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Research Article

Detection of Ship Fuel Sulfur Contents in Exhaust Plumes at the Kanmon Straits, Japan, before and after the Global Sulfur Limit 2020

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ABSTRACT The global limit on the sulfur content of ship fuel was reduced from 3.50% to 0.50% in January 2020 to reduce ship emissions of SO_2 and particulate matter. We conducted observational campaigns before and after the new global limit was introduced to detect changes in coastal air quality. We measured ambient concentrations of SO_2 and CO_2 ship plumes on shore with the sniffing method under the Kanmon Bridge over the Kanmon Straits between Honshu and Kyusyu Islands, Japan, for several weeks in August to September in 2019 and 2020. The fuel sulfur content (FSC) estimated from our measurements mainly varied from 0.50% to 3.00% in 2019, whereas the range narrowed to 0.10% to 0.40% in 2020, showing that all the ships complied. The mean FSC in 2020 was reduced to 16% of that in 2019, which was consistent with the reduction in the ambient SO_2 concentration. Sakurai *et al.* (2021) estimated that after the 2020 global limit was brought in, SO_2 emissions from ships were reduced to 24% of their previous values by assuming that all ships have a FSC of 0.50%. Our results indicate the 2020 global limit led to much greater reductions in SO_2 emissions from ships than expected.

KEY WORDS Fuel sulfur content, MARPOL Annex VI, SO_2, Sniffing method, Kanmon Straits

1. INTRODUCTION

On January 1, 2020, the global limit for fuel sulfur content (FSC) was reduced from 3.50% to 0.50% (mass/mass. Hereinafter, m/m) for all ships. This new regulation (hereinafter, the 2020 global limit) was agreed at the 70^{th} International Maritime Organization Marine Environment Protection Committee (MEPC) in 2016 to reduce emissions of sulfur oxides and particulate matter from ships. Ship emissions of sulfur dioxide (SO_2) accounted for 11% of the global anthropogenic total emissions (105,258 Gg) in 2015 (Crippa *et al.*, 2019). Stricter regulations can be set in Sulfur Emission Control Areas (SECA) introduced in MEPC. Currently, SECA operate in the Baltic Sea, the North Sea, the English Channel, and the United States of America and Canada, where the FSC has been limited to below 0.1% since 2015. Similar local regulations are also in force in Europe, China, and South Korea.

In Japan, the government decided that there was no need to introduce SECA because the effects of SECA on air quality would be limited or uncertain (Technical

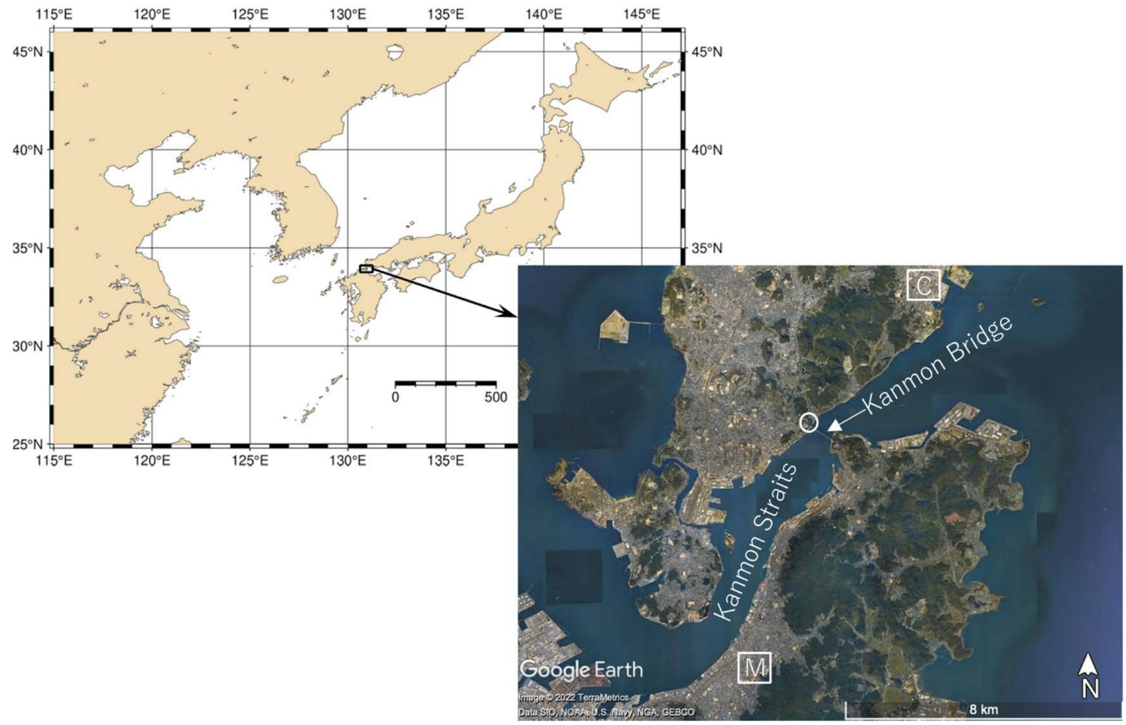

Fig. 1. Location of our monitoring site (white circle) along with the governmental monitoring stations (M: Moji and C: Chofu).

Committee on ECA, 2013). However, ships are still a dominant source of SO_2 in Japan, accounting for 16% of the total anthropogenic emission (1,113 Gg) in 2015 (Crippa *et al.*, 2019). More locally, the ship emissions are comparable to emissions from onshore stationary sources. According to Sakurai *et al.* (2021), the SO_2 emissions from ships (both navigating and anchored) were estimated as 14.7 Gg yr^{-1} in Tokyo Bay (1,380 km^2) and 55.0 Gg yr^{-1} in the Seto Inland Sea (21,827 km^2) in the Japanese fiscal year of 2015 (April to the following March), which corresponded to 56% and 41% of the total emissions in the surrounding onshore areas (9,612 km^2 and 61,260 km^2, respectively). Nakatsubo *et al.* (2020) analyzed ambient concentrations of SO_2 and fine particulate matter ($PM_{2.5}$) measured in 2016 and 2017 at three air quality monitoring stations along the coast of the Seto Inland Sea. They concluded that SO_2 mostly originated from ships and 17.3% to 21.4% of $PM_{2.5}$ concentrations came from ships. Ship emissions are so large that coastal air quality may be affected at certain fractions (Sorte *et al.*, 2020). Sakurai *et al.* (2021) also estimated that the 2020 global limit would reduce the ship SO_2 emissions to 24% of their pre-limit value in the Seto Inland Sea. It is expected that the 2020 global limit will have reduced the coastal ambient SO_2 concentration to 24% or less of its pre-limit value. The present study examines whether such drastic reductions really occurred through monitoring ship emissions before and after the 2020 global limit.

2. EXPERIMENTS

2.1 Observational Conditions

Our field campaigns were conducted at the Kanmon Straits (Fig. 1). The Kanmon Straits divide Kyusyu and Honshu islands and are about 600 m wide at the narrowest point near the Kanmon Bridge. Nearly 50,000 ships over 500 gross tonnage (G/T) pass through the straits per year (Kanmon Waterway Office, 2022). The waterways in the Kanmon Straits are the right-hand traffic, and ships on each lane run about 200 m from the shore at the Kanmon Bridge.

The field campaigns were performed before and after the 2020 global limit came into effect, from August 29 to September 11, 2019, and from August 26 to September 15, 2020. Ships may need to refill their tanks several times with low-sulfur fuel to replace old high-sulfur fuel remaining in the tanks completely. The 2019 campaign ended nearly 3.5 months before the 2020 global limit started. The ships were not expected to have changed their fuel at the time of the 2019 campaign.

2.2 Measurement Method

Much research has focused on measuring the atmospheric concentrations of the pollutants emitted from ships, especially in parts of Europe and North America where SECA have been set (e.g., Sorte *et al.*, 2020). Optical and sniffing methods have been used to measure these pollutants (Balzani Lööv *et al.*, 2014). The optical methods include differential optical absorption spectroscopy, differential absorption light detection and ranging, and ultraviolet cameras; however, these methods need a model for fuel combustion to estimate the FSC. The sniffing method is based on measurements by analyzers used for routine air quality monitoring. The sniffing method is more convenient and has the advantage that the FSC can be calculated directly. Both the optical and sniffing methods can be used on either fixed land-based or mobile platforms. Fixed platforms are good for continuous monitoring but may miss plumes that flow aloft or in different directions from ships. Mobile platforms can increase the likelihood of sampling by chasing plumes or ships from airborne or seaborne vehicles. We chose the sniffing method and set the analyzers at a fixed land-base station as described in Section 2.3.

In the sniffing method, the FSC can be obtained from carbon dioxide (CO_2) and SO_2 concentrations measured in the plume, as follows (Mellqvist *et al.*, 2017).

$$\mathrm{FSC} = 0.232 \frac{\int([\mathrm{SO_2}]-[\mathrm{SO}_{2,bkg}])dt}{\int([\mathrm{CO_2}]-[\mathrm{CO}_{2,bkg}])dt}$$

Here, the unit of FSC is % (m/m); $[CO_2]$ and $[SO_2]$ are the gas concentrations of CO_2 and SO_2 expressed in ppm and ppb, respectively; and the subscript *bkg* indicates the baseline concentrations. The constant 0.232 is the molecular weight ratio of sulfur (32 g mol^{-1}) to carbon (12 g mol^{-1}) multiplied by the carbon mass percent in the fuel (87%) and 0.001 ppb/ppm. The integral range may differ between CO_2 and SO_2, depending on the response time of instruments used. The estimated FSC is slightly lower (~5%) than the true FSC, because the fuel sulfur is not fully converted to SO_2 during combustion (Mellqvist *et al.*, 2017). However, we did not correct the estimated FSC. Mellqvist *et al.* (2017) estimated the overall uncertainty for FSCs of 0.1% and 1% obtained by the sniffing method as $0.1 \pm 0.04\%$ and $1 \pm 0.19\%$, respectively. The monitoring site at which Mellqvist *et al.* (2017) conducted their study was the Great Belt Bridge, Denmark, which has no stationary, industrial sources of SO_2 in the vicinity. In contrast, the Kanmon Bridge is more likely to be affected by sources other than ships. Our uncertainty in the FSC estimates may be higher than those reported by Mellqvist *et al.* (2017). Therefore, we decided to calculate FSCs only for apparent peaks. The criteria are described in Section 3.2.

2.3 Instruments

The SO_2 concentration was analyzed by the ultraviolet fluorescence method (43C-TLE, Thermo Electron). The SO_2 instrument was equipped with a hydrocarbon kicker, which removed hydrocarbons from the sampled air to reduce interference. For the sniffing method, the instrument is often operated without the hydrocarbon kicker to obtain a fast response time, (e.g., Mellqvist *et al.*, 2017). However, in our preliminary laboratory experiments before the 2019 campaigns, we found that the SO_2 measurements became unstable without the hydrocarbon kicker. The CO_2 concentration was measured with a non-dispersive infrared instrument (LI-7000, LI-COR). We also measured concentrations of nitrogen oxide (NO) and nitrogen dioxide (NO_2) with a chemiluminescence instrument (42C-TL, Thermo Electron). The response time is about 40 sec for the SO_2 monitor, below 30 sec for the NOx monitor, and below 1 sec for the CO_2 monitor. Instantaneous winds were measured with a vane anemometer (MVS-350, Koshin). Video cameras were set to

Fig. 2. View of the Kanmon Straits from the monitoring site under the Kanmon Bridge. All the instruments were stored in the shed.

record ships passing in front of the instruments. All the instruments were stored in a shed placed on shore facing the Kanmon Straits at ca. 5 m above sea level under the Honshu-side piers of the Kanmon bridge (Fig. 2). Signals from the instruments were sampled every second, and 10 s means were logged. Identification, position, course, speed and other information on the ships passing in front of the monitoring site was obtained from the Automatic Identification System (AIS).

3. RESULTS AND DISCUSSION

3.1 Meteorological and Air Quality Conditions

During the 2019 and 2020 campaigns, mean temperatures were 26.8 and 27.2°C and total rainfall amounts were 178.5 and 130.5 mm, respectively, at the nearest meteorological observatory in Shimonoseki. Fig. 3 shows the wind roses at the study site during the campaigns. The most dominant wind directions of ENE and E were from the straits. The frequencies and mean wind velocities had similar patterns and magnitudes in both campaigns. Therefore, the meteorological conditions were similar enough not to influence the air quality in the campaigns. However, both campaigns were affected by typhoons, and in the 2020 campaign, all the instruments were removed for three days from September 5, 2020, to avoid high wind speeds and tidal waves caused by Typhoon 202010 (Haishen). On the other days, the instruments fully worked except for calibration. In total, measurements of SO_2, NOx, and CO_2 were obtained for more than 353 and 346 h in the 2019 and 2020 campaigns, respectively.

Box plots in Fig. 4 show the ranges of the SO_2, NOx, and CO_2 concentrations during each of the campaigns. The NO concentration in the 2020 campaign reached the highest range of 200 ppb occasionally. The CO_2 concentration remained almost constant during the two campaigns, and the NOx concentration decreased slightly. In contrast, the SO_2 concentration decreased considerably, from 6.3 to 0.9 ppb for the mean, from 4.1 to 0.6 ppb for the median, and from 166.5 to 16.3 ppb for the maximum. Fig. 5 shows the frequencies of SO_2 by wind direction in the 2019 and 2020 campaigns. Although the absolute levels of SO_2 concentrations were different between the campaigns, SO_2 was mostly observed in wind directions from NE to ESE.

In general, the SO_2 concentration varies with meteorological factors, including wind direction, velocity, and stability, as well as by emissions from ships and other sources. According to the AIS data, ships passed the straits 3,060 and 4,170 times in total during the 2019 and 2020 campaigns, respectively. The number of passing ships per day was consistently around 200 for both years, of which

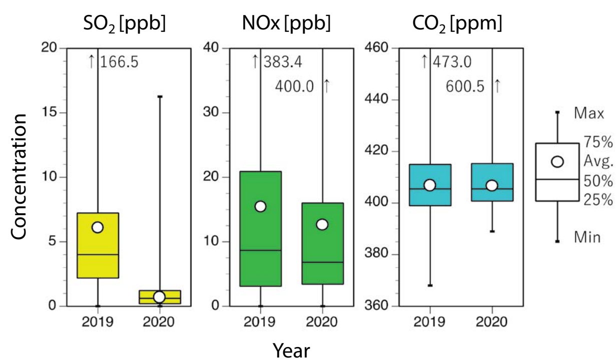

Fig. 4. Box plots of SO_2, NOx, and CO_2 concentrations in the 2019 and 2020 campaigns.

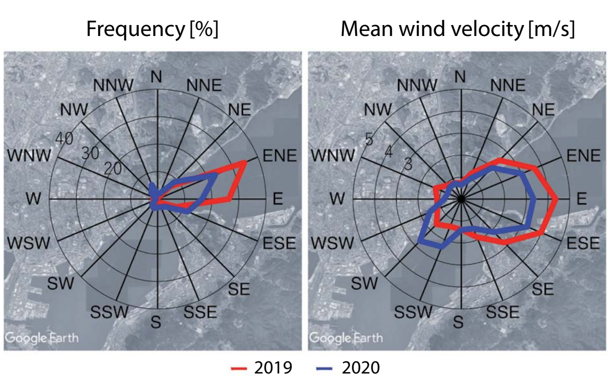

Fig. 3. Wind roses at the monitoring site in the 2019 and 2020 campaigns.

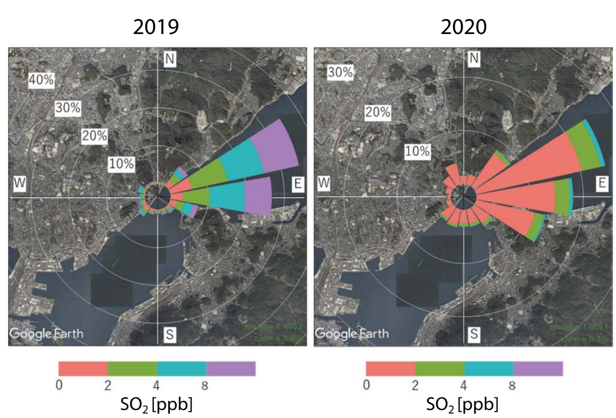

Fig. 5. Occurrence rate of SO_2 concentrations by wind direction.

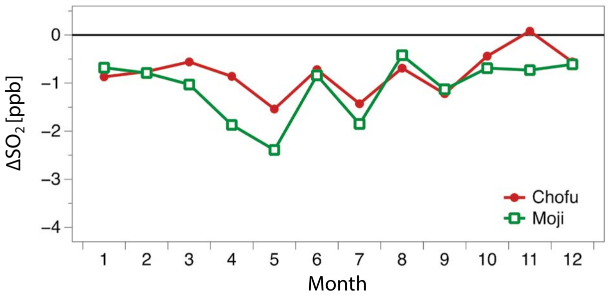

Fig. 6. Changes in the monthly mean SO_2 concentration (ΔSO_2) from 2019 to 2020 at the governmental monitoring stations.

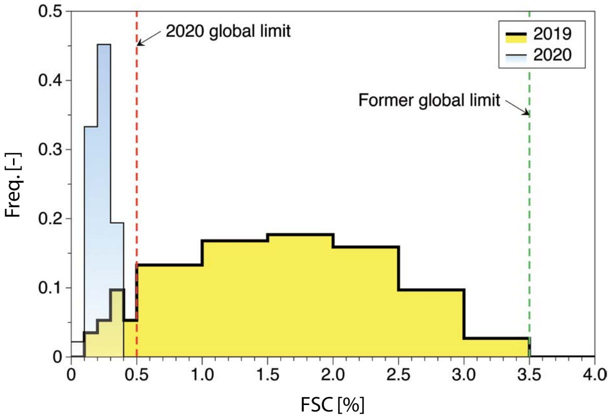

Fig. 7. Frequency distribution of FSC. The frequency is normalized by the total number of the identified peaks for each year.

about 60% were counted as cargo ships and nearly 30% as tankers. These results indicate that the activity of ships passing through the Kanmon Straits was similar during the 2019 and 2020 campaigns. In addition to the consistency in wind roses (Fig. 3), the results showed that the remarkable reduction in the SO_2 concentration compared with the tiny changes in the NOx and CO_2 concentrations was attributed to the 2020 global sulfur limit.

The reductions in the SO_2 concentration from 2019 to 2020 were also confirmed at governmental monitoring stations. Fig. 6 shows changes in monthly mean concentrations ($\Delta SO_2 = [SO_2]_{2020} - [SO_2]_{2019}$) measured at the two stations nearest to the Kanmon Straits, Chofu and Moji (see Fig. 1 for their locations). Even though the governmental monitoring stations record hourly mean concentrations of SO_2 in units of 1 ppb, the monthly mean concentrations decreased from 2019 to 2020 for nearly all the months. The monthly mean concentrations of SO_2 in September at the two stations decreased by 66% for Chofu and 40% for Moji. This reduction is smaller than that in our measurements at the Kanmon Straits (86%), probably because these two stations are more affected by sources other than ships. The large reductions in May might be associated with the national state of emergency declared in response to the COVID-19 pandemic, which needs further analysis.

3.2 FSC

The dominant wind directions for SO_2 were NE, ENE, E, and ESE (Fig. 5). The most frequent wind direction was ENE, which is nearly along the sea traffic lanes in the Kanmon straits. In this wind direction, plumes from ships in the lane arrive continuously at the measurement site over a long time, resulting in broad, undefined, or overlapping peaks in concentration. Therefore, we analyzed the FSC for peaks in wind directions between E and S. In addition, we set the following criteria in calculating the FSC: a CO_2 peak with height over 3 ppm and temporal duration within 3 min, and the corresponding SO_2 peak over 4 ppb with temporal duration within 3 min. Here, the background concentration was obtained by taking an average of concentrations at the beginning and ending of each peak. In the 2019 and 2020 campaigns, 113 and 93 peaks, respectively, were clearly identified and analyzed.

Fig. 7 shows the frequency distributions of the FSC normalized by the total number of the identified peaks for the respective campaigns. The FSCs in the 2019 campaign formed a broad distribution from 0.50% to 3.00% and were below the former global limit of 3.50%. For the 2020 campaign, all the FSCs were aggregated below the 2020 global limit of 0.50%. According to a governmental survey on maritime fuels (Maritime Bureau of the Ministry of Land, Infrastructure, Transport and Tourism, 2019), high-sulfur and low-sulfur fuels contain 0.61% to 2.86% (m/m) and 0.17% to 0.46% (m/m) sulfur, respectively. Our FSC results indicated high-sulfur fuels were used before the 2020 global limit and low-sulfur fuels were used after. Our results also suggested that all the analyzed ships complied with the global sulfur regulations of the time.

A bump in the FSC distribution below 0.5% in the 2019 results indicated that some ships used low-sulfur fuels before the 2020 global limit. Fig. 8 summarizes the number of ships by gross tonnage for the FSC below and over 0.50% from the AIS data. For FSCs of less than 0.50 %, 85% of ships were below 1,000 G/T. However, these small ships account for only 26% of ships, indicating that

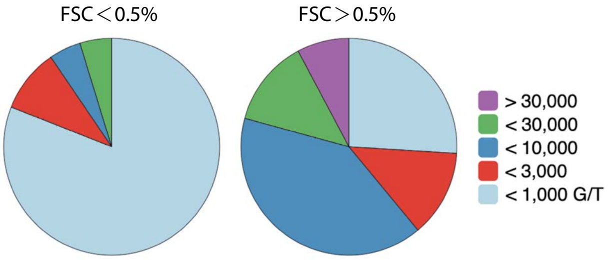

Fig. 8. Proportions of ships by gross tonnage (G/T) for FSC under and over 0.5%.

larger ships had FSCs larger than 0.50%. Thus, smaller ships tended to use low-sulfur fuels before the 2020 global limit.

There was a large reduction in the mean SO_2 concentration at the Kanmon Bridge from 6.3 to 0.9 ppb (Fig. 4), which corresponded to a reduction to 14% of the former concentration. The mean FSC changed from 1.40% to 0.23%, which corresponded to a reduction to 16% of the former concentration. The SO_2 concentration at our measurement site was mostly influenced by ships. Sakurai *et al.* (2021) estimated that the ship emissions of SO_2 would be reduced to 24% of that before the 2020 global limit. That is, the reduction ratios in the FSC and ambient SO_2 concentration were greater than that in the ship emissions of SO_2. The estimates by Sakurai *et al.* (2021) assumed that high-sulfur fuels with an FSC of 2.45% would be replaced with low-sulfur fuels with an FSC of 0.50%. Commercially available low-sulfur fuels contain 0.17% to 0.46% sulfur, which is lower than the 2020 global limit. Our results suggest that the ship emissions of SO_2 after the 2020 global limit was introduced may be lower than the estimates by Sakurai *et al.* (2021). That is, the 2020 global limit led to greater reductions in SO_2 emissions from ships than expected.

4. CONCLUSIONS

Ambient concentrations of SO_2 and CO_2 were measured under the Kanmon Bridge for several weeks in 2019 and 2020, before and after the 2020 global sulfur limit was introduced. The sniffing method detected ship plumes and monitored compliance with the FSC well. The FSC estimated generally varied from 0.50% to 3.00% in 2019, but the range narrowed to 0.10% to 0.40% in 2020. The mean FSC in 2019 was reduced in 2020 to 16% of that before the limit, which was consistent with a reduction ratio in the ambient SO_2 concentration. Sakurai *et al.* (2021) may have overestimated SO_2 emissions from ships after the 2020 global limit because they assumed a higher FSC of 0.50% than our measured results.

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