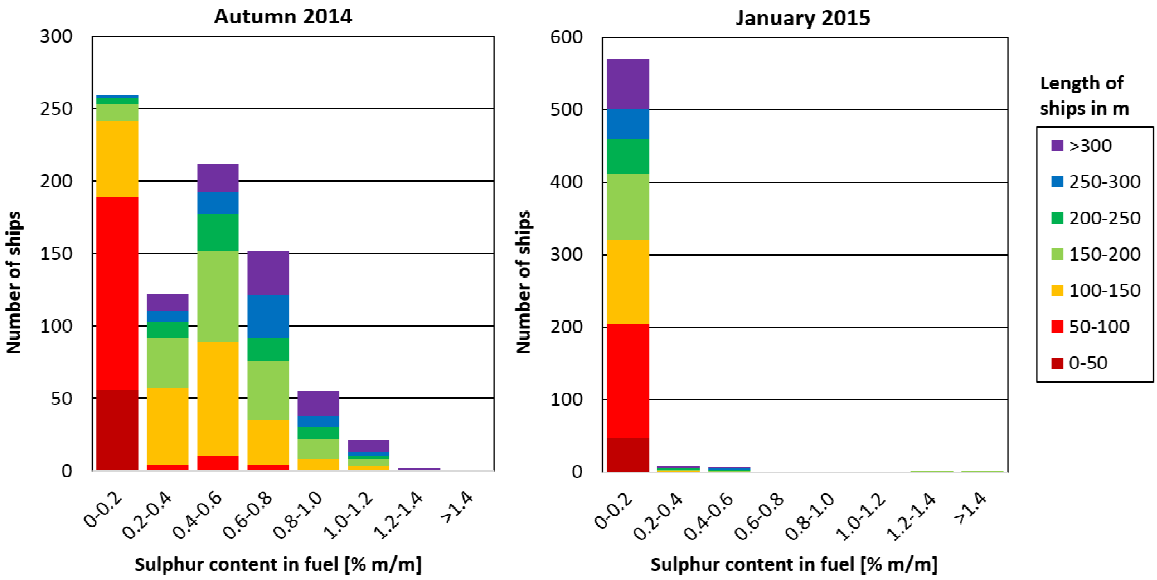


Figure 2: Comparison of absolute NO and SO_2 volume mixing ratio values measured over two weeks, one week in December 2014 and one in January 2015 with comparable wind conditions. Each peak belongs to one emission plume of an individual ship. The reduction in SO_2 in 2015 is obvious, while for NO no reduction can be observed.



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3 Figure 3. Sulphur fuel content in autumn 2014, and in January 2015, after the change of fuel
4 regulations. Colour-coded is the length of the ships that have been analysed. While in 2014
5 only small ships had fuel sulphur contents below 0.2%, nearly all ships fell into this category
6 in January 2015.

7

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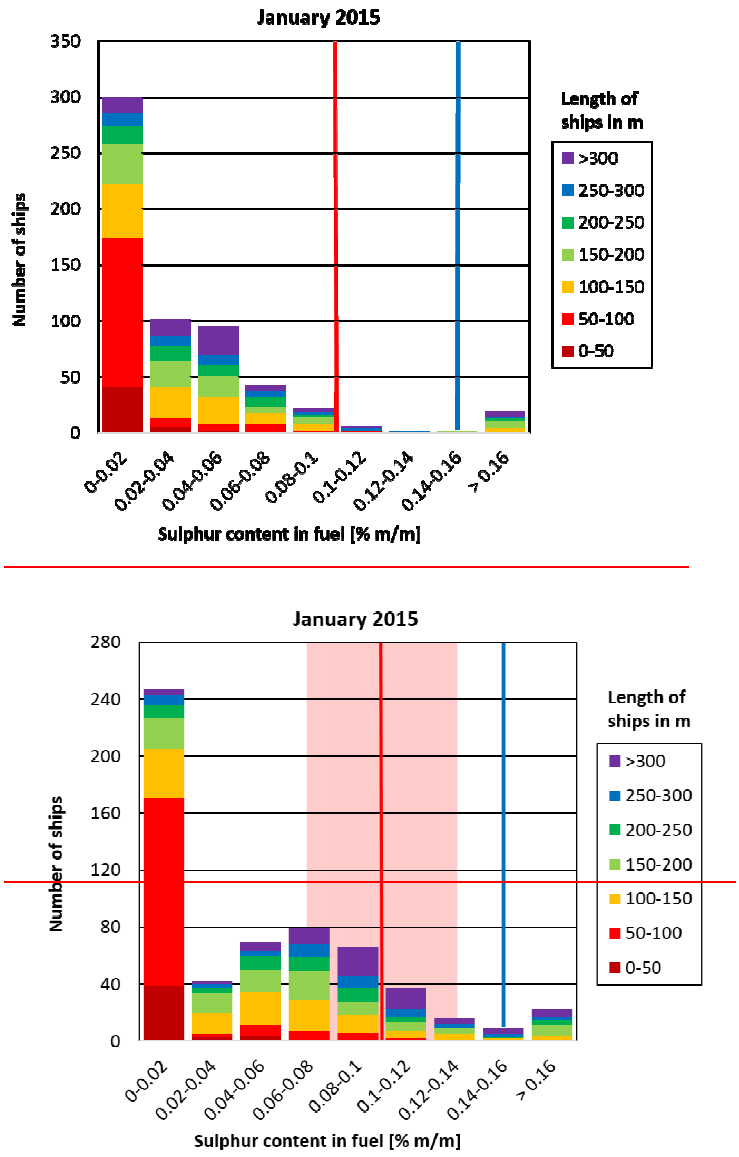

Figure 4: Detailed view of the January 2015 data set. Colour-coded is the length of the ships, the red line indicates the 0.1 % limit with the shaded area representing the upper limit uncertainty of 30 %. The blue line indicates the suggested limit of 0.15 % for flagging ships as exceeding the allowed sulphur fuel content limit.