Response to reviewers' comments

We thank the reviewers for the constructive comments and suggestions, which are very positive to improve scientific contents of the manuscript. We have revised the manuscript appropriately and addressed all the reviewers' comments point-by-point for consideration as below. The remarks from the reviewers are shown in black, and our responses are shown in blue color. All the page and line numbers mentioned following are refer to the revised manuscript without change tracked.

The authors present a long time series of SO_2 observations using active DOAS instruments at two measurement sites in Shanghai. The first measurement site is located at a river, while the second one is an urban background site. During the observation period, the SO_2 emissions of ships were restricted twice and the changes on the ambient SO_2 levels as a result of these changes were evaluated and interpreted. In order to interpret the measurements, two machine learning models were used to first interpolate data gaps and then to eliminate the influence of different weather conditions on the measured SO_2 levels. The manuscript is generally well written and of high interest for scientists and policymakers, but I would suggest some improvements before publication in ACP.

General comments:

1. I would highly recommend adding some more explicit information how ship traffic changed and evolved at the measurement site during the years, e.g. average number of ship passages per year and the composition of ship types throughout the years. Changes in ship traffic density or fleet composition are often mentioned and used for interpretation of results, but never explicitly shown to the reader. Figure 8 somewhat reflects this, but only for ships where the plumes were captured with the DOAS instrument.

Response: Thank you for your comment. We have added information on ship traffic at the WSW site (2018–2023). Over a thousand vessels pass daily, including cargo, passenger, fishing, and tanker ships. Figures S1, S2 and Text S1 summarize daily vessel numbers, moving vs. stationary ships, vessel type composition, main and auxiliary engine power, and typical speeds. Seasonal and long-term trends, as well as the presence of larger, higher-powered vessels.

In the manuscript:

"where over a thousand vessels pass daily, including cargo ships, passenger ships, fishing boats, oil tanker and other ships in various operating conditions. Shipping activities are the primary source of ambient pollution at this site. Fig S1, S2 and Text S1 give an overview of ship activity in the WSW Channel." Please refer to Line 81-89.

In the Supporting document:

Text S1. Overview of Ship Activity in the WSW Channel.

"To provide background information on local ship traffic conditions relevant to the observed SO_2 variations, this section summarizes key characteristics of vessel activity in the WSW channel based on AIS data from 2018 to 2023.

Figure S1 presents the temporal evolution of daily vessel numbers in the channel, including total ships, moving ships, and stationary ships. Seasonal reductions in traffic are evident around the time of the Chinese New Year each year, reflecting holiday-related slowdowns. Throughout the period, the overall number of ship traffic shows a gradual increasing trend. The vessel type composition is *also illustrated, showing that cargo ships and passenger boats have remained the predominant categories.*

Figure S2 shows daily statistics of the main engine (ME) and auxiliary engine (AE) power of vessels passing through the channel. The ME power is generally much higher than AE power, reflecting the dominant role of propulsion engines in energy consumption and emissions. The large standard deviations in both ME and AE power reflect the diversity of ship types in the WSW channel—ranging from large cargo ships and cruise vessels (with ME power up to 50,000–70,000 kW) to small fishing and harbor boats (tens of kW). In recent years, the upper percentiles of both ME and AE power have increased, suggesting a growing presence of larger or higher-powered vessels in the area.

Vessel speed is another relevant operational parameter. Although instantaneous speed can vary significantly within a single ship's trajectory, it is observed that the maximum speed of vessels operating in this region can reach up to 52.6 knots. At the same time, many ships remain stationary near the shore or move slowly within the channel, typically maintaining speeds around 5–6 knots."

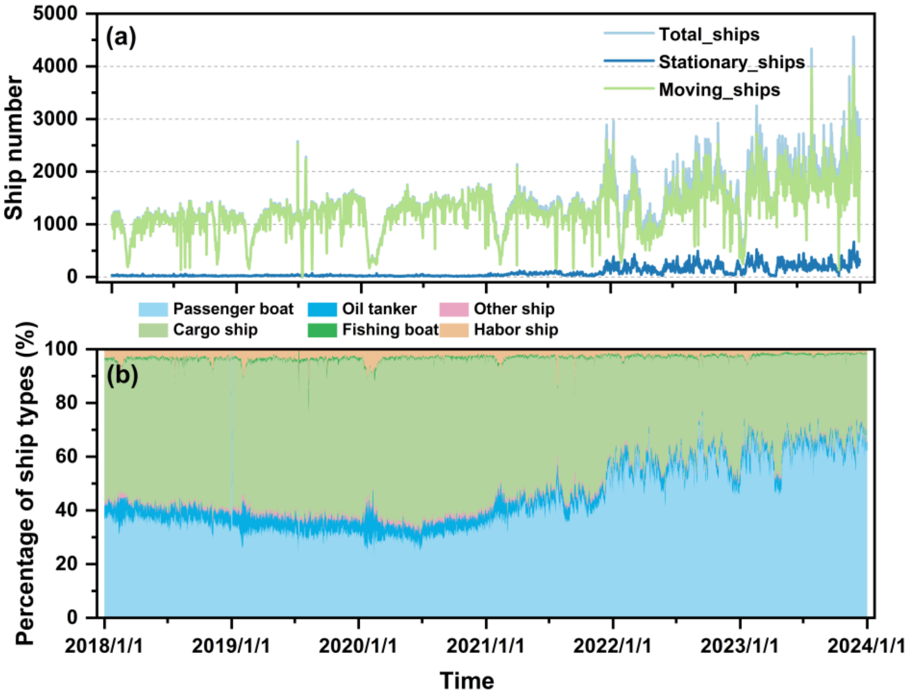

Figure S1. Temporal dynamics of daily ship traffic and ship type composition in the WSW channel (2018–2023). (a) Daily number of total ships, moving ships, and stationary ships detected from AIS records. (b) Percentage composition of different ship types over time, including passenger boats, cargo ships, oil tankers, shipping boats, harbor ships, and other vessels.

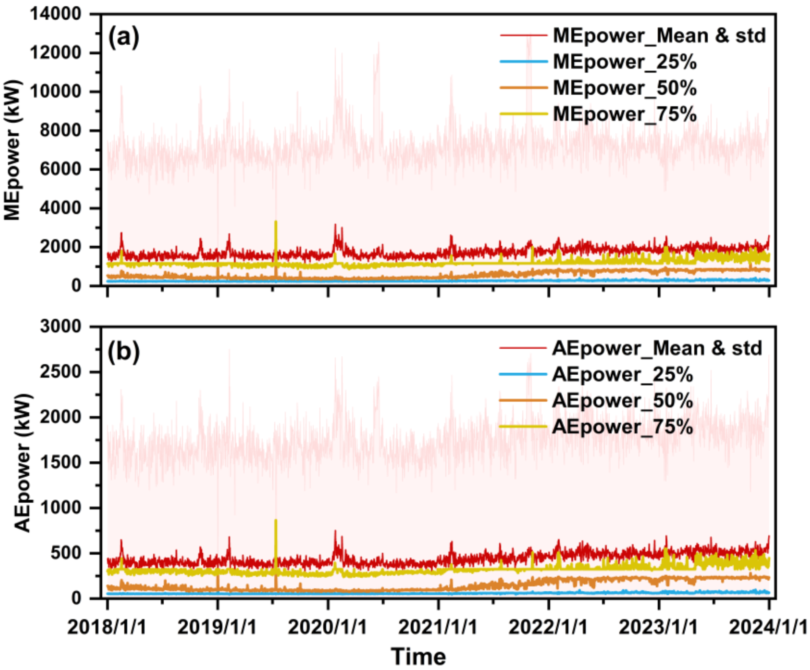

Figure S2. Temporal statistics of main engine and auxiliary engine power of vessels in the WSW channel (2018–2023). (a) Time series of main engine (ME) power, showing the mean ± standard deviation (shaded area) and the 25th, 50th, and 75th percentiles of power (kW). (b) Time series of auxiliary engine (AE) power, showing the mean ± standard deviation (shaded area) and the 25th, 50th, and 75th percentiles of power (kW).

2. I would suggest adding Figure S6 of the Supplement to Figure 6 because it's an important piece of information.

Response: Thank you for your comment. We already adding Figure S6 of the Supplement to Figure 6. In the revised manuscript, you can see the trend of the concentration distribution of SO_2 ranging from 2 ppbv to 30 ppbv. The corresponding description has also been modified.

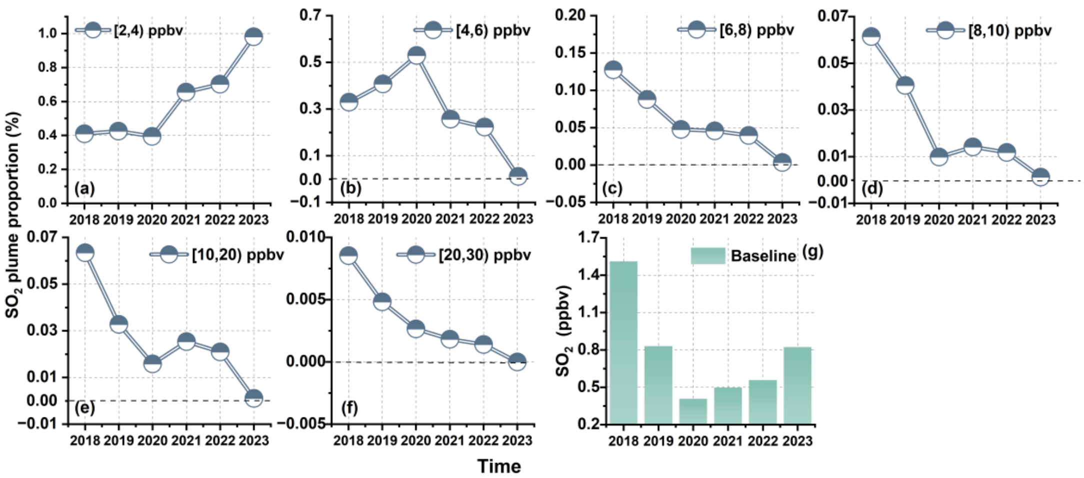

Figure 6: *Yearly variation in SO_2 plume proportions and baseline level from 2018 to 2023. (a-f) Number of SO_2-rich plumes within different concentration ranges divided by the total valid spectra for each year. (g) Annual baseline concentrations of SO_2 obtained through the BEADs algorithm. Please refer to Line 307-310.*

3. How are ship emissions treated in the machine learning gap-filling algorithm? Does the gap-filling only reproduce the baseline SO_2 signal from other sources than ships? Can you provide a comparison of the result of the gap-filling algorithm with measured data?

Response: Thank you for your comment. We address your three sub-questions as follows. As the first two questions are closely related, we discuss them together.

Question 1: How are ship emissions treated in the machine learning gap-filling algorithm? In designing the machine learning (ML)-based gap-filling algorithm, we considered it essential to include parameters that could reflect ship emissions. Initially, we attempted to use two types of input variables: (a) the number of vessels derived from AIS data, and (b) an hourly ship emission inventory based on bottom-up estimates within a 4 km radius around the WSW (LP-DOAS) site.

However, we found that at the hourly scale, neither of these indicators showed meaningful correlation with Observed_SO_2 concentrations in the shipping channel. Changing these variables had negligible impact on the ML model outputs, suggesting that they could not effectively represent hourly variations in ship emissions. We believe this is due to the coarse nature of AIS-based indicators: ship numbers do not capture ship type, size, or operational status.

Likewise, raw bottom-up emission inventories are spatially aggregated and cannot be readily matched to the high temporal resolution of hourly LP-DOAS measurements. Although it is indeed meaningful to relate emission inventories obtained over a certain area to concentrations measured along a single LP-DOAS path, establishing such a correspondence at an hourly scale is highly challenging and beyond the scope of this study (Although it is difficult to establish a correspondence at an hourly scale, we find that at coarser temporal resolutions, emission inventory data can be used to validate the Ship_related_SO_2 identified in this study in terms of overall trends. As shown in Text S5.)

Therefore, in a second round of modeling, we took advantage of the fact that the observed pollutants at WSW—including SO_2, NO_2, HONO, HCHO, and O_3—are themselves strongly influenced by ship emissions. These co-measured pollutants were used as predictors to reconstruct missing SO_2 values via cross-species learning within the ML framework. That is, when SO_2 data were missing, its temporal patterns were inferred from other concurrent trace gases. This allows the model to retain the signal of ship emissions implicitly present in the co-measured species. As shown in Figure 2.

Question 2: Does the gap-filling only reproduce the baseline SO_2 signal from other sources than ships? Regarding the second question specifically: no, the gap-filling model does not only reproduce a “baseline” SO_2 signal excluding ship emissions. Instead, the reconstructed SO_2 values at WSW reflect the combined influence of three major sources: (a) Direct ship emissions, which are captured via learned associations with co-pollutants (such as NO_2 and HONO), as previously addressed in the response to the first question; (b) land-based sources from urban areas, which also known as Deweahthered_FDU and (c) Meteorological influences, which are incorporated using feature representations derived from ERA5 reanalysis data.

The model was trained using feature vectors representative of all these sources, ensuring that the gap-filled SO_2 values capture the variability of emissions, including those originating from ships.

Question 3: Can you provide a comparison of the result of the gap-filling algorithm with measured data? Of course, now we are very pleased to present to you the comparison between the gap-filling algorithm and the actual observed values. To evaluate the performance of the machine learning gap-filling algorithm, we conducted a point-to-point comparison between predicted and observed SO_2 concentrations using a dataset from 2024, comprising 641 valid hourly measurements. As shown in Figure S6. This segment was selected as a new representative test case, given the lack of long continuous observations during earlier periods. The data were not arbitrarily selected or artificially stitched together; rather, they were drawn from the naturally continuous measurement windows available in January, February and March of 2024. Although data is still incomplete, the period we selected represents the longest and most continuous segment of real observations available.

Figure S6 demonstrate strong consistency between the predicted and observed data, with an R^2 of 0.84, RMSE of 0.41 ppbv, and MAE of 0.29 ppbv. The overall mean SO_2 concentration was 1.42 ppbv from observations and 1.38 ppbv from model predictions, indicating minimal systematic bias. Across different concentration ranges, the model reproduced observed values accurately: for example, in the 1–3 ppbv range, both predicted and observed means were nearly identical (1.74 vs. 1.75 ppbv), and even for higher values (3–5 ppbv), the agreement remained robust (3.66 vs. 3.88 ppbv). We further examined the model's ability to reproduce short-term SO_2 episodes, which are of particular importance for ship plume characterization. Among the data, 1.25% of points exceeded 5 ppbv. For this subset, the predicted mean was 4.71 ppbv, compared to an observed mean of ~5.45 ppbv. The predicted maximum SO_2 also closely approached the observed maximum (5.94 vs. 6.08 ppbv). Although the reproduction effect of high concentrations is slightly lower than that of low concentrations (this is usually due to the relatively lower occurrence frequency of high concentrations, resulting in fewer opportunities to provide learning samples), in general, the model can well reproduce the changes in SO_2 concentration in the waterway environment. These results indicate that the model is capable of recovering both baseline concentrations and elevated episodes associated with local sources such as ship emissions. Importantly, although the algorithm does not rely on explicit ship indicators (e.g., AIS or emission inventories), it incorporates co-measured species (NO_2, HONO, HCHO, O_3) and meteorological factors that reflect shared influences from ship activity. This design enables the model to retain shipping-related signals in an implicit but effective way.

We have now added detailed explanations regarding the treatment of ship emissions in the methodology section of manuscript. At the same time, in the supplementary materials, we have added our considerations when selecting indicators to represent the emissions of the ship, and also included a new comparison diagram and textual explanation of the gaps-filling algorithm. These pieces of information are intended to help future readers better understand the role and performance of the gap-filling algorithm used in this study.

In the manuscript:

"As illustrated in Fig. 2a, the gap-filling model for WSW SO_2 incorporates several predictive features representing three major types of environmental influences: including meteorological conditions, ship emissions, and urban land-based emissions. Specifically, co-measured pollutants at WSW (NO_2, HCHO, HONO, O_3) help represent shipping-related emissions through cross-species learning, while SO_2 measured at FDU—after meteorological normalization (Deweathered_FDU)—accounts for urban land-based emission influences." *Please refer to Line 117-121.*

In the supplementary materials

Text S3. Machine learning data input, model tuning, and performance evaluation.

"When training the model to fill the missing SO_2 values at WSW, three categories of input features were incorporated to comprehensively capture environmental influences from different sources: meteorological conditions, ship emissions, and urban land-based emissions. Specifically, these consisted of: seven meteorological variables from the ERA5 reanalysis dataset; co-measured pollutant data (including HCHO, HONO, O_3, and NO_2) obtained via DOAS at the WSW site—which facilitated indirect capture of ship emission signals through cross-species learning; and meteorologically normalized SO_2 data from the FDU site (Deweathered_FDU), representing background variations associated with urban land-based emissions. The model achieved an R^2 of 0.76 and an RMSE of 0.65 ± 0.21. The completed SO_2 concentration time series is presented in Figure S3.

The selection of predictor variables to represent ship emissions involved multiple rounds of testing and evaluation. Initial attempts to incorporate AIS-derived indicators, such as ship number and hourly bottom-up emission inventories within a 4 km radius around the WSW site, showed no significant correlation with observed SO_2 concentrations at the hourly scale—their inclusion resulted in negligible improvement in model performance. This outcome is attributed to the fact that AIS-based ship number do not capture distinctions in ship type, size, or operational status. For raw bottom-up emission inventories, it's spatially aggregated and cannot be readily matched to the high temporal resolution of hourly LP-DOAS measurements. Consequently, the approach shifted toward using co-measured pollutants (NO_2, HCHO, HONO, O_3) obtained at the same WSW site, which are strongly influenced by ship activities.

To evaluate the performance of the machine learning-based gap-filling algorithm, a point-to-point comparison was conducted between predicted and observed SO_2 concentrations. The evaluation used an independent validation dataset from 2024, consisting of 641 hourly measurements obtained during naturally continuous observation windows in January, February, and March. As shown in Figure S6, the gap-filled SO_2 concentrations (Predicted SO_2) demonstrate strong agreement with observed SO_2. The model achieved an R^2 of 0.84, with an RMSE of 0.41 ppbv and MAE of 0.29 ppbv. The overall mean observed SO_2 concentration was 1.42 ppbv, compared to a predicted mean of 1.38 ppbv. The model accurately reproduced observed values across different concentration ranges: within the 1–3 ppbv interval, the predicted mean (1.75 ppbv) was nearly identical to the observed mean (1.74 ppbv), and for higher concentrations (3–5 ppbv), the predicted mean (3.88 ppbv) remained close to the observed value (3.66 ppbv). The model's ability to capture short-term SO_2 episodes—critical for characterizing ship plumes—was also evaluated. Among all data points, 1.25% exceeded 5 ppbv. For these high-concentration events, the predicted mean was 4.71 ppbv compared to an observed mean of 5.45 ppbv. The predicted maximum (5.94 ppbv) closely matched the observed maximum (6.08 ppbv). Although the reconstruction of peak concentrations shows a slight underestimation—likely due to the lower frequency of high -concentration events limiting training examples—the model overall captures the temporal variations in SO_2 concentrations well in the waterway environment."

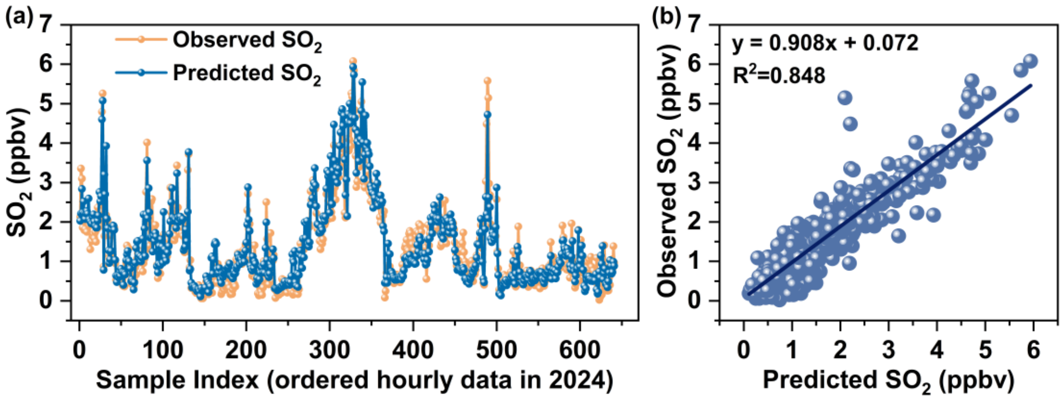

Figure S6. Comparison between observed and machine learning-predicted hourly SO_2 concentrations at WSW in 2024. (a) Temporal variation using ordered sample index. (b) Regression plot showing strong agreement (R^2 = 0.848) between predicted and observed values.

4. Also, in the supplement it looks like, there were almost no measurements at WSW in 2020 and from July 2022 to July 2023, how does this influence the results?

Response: Thank you for your comment. We acknowledge the limited measurement coverage at the WSW site in 2020 and between July 2022 and July 2023, which may raise concerns about the reliability of model-filled values and their influence on trend analysis. However, this limitation is unlikely to affect the overall conclusions. Our gap-filling model has been validated to reproduce long-term variations in channel SO_2 concentrations reliably. Based on an independent 2024 validation dataset, the entire bias observed was about –0.04 ppbv (Predicted_SO_2 minus Observed SO_2) Although this estimate may be slightly large due to the relatively short validation period, a longer-term comparison (2018–2023) between predicted and observed SO_2 shows extremely small residuals at WSW (-0.0032 ppbv; see Text S3 and Figures S4, S5). This indicates that over broader temporal scales, the gap-filling values are very close to the real measurements. Even if SO_2 levels in 2020–2022 were uniformly adjusted upward by 0.04 ppbv, the key findings—namely, a decrease from 2018 to 2020 followed by an increase from 2020 to 2023—would remain unchanged. Furthermore, the gap-filling data were only used in Section 3.1 for long-term trends; the analyses in Sections 3.2 and 3.3 relied solely on observed data and are therefore unaffected.

While the missing periods may have reduced the number of high- SO_2 plumes captured at WSW in 2020–2022, this effect is expected to be limited. The analyses in Sections 3.2 and 3.3 are based on the relative contribution of high-concentration plumes rather than the absolute number of plumes, which helps mitigate the influence of incomplete sampling.

To address this issue, we compared the differences between the predicted and observed values during 2018–2023. We also conducted a limitation analysis of the article in the supplementary material and main text.

In the supplementary materials:

Text S3. Machine learning data input, model tuning, and performance evaluation.

"Figure S4 presents the residual error plots and their frequency distribution between the predicted and observed SO_2 concentrations for both sites. Figure S5 shows the scatter plots of the predicted versus observed SO_2, along with the correlation coefficients (R^2). The results demonstrate that the

mean residuals are negligible (-0.0032 ppbv at WSW and -1.16×10⁻⁵ ppbv at FDU). The majority of daily residuals (59.36% at WSW and 86.9% at FDU) fall within ±0.2 ppbv, and the high R^2 values (above 0.9) confirm a strong model-observation agreement at both locations"

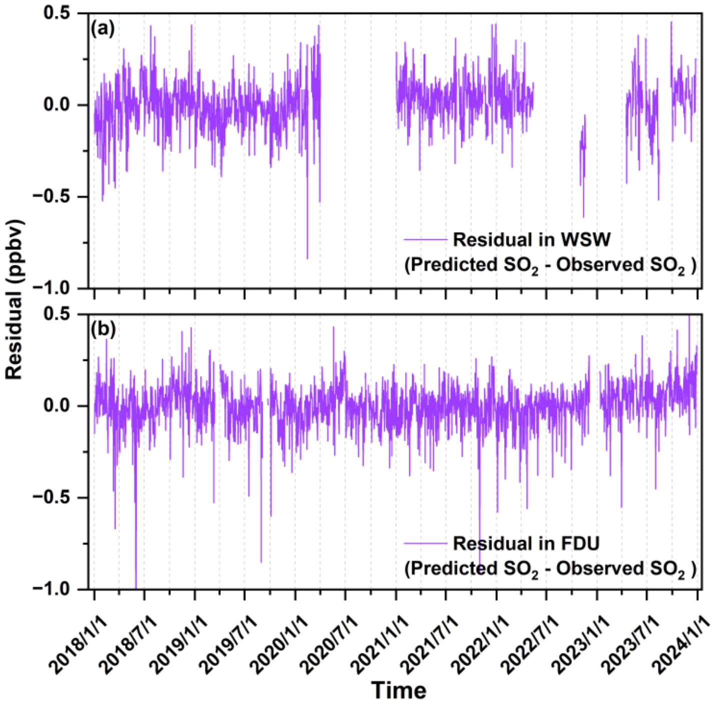

Figure S4. Time series and frequency distribution of residuals (Predicted SO_2 minus Observed SO_2) at the daily mean scale for (a, c) WSW and (b, d) FDU during 2018–2023.

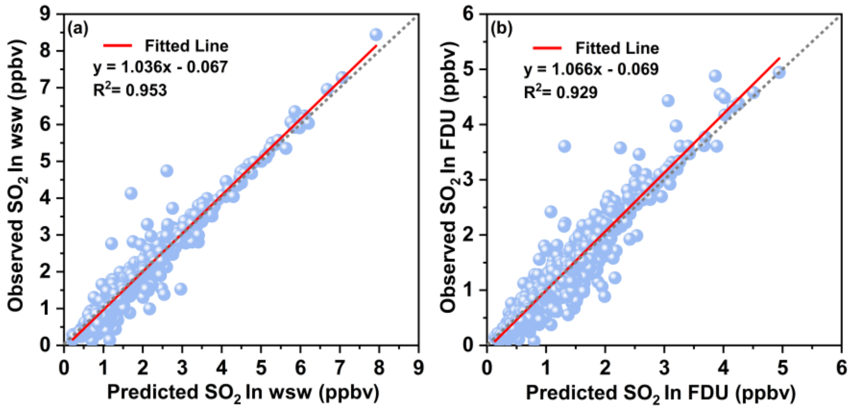

Figure S5. Scatter plots between predicted and observed SO_2 concentrations at the daily mean scale for (a) WSW and (b) FDU.

Text S7. Limitations and Uncertainties

"From a measurement coverage perspective, another source of uncertainty arises from the limited measurements at the WSW site in 2020 and between July 2022 and July 2023, during which reconstructed values were used to fill missing periods. Our validation analysis shows that the gap-filling model reproduces long-term SO_2 variations reliably, with a mean residual of –0.0032 ppbv over 2018–2023 (see Text S3, Figures S4, S5), although short validation samples (e.g., in 2024) suggest that biases of up to –0.04 ppbv may occasionally occur. Even if the concentrations during

2020–2022 were uniformly adjusted by this margin, the main interannual trends—a decrease from 2018 to 2020 followed by an increase from 2020 to 2023—would remain unchanged. We note, however, that the absence of measurements may reduce the number of high-SO_2 plumes captured during these years. Because our plume-related analyses in Sections 3.2 and 3.3 are based on relative contributions rather than absolute plume counts, this influence is expected to be limited, but some degree of bias cannot be fully excluded.”

5. What is the main wind direction at FDU and WSW? Even though FDU is a background station I would assume ship traffic could influence the SO_2 signal at this station, when the wind blows somewhat from the direction of the river.

Response: Thank you for your comment. The WSW and FDU stations are only about 4 km apart and both fall within the same ERA5 grid cell. Therefore, they are subject to broadly the same prevailing wind patterns, which are predominantly from the northeast (NE) and southeast (SE) sectors throughout 2018–2023. These directions are aligned with the Yangtze River channel, where ship traffic is concentrated, meaning that in principle both stations can be affected by ship emissions under such wind conditions.

However, we would like to clarify that in our methodology, we did not directly use the raw observed SO_2 at FDU as the background signal. Instead, we applied meteorological normalization (Deweathered model) to the FDU data, using machine learning to model and remove the effects of meteorology (including wind direction and speed, temperature, boundary layer height, etc.) on pollutant levels. This process effectively captures and accounts for episodes where ship-related air masses might lead to elevated concentrations due to directional transport. By removing these meteorologically driven variations, the residual signal at FDU reflects the underlying background pollution trend, excluding short-term transport effects such as those from the river channel. Therefore, in our study, the Deweathered SO_2 concentration at FDU (Deweathered_FDU) is used as the background station level, not the direct Observed_FDU. This ensures that our background estimation is robust against meteorological and directional influences, including potential ship traffic impacts.

We have added a wind rose figure (Figure S8) in the Supplement to illustrate the prevailing wind directions in WSW and FDU, and we have refined the description of Deweathered_FDU in the manuscript to clarify that while southeast winds can theoretically transport ship emissions to FDU, the deweathering procedure minimizes such influences, ensuring that FDU represents the urban background.

In the manuscript:

“For the FDU site, however, the Deweathered model effectively removes the influence of transported pollution under different wind directions (Fig. S8)—for example, ship-related SO_2 transported from the northeast channel—so that the residual values can represent the locally generated SO_2 level. Given that both FDU and WSW are located in similar environments, primarily surrounded by residential areas and typical urban roads, the Deweathered_ SO_2 concentrations at FDU are therefore taken as the background level for Shanghai’s urban region. Thus, by subtracting the background (Deweathered_FDU) from the Deweathered_WSW, the contribution of

ship_related_SO₂ can be effectively determined." Please refer to Line 145-150.

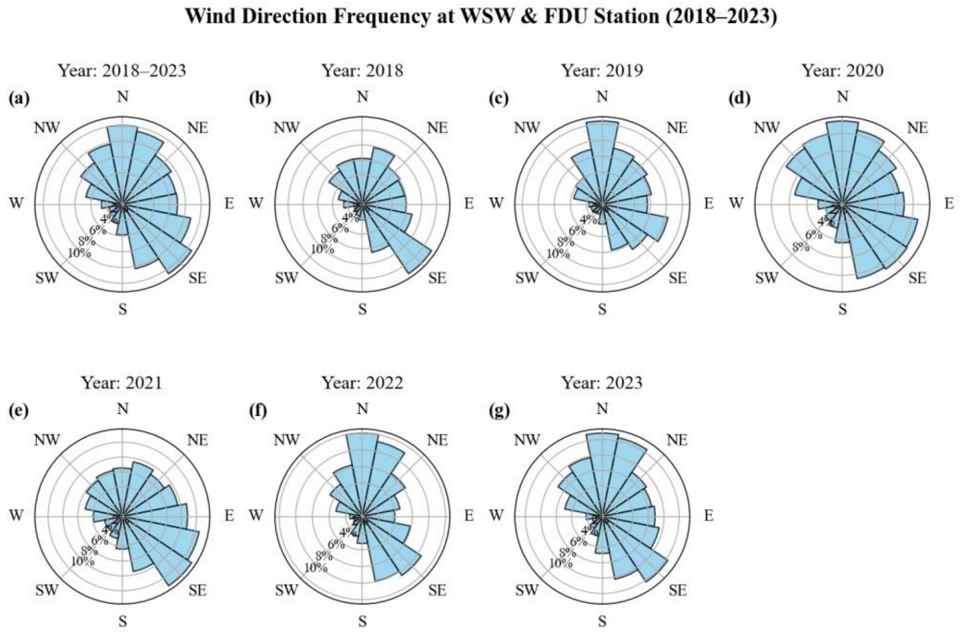

Figure S8. Wind direction frequency distribution at WSW and FDU station from 2018 to 2023. (a) The aggregated wind distribution for all years. (b)–(g) The show annual wind patterns from 2018 to 2023. Wind direction is plotted in polar coordinates with percentage frequency indicated by concentric circles.

6. Could you elaborate a little bit on what measures the ships can use to reduce SO_2 emissions in this control area (e.g., change of fuel to lower sulphur fuels, scrubbers, …)

Response: Thank you for your comment. In this emission control area, ships can adopt several technical and operational measures to reduce SO_2 emissions, in line with both international and domestic regulations, including Switch to low-sulfur fuels, use of exhaust gas cleaning systems (scrubbers), use of alternative fuels and Operational measures.

One of the most common approaches is switching to low-sulfur fuels, such as marine gas oil (MGO) (Corbett et al., 2008), very low sulfur fuel oil (VLSFO) (Sultanbekov et al., 2022), or ultra-low sulfur fuel oil (ULSFO) (Ershov et al., 2022). Using these fuels can directly and effectively reduce the emission of SO_2.

Another widely used method is the installation of exhaust gas cleaning systems, known as scrubbers, which can effectively remove SO_2 from exhaust gases, allowing continued use of high-sulfur fuels while still complying with emission standards (Lunde Hermansson et al., 2024; Andreasen and Mayer, 2007). However, it is worth noting that the promotion of scrubbers has been limited due to environmental concerns associated with their use, including potential impacts such as slowed growth and increased mortality of marine organisms (Koski et al., 2017; Thor et al., 2021), as well as the acidification of surrounding waters (Hassellöv et al., 2013; Claremar et al., 2017).

In addition, the use of alternative fuels such as liquefied natural gas (LNG) (Pavlenko et al., 2020; Attah and Bucknall, 2015), methanol(Svanberg et al., 2018; Shi et al., 2023), or biofuels(Cesilla De

Souza and Eugênio Abel Seabra, 2024; Ahmed et al., 2025) has also emerged as a cleaner option, with LNG being particularly effective in reducing sulfur oxide emissions. However, the adoption of such fuels remains limited due to infrastructure and economic constraints.

Operational strategies such as speed reduction (slow steaming), route optimization, and the use of shore power while berthed can also significantly reduce fuel consumption and thus SO_2 emissions, especially in coastal and port areas. These measures are often implemented in combination, depending on ship characteristics, route planning, and regulatory requirements (Zis et al., 2015; Zis et al., 2014).

We introduce several methods to reduce SO_2 emissions from ships in the manuscript, and discuss the limitations of these approaches in Section 3.2

In the manuscript:

"In 2015, China launched its Domestic Emission Control Area (DECA 1.0) policy, requiring ships with compatible facilities in the Pearl River Delta, Yangtze River Delta, and Bohai Rim (Beijing-Tianjin-Hebei) regions to use fuel with ≤0.5% sulfur content during berthing periods from January 2016 (Zou et al., 2020; Zhang et al., 2019; Wang et al., 2021). By late 2018, China upgraded the policy to DECA 2.0, mandating that all ships operating within China's territorial sea (12-nautical-mile zone) must use fuel with ≤0.5% sulfur content while sailing from January 2019 onward, and ≤0.1% sulfur content while at berth, or adopt equivalent emission control measures. For example, installing exhaust gas cleaning systems (scrubbers) (Lunde Hermansson et al., 2024; Andreasen and Mayer, 2007), adopting alternative fuels like LNG(Pavlenko et al., 2020; Attah and Bucknall, 2015), methanol(Svanberg et al., 2018; Shi et al., 2023) and biofuels(Cesilla De Souza and Eugênio Abel Seabra, 2024; Ahmed et al., 2025), and applying operational strategies such as slow steaming and shore power use(Zis et al., 2015; Zis et al., 2014)." Please refer to Line 46-56.

"Some ships may have started using fuels with slightly lower sulfur content, which led to an increase in the frequency of low SO_2 plumes. The adoption of low-sulfur fuels was the most common choice during this period, as it required little or no modification of existing engine systems (Vedachalam et al., 2022; Slaughter et al., 2020). In contrast, due to the high retrofitting costs of engine systems and the limited number of ships using LNG, most ports currently do not provide bunkering facilities for LNG and other alternative fuels, including biofuels (Vedachalam et al., 2022). Although scrubbers allowed the continued use of high-sulfur fuels, their application was constrained by high installation costs, long retrofitting times (up to 9 months) (Slaughter et al., 2020), and concerns about secondary environmental impacts from waste discharges (Hassellöv et al., 2013; Claremar et al., 2017; Thor et al., 2021). Only 3,000/60,000 vessels have been retrofitted with a scrubber system, as reported by Slaughter et al. (2020)." Please refer to Line 276-285.

Specific comments:

1. L159: If these differences are caused by irregular ship traffic, this should be assessable in the AIS data and should be shown (as already mentioned in general comment 1)

Response: Thank you for your comment. We have added explicit information on ship traffic at the WSW site (2018–2023). As mentioned in our response to General Comment # 1, the newly added Figures S1–S2 and Text S1 summarize detailed AIS-based statistics, including daily vessel counts (typically over a thousand per day), the proportions of moving vs. stationary ships, vessel-type composition (cargo, passenger, fishing, and tanker ships), main and auxiliary engine power, typical speeds, as well as seasonal and long-term trends and the occurrence of larger, higher-powered vessels.

In addition, rather than directly using the raw AIS data, we compared the AIS-based bottom-up emission inventory with the Ship_related_SO_2 derived in this study (which reflects the SO_2 concentration variations attributable to irregular ship activities). The results show that Ship_related_SO_2 exhibits a stronger correlation with the inventory than either the directly observed_SO_2 or the Deweathered_SO_2, further confirming the validity of our approach. Furthermore, the conclusions drawn from our study may provide valuable insights for refining future ship emission inventories. Our approach, reason and result are detailed in the supplementary materials: (Text S5, S6; Figs. S15, S16), where we describe how AIS data were processed, integrated, and converted into emission inventory data to explain temporal variations in Ship_related_SO_2. Below, we clarify our methodology in two key aspects:

Firstly, why we did not use raw AIS data such as ship numbers? while the WSW channel experiences high vessel traffic (1,000–5,000 ships per day), raw ship counts alone are an inadequate proxy for SO_2 emissions. This is because vessels vary considerably in operational status (e.g., moving vs. stationary, high vs. low speed), size, and proximity to the measurement path. For example, two ships passing through the channel may both be counted as "1" in AIS statistics, yet their actual SO_2 emissions could differ by orders of magnitude due to differences in operational conditions and types.

Secondly, why we used an emission inventory? This inventory integrates multiple ship parameters—including position, speed, type, and main and auxiliary engine power—to estimate hourly SO_2 emissions. As demonstrated in supplementary materials Figure S15, this method yields significantly stronger correlations with ship_related_SO_2 (R^2 = 0.32–0.54) than raw SO_2 concentrations (R^2 = 0.04–0.06). Supplementary materials Figure S16 further shows synchronized temporal trends between the inventory estimates and observed Ship_related_SO_2, validating the effectiveness of this approach.

It is also worth noting that the development of ship emission inventories from AIS data remains an active and complex research field. While methodological refinements are beyond the scope of this study, we adopted a well-established inventory methodology (detailed in the Text S6) to ensure a meaningful and practical comparison with our observed results. We have also added clarifications in the manuscript and updated the supplementary material (Text S5, S6) to explain our AIS data processing methodology and justify the use of the emission inventory as the most representative dataset for shipping activity.

In the manuscript:

"In addition, a ship emission inventory based on AIS data was constructed, which further supports the interpretation of the variability observed at WSW (Text S5)." Please refer to Line 205-206.

In supplementary materials:

Text S5. Comparison Between Observational Data and AIS-Based Ship Emission Inventory.

"In the paragraph of this supplementary material, we compared Ship_related_SO_2 derived from DOAS observations with those estimated by traditional bottom-up ship emission inventories,

discussed the similarities and differences in outcome trends between the two approaches, and identified the underlying causes. AIS data provides detailed information on ship activities and is commonly used for calculating ship emission inventories on large spatiotemporal scales (Mao et al., 2020; Zou et al., 2020).

The reason for employing a comprehensive ship emission inventory from AIS, rather than relying on any single ship parameter (e.g., ship count, engine power, or speed), is as follows: While parameters like ship count, main engine power, and speed are valuable indicators, they are independently insufficient to accurately represent actual SO_2 emissions. This is because emissions are the product of a complex interplay of these factors. For instance: A high-powered ship moving slowly may emit similarly to a lower-powered ship at high speed; A stationary ship using its auxiliary engine for onboard services may emit more than a ship maneuvering at low speed with its main engine at idle; Simply counting all vessels equally ignores the vast differences in emission potential between a large container ship and a small fishing boat.

Therefore, a bottom-up emission inventory methodology was adopted (Text S6). This approach synthesizes the key parameters derived from AIS data—including ship type, instantaneous position and speed, and installed main and auxiliary engine power—into a holistic framework. By applying standardized emission algorithms and fuel sulfur content assumptions, this inventory translates dynamic ship activity into estimated hourly SO_2 emissions.

The scatter plots in Figure S15 illustrate the correlation (R^2) between ship emission inventory-based SO_2 emissions and the 14-day mean SO_2 concentrations based on observation at the WSW site. In the process of removing meteorological influences and land-based emissions, the correlation between the ship emission inventory and SO_2 concentrations progressively improves step by step. For the period from 2018 to 2020, the R^2 increases from 0.064 (Observed_SO_2) to 0.154 (Deweathered_SO_2), and further to 0.32 (Ship_related_SO_2). Similarly, for the period from 2021 to 2023, the R^2 rises from 0.043 (Observed_SO_2) to 0.163 (Deweathered_SO_2), and ultimately reaches 0.54 (Ship_related_SO_2). This trend underscores the effectiveness of the combined meteorological normalization and land-based emissions subtraction processes in refining our understanding of Ship_related_SO_2 contributions. Compared with directly observed_SO_2, the emissions inventory explains the trend of Ship_related_SO_2 changes better.

Figure S16 illustrates the 14-day mean variations of Ship_related_SO_2 concentrations and ship emission inventory in the WSW from 2018 to 2023. During the policy adjustment period (2018–2020), both the Ship_related_SO_2 and the corresponding SO_2 emissions in the inventory showed a gradual decline. If all ships had complied with the low-sulfur fuel policy, SO_2 emissions from ships would have shown a sharp decrease at the early stage of policy implementation, as illustrated in Figure S16c. However, due to the presence of non-compliant ships (as discussed in Sections 3.2 and 3.3), the reduction in SO_2 emissions from ships has been a gradual process, as shown in Figure S16a. While the consistency between Ship_related_SO_2 and the inventory improved during the policy stabilization period (2021–2023) in Figure S15f, which means that the fuel use of ships is closer to the policy requirements."

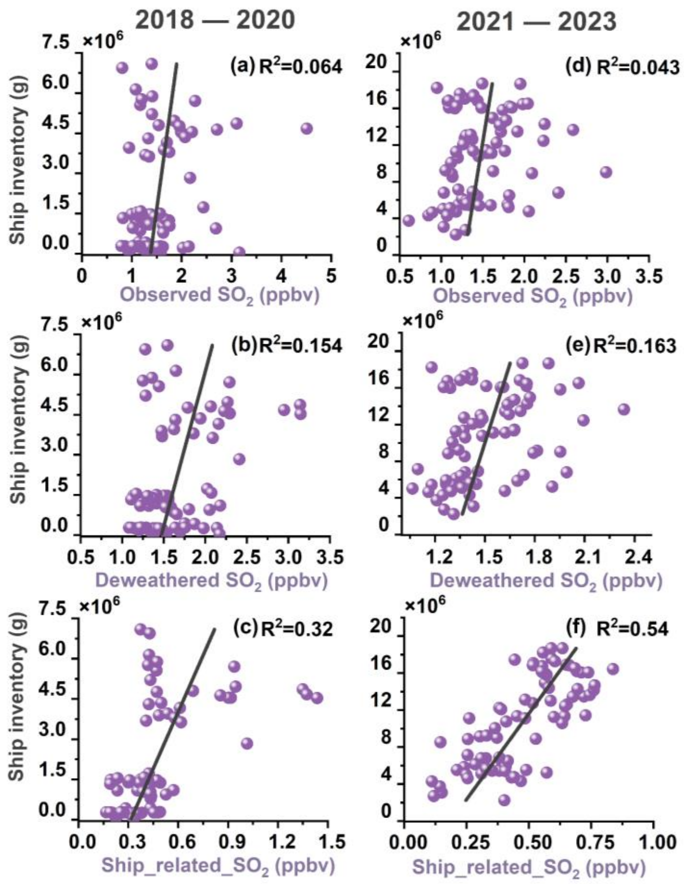

Figure S15. Correlations between 14-day mean SO_2 concentrations (x-axis) at WSW site and ship SO_2 inventory (y-axis), divided into three categories: (a, d) Observed_SO_2 concentrations, (b, e) Deweathered_SO_2 concentrations, and (c, f) Ship_related_SO_2 concentrations. (a–c) correspond to the policy adjustment period from 2018 to 2020, while panels (d–f) represent the policy stabilization period from 2021 to 2023.

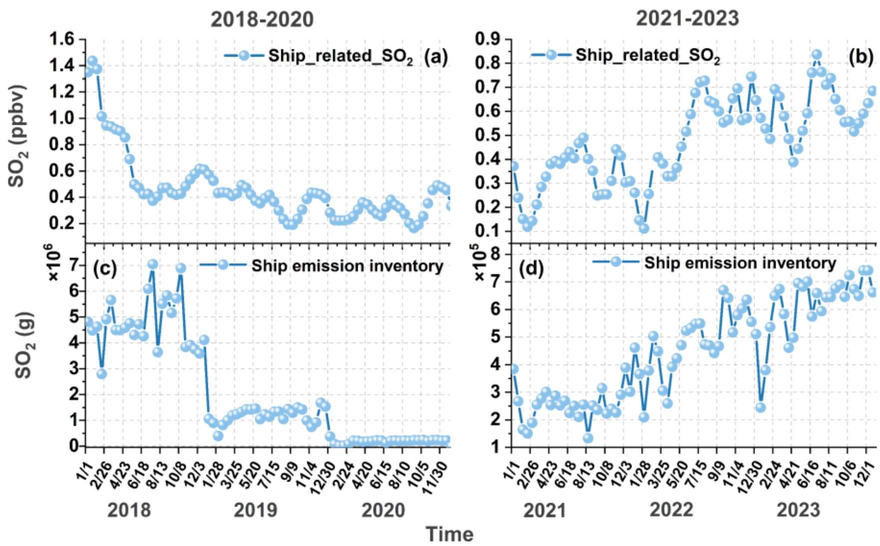

Figure S16. 14-day mean variations of Ship_related_SO_2 concentrations and emission inventory in the Wusong channel from 2018 to 2023. (a) and (b) represent the 14-day mean Ship_related_SO_2 derived from observations for 2018–2020 and 2021–2023, respectively. (c)

and (d) show the corresponding 14-day mean SO_2 emissions from the ship emission inventory during the same periods.

2. L173: Was there a strong reduction in ship traffic in 2020 due to COVID19 compared to the other years? Is this decrease in WSW data maybe influenced by the lack of observational data in 2020?

Response: Thank you for your comment. According to the newly added Figure S10, ship traffic in the 4-km radius around the WSW site showed a clear increasing trend from 2018 to 2023, with daily average ship number of 1037 ± 247, 1178 ± 312, 1223 ± 353, 1268 ± 363, 1507 ± 489, and 1939 ± 594, respectively. This supports our description of steadily growing shipping activity at WSW. Although the total number of vessels in 2020 remained higher than in 2018 and 2019, there was indeed a temporary reduction during the most severe COVID-19 lockdown period (January–April 2020), when the daily average dropped to 916 ± 406 vessels, lower than in the same months of 2018 (937 ± 264) and 2019 (1002 ± 309). Therefore, while the pandemic temporarily suppressed traffic, it did not reverse the long-term growth trend of shipping activity at WSW.

Regarding your second question on whether the observed decrease at WSW may be influenced by the lack of measurements in 2020: as also discussed in our response to General Comment #4, we acknowledge that the gap-filled values could slightly underestimate SO_2 concentrations. A short validation using an independent 2024 dataset suggested a possible bias of about –0.04 ppbv (Predicted minus Observed). However, a longer-term comparison over 2018–2023 showed an extremely small residual at WSW (–0.0032 ppbv; see Text S3 and Figures S4–S5), indicating that the model reproduces long-term variations reliably. Even if SO_2 concentrations in 2020 were adjusted upward by 0.04 ppbv, the key interannual trend—namely, a decrease from 2018 to 2020 followed by an increase from 2020 to 2023—would remain unchanged.

We have added a description of the changes in ship numbers from 2018 to 2023 in the main text, presented the temporal variations in ship numbers during this period in Figure S10 of the Supplement, and discussed the limitations and uncertainties arising from missing observations in Supplementary Text S7.

In the manuscript:

"Fig. S12 shows the overall increasing trend in the number of ships from 2018 to 2023, with irregular fluctuations within each year." Please refer to Line 253-254.

In supplementary materials:

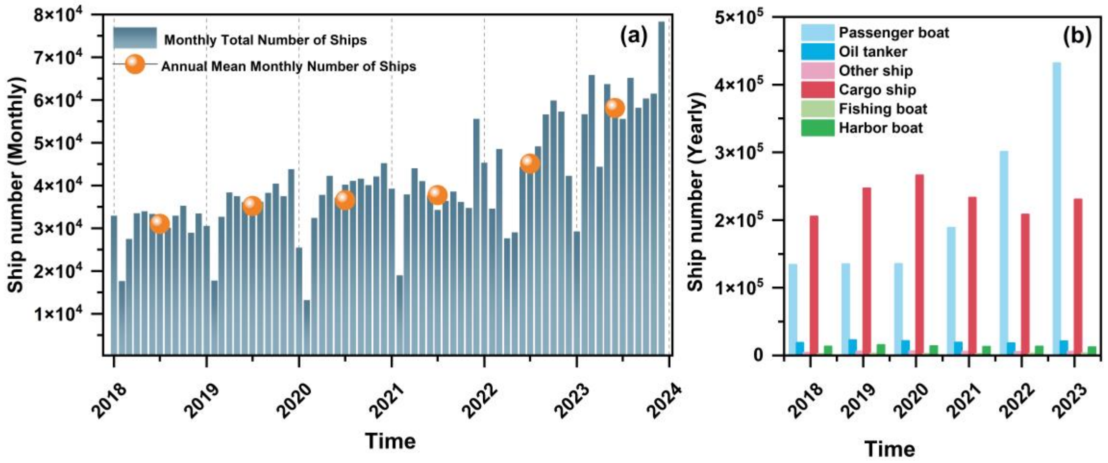

Figure S12. Annual variation of shipping activity in the channel from 2018 to 2023. (a) Monthly total number of ships and annual mean values. (b) Yearly ship number by ship type (cargo, oil tanker, passenger boat, fishing boat, and harbor boat). (For a more robust parameter of activity, a ship emission inventory (Text S7) was created, incorporating ship number, type, ME & AEpower, and speed for comparison with Ship_related_SO_2)

Text S7. Limitations and Uncertainties

"From a measurement coverage perspective, another source of uncertainty arises from the limited measurements at the WSW site in 2020 and between July 2022 and July 2023, during which reconstructed values were used to fill missing periods. Our validation analysis shows that the gap-filling model reproduces long-term SO_2 variations reliably, with a mean residual of –0.0032 ppbv over 2018–2023 (see Text S3, Figures S4–S5), although short validation samples (e.g., in 2024) suggest that biases of up to –0.04 ppbv may occasionally occur. Even if the concentrations during 2020–2022 were uniformly adjusted by this margin, the main interannual trends—a decrease from 2018 to 2020 followed by an increase from 2020 to 2023—would remain unchanged. We note, however, that the absence of measurements may reduce the number of high-SO_2 plumes captured during these years. Because our plume-related analyses in Sections 3.2 and 3.3 are based on relative contributions rather than absolute plume counts, this influence is expected to be limited, but some degree of bias cannot be fully excluded."

3. L196 to L199: FDU shows a decrease and stabilization at a lower level, while WSW shows a decrease and then increases again in 2022 and 2023. Please clarify. L196 ~ L199:

Response: Thank you for pointing this out. We have carefully re-examined the trend descriptions and revised the text accordingly to ensure consistency between the observed data and the written interpretation. Specifically, we have clarified that:

"After normalizing for meteorological influences, the deweathered SO_2 concentrations (Deweathered_WSW and Deweathered_FDU) represent a time series with meteorological variability removed. These deweathered values is overall higher than the observed concentrations. Deweathered_FDU shows a decreasing trend in 2022 followed by a stabilization in 2023, while Deweathered_WSW exhibits a decline since 2018 and an increase again in 2022 and 2023." Please refer to Line 222-225.

4. Add Figure S6 to Figure 6, because it is an important piece of information for your reasoning.

Response: Thank you for your comment. We already adding Figure S6 of the Supplement to Figure 6. In the revised manuscript, you can see the trend of the concentration distribution of SO_2 ranging from 2 ppbv to 30 ppbv. The corresponding description has also been modified.

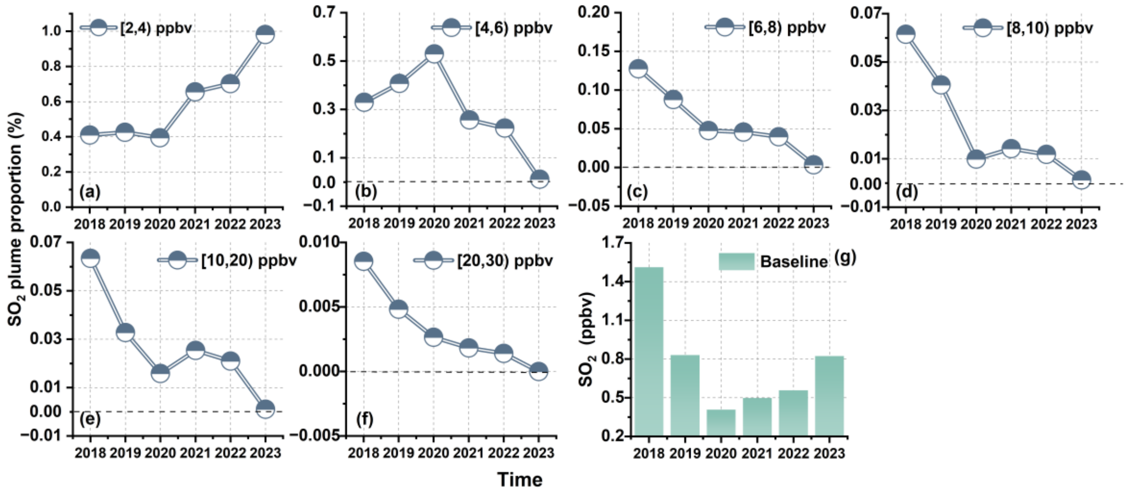

Figure 6: *Yearly variation in SO_2 plume proportions and baseline level from 2018 to 2023. (a-f) Number of SO_2-rich plumes within different concentration ranges divided by the total valid spectra for each year. (g) Annual baseline concentrations of SO_2 obtained through the BEADs algorithm. Please refer to Line 307-310.*

Technical corrections:

1. L12: Zhou should be capitalized.

Response: Thank you for pointing this out. We have corrected the capitalization and now "Zhou" is properly capitalized. *Please refer to Line 12.*

2. L125: please add a reference for the ERA5 dataset.

Response: Thank you for your comment. We have revised the manuscript to include a description of the ERA5 dataset and added appropriate references. The revised sentence now reads:

"All meteorological data used in this study were obtained from the fifth-generation European Centre for Medium-Range Weather Forecasts (ECMWF) atmospheric reanalysis, known as ERA5, which provides hourly around-the-clock meteorological factors from surface up to 0.01 hPa (spanning 137 vertical levels) with a spatial resolution of 0.25° × 0.25° (Marshall, 2000; Hersbach et al., 2020)."

Please refer to Line 164-169.

3. Figure 4: here CDECA is mentioned, but this is not mentioned or explained anywhere else, please clarify. Also, there is a typo in "low-sulfur fuel oil" right before "CDECA" in this Figure.

Response: Thank you for your comment. We have revised the Introduction to explicitly clarify the timeline and sulfur content limits of China emission control policies. The previously used abbreviation "CDECA" has been replaced by "DECA 2.0" for consistency. We also revised the description in Figure 4 about "low-sulfur fuel oil".

"In 2015, China launched its Domestic Emission Control Area (DECA 1.0) policy, requiring ships with compatible facilities in the Pearl River Delta, Yangtze River Delta, and Bohai Rim (Beijing-Tianjin-Hebei) regions to use fuel with ≤0.5% sulfur content during berthing periods from January 2016 (Zou et al., 2020; Zhang et al., 2019; Wang et al., 2021). By late 2018, China upgraded the policy to DECA 2.0, mandating that all ships operating within China's territorial sea (12-nautical-mile zone) must use fuel with ≤0.5% sulfur content while sailing from January 2019 onward, and ≤0.1% sulfur content while at berth, or adopt equivalent emission control measures." Please refer to Line 47-53.

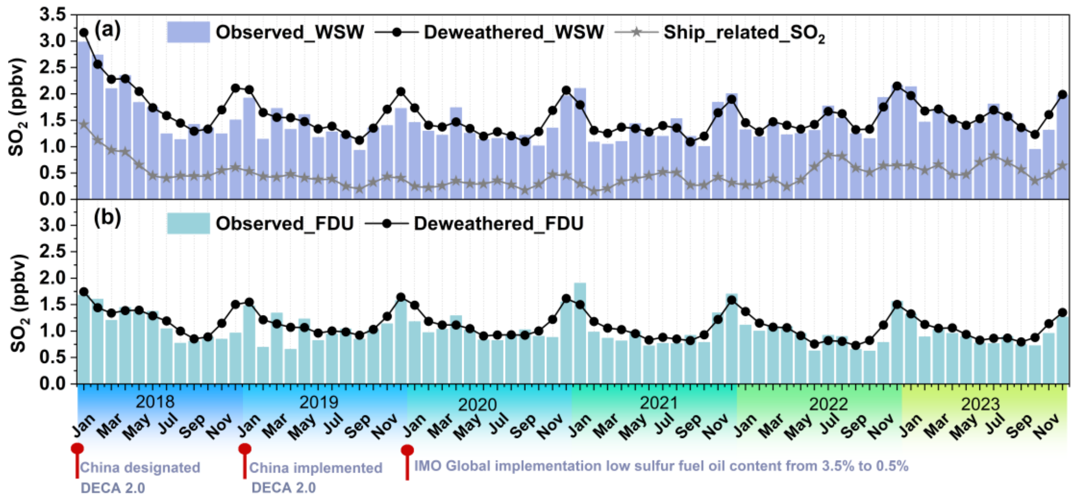

Figure 4: *Monthly Observed_SO_2 concentrations based on DOAS and Deweathered_SO_2 after weather normalization in WSW and FDU, and Ship_related_ SO_2 contributions during 2018-2023. (a) The light purple bars represent the monthly average Observed_SO_2 concentration at WSW; The solid black circles represent the deweathered SO_2 concentration at WSW after removing meteorological influences. The gray star symbols indicate the monthly average contribution of Ship_related_SO_2. (b) The light blue bars represent the monthly average observed SO_2 concentration at FDU; The solid black circles represent the Deweathered_SO_2 concentration at FDU removing meteorological influences. Please refer to Line 209-216.*

4. L250: Please verify 2023, I think it should be 2021.

Response: Thank you for pointing this out. We have carefully checked the sentence and confirmed that the correct year should indeed be 2021. This has now been corrected in the revised manuscript.

"The baseline was highest in 2018 and subsequently exhibited a declining trend from 2018 to 2021, followed by an increase from 2021 to 2023, consistent with the variation in Ship_related_SO_2 observed in Section 3.1." Please refer to Line 287-288.

5. Supplement:

"mLF/aEF: Main engine/auxiliary engine emission factor, g/kWh", I think mLF needs to be changed to mEF.

Response: Thank you for pointing this out. We have corrected the typo in the Supplement: "mLF" has been changed to "mEF" to accurately represent the main engine emission factor.

"mEF/aEF: Main engine/auxiliary engine emission factor, g/kWh;"

Reference:

Ahmed, S., Li, T., Zhou, X. Y., Yi, P., Chen, R. J. R., and Reviews, S. E.: Quantifying the environmental footprints of biofuels for sustainable passenger ship operations, Renewable and Sustainable Energy Reviews 207, 114919, https://doi.org/10.1016/j.rser.2024.114919, 2025.

Andreasen, A. and Mayer, S.: Use of Seawater Scrubbing for SO_2 Removal from Marine Engine Exhaust Gas, Energy & Fuels, 21, 3274-3279, http://10.1021/ef700359w, 2007.

Attah, E. E. and Bucknall, R. J. O. E.: An analysis of the energy efficiency of LNG ships powering options using the EEDI, Ocean Engineering, 110, 62-74, https://doi.org/10.1016/j.oceaneng.2015.09.040, 2015.

Cesilla de Souza, L. and Eugênio Abel Seabra, J.: Technical-economic and environmental assessment of marine biofuels produced in Brazil, Cleaner Environmental Systems, 13, 100195, https://doi.org/10.1016/j.cesys.2024.100195, 2024.

Claremar, B., Haglund, K., and Rutgersson, A.: Ship emissions and the use of current air cleaning technology: contributions to air pollution and acidification in the Baltic Sea, Earth Syst. Dynam., 8, 901-919, http://10.5194/esd-8-901-2017, 2017.

Corbett, J. J., Winebrake, J. J. J. J. o. t. A., and Association, W. M.: Emissions tradeoffs among alternative marine fuels: Total fuel cycle analysis of residual oil, marine gas oil, and marine diesel oil, J Air Waste Manag Assoc, 58, 538-542, http://doi.org/10.3155/1047-3289.58.4.538., 2008.

Ershov, M. A., Savelenko, V. D., Makhmudova, A. E., Rekhletskaya, E. S., Makhova, U. A., Kapustin, V. M., Mukhina, D. Y., and Abdellatief, T. M. M.: Technological Potential Analysis and Vacant Technology Forecasting in Properties and Composition of Low-Sulfur Marine Fuel Oil (VLSFO and ULSFO) Bunkered in Key World Ports, Journal of Marine Science and Engineering, 10, 1828, https://doi.org/10.1016/j.resourpol.2022.102636, 2022.

Hassellöv, I.-M., Turner, D. R., Lauer, A., and Corbett, J. J.: Shipping contributes to ocean acidification, Geophysical Research Letters, 40, 2731-2736, https://doi.org/10.1002/grl.50521, 2013.

Koski, M., Stedmon, C., and Trapp, S.: Ecological effects of scrubber water discharge on coastal plankton: Potential synergistic effects of contaminants reduce survival and feeding of the copepod Acartia tonsa, Marine Environmental Research, 129, 374-385, https://doi.org/10.1016/j.marenvres.2017.06.006, 2017.

Lunde Hermansson, A., Hassellöv, I.-M., Grönholm, T., Jalkanen, J.-P., Fridell, E., Parsmo, R., Hassellöv, J., and Ytreberg, E.: Strong economic incentives of ship scrubbers promoting pollution, Nature

Sustainability, 7, 812-822, http://doi.10.1038/s41893-024-01347-1, 2024.

Mao, J., Zhang, Y., Yu, F., Chen, J., Sun, J., Wang, S., Zou, Z., Zhou, J., Yu, Q., Ma, W., and Chen, L.: Simulating the impacts of ship emissions on coastal air quality: Importance of a high-resolution emission inventory relative to cruise- and land-based observations, Science of The Total Environment, 728, 138454, https://doi.org/10.1016/j.scitotenv.2020.138454, 2020.

Pavlenko, N., Comer, B., Zhou, Y., Clark, N., and Rutherford, D. J. S. E. P. A. S., Sweden: The climate implications of using LNG as a marine fuel, Swedish Environmental Protection Agency: Stockholm, 2020.

Shi, J., Zhu, Y., Feng, Y., Yang, J., and Xia, C. J. A.: A prompt decarbonization pathway for shipping: green hydrogen, ammonia, and methanol production and utilization in marine engines, Atmosphere, 14, 584, https://doi.org/10.3390/atmos14030584, 2023.

Slaughter, A., Ray, S., and Shattuck, T. J. D. D. L.: International Maritime Organization (IMO) 2020 strategies in a non-compliant world, 1-14, 2020.

Sultanbekov, R., Denisov, K., Zhurkevich, A., and Islamov, S.: Reduction of Sulphur in Marine Residual Fuels by Deasphalting to Produce VLSFO, 10, 1765, 2022.

Svanberg, M., Ellis, J., Lundgren, J., Landälv, I. J. R., and Reviews, S. E.: Renewable methanol as a fuel for the shipping industry, Renewable and Sustainable Energy Reviews, 94, 1217-1228, https://doi.org/10.1016/j.rser.2018.06.058, 2018.

Thor, P., Granberg, M. E., Winnes, H., and Magnusson, K.: Severe Toxic Effects on Pelagic Copepods from Maritime Exhaust Gas Scrubber Effluents, Environmental Science & Technology, 55, 5826-5835, http://doi.org/10.1021/acs.est.0c07805, 2021.

Vedachalam, S., Baquerizo, N., and Dalai, A. K.: Review on impacts of low sulfur regulations on marine fuels and compliance options, Fuel, 310, 122243, https://doi.org/10.1016/j.fuel.2021.122243, 2022.

Wang, X., Yi, W., Lv, Z., Deng, F., Zheng, S., Xu, H., Zhao, J., Liu, H., and He, K.: Ship emissions around China under gradually promoted control policies from 2016 to 2019, Atmos. Chem. Phys., 21, 13835-13853, http://doi.org/10.5194/acp-21-13835-2021, 2021.

Zhang, X., Zhang, Y., Liu, Y., Zhao, J., Zhou, Y., Wang, X., Yang, X., Zou, Z., Zhang, C., Fu, Q., Xu, J., Gao, W., Li, N., and Chen, J.: Changes in the SO_2 Level and $PM_{2.5}$ Components in Shanghai Driven by Implementing the Ship Emission Control Policy, Environmental Science & Technology, 53, 11580-11587, http://doi.org/10.1021/acs.est.9b03315, 2019.

Zis, T., North, R. J., Angeloudis, P., Ochieng, W. Y., and Bell, M. G. J. T. R. R.: Environmental balance of shipping emissions reduction strategies, Transportation Research Record Journal of the Transportation Research Board, 2479, 25-33, http://doi.org/10.3141/2479-04, 2015.

Zis, T., North, R. J., Angeloudis, P., Ochieng, W. Y., Harrison Bell, M. G. J. M. E., and Logistics: Evaluation of cold ironing and speed reduction policies to reduce ship emissions near and at ports, Maritime Economics & Logistics, 16, 371-398, http://doi.org/10.1057/mel.2014.6, 2014.

Zou, Z., Zhao, J., Zhang, C., Zhang, Y., Yang, X., Chen, J., Xu, J., Xue, R., and Zhou, B.: Effects of cleaner ship fuels on air quality and implications for future policy: A case study of Chongming Ecological Island in China, Journal of Cleaner Production, 267, 122088, https://doi.org/10.1016/j.jclepro.2020.122088, 2020.