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 conducted long-term observations of ship-related SO_2 concentrations in the Shanghai shipping channel from 2018 to 2023 by the DOAS technique. Meteorological effects and urban background emissions were removed by machine learning techniques. The paper focuses on evaluation the effectiveness of low-sulfur policies in mitigating the SO_2 emissions from maritime activities, since during the time period marine fuel sulfur content (FSC) was restricted twice. The authors suggest that this DOAS-based approach is cost-efective tool for monitoring ship emissions and can be easily applied to other coastal regions. The paper is scientifically sound and well organized. It is very important not only for scientists but also for policy makers. However, I have some issues that should be addressed before recommending the paper to be accepted for publication in ACP.

Main comments

1. In Introduction, lines 40-50 considering the restrictions in FSC is somewhat confused and should be elaborated. Please, give explicitly the years when China designated and implemented ECA, DECA and CDECA (the abbreviation is mentioned in Fig.4 but not explained anywhere) areas, and the maximum sulfur content percents in the regions. Regarding Fig. 4 you should clarify the red bars. What happened in Jan 2020 (IMO regulation, FSC from 3.5% to 0.5%
) in the Yantze River Delta since if I understood correctly that region implemented the FSC of 0.1% already in Jan 2019.

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. Specifically, we now explain that in the manuscript. We also revised Fig. 4 accordingly: the previously used abbreviation "CDECA" has been replaced by "DECA 2.0" for consistency, and the meaning of the red bars is now clarified in the figure caption. The three red bars indicate key milestones in fuel sulfur control policies: In 2018 China designated DECA 2.0 (policy announced). In 2019 China implemented DECA 2.0, requiring ≤0.5% sulfur while sailing and ≤0.1% at berth within its territorial sea. In 2020, The International Maritime Organization (IMO) regulation came into effect globally, reducing the maximum fuel sulfur content from 3.5% to 0.5%.

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.

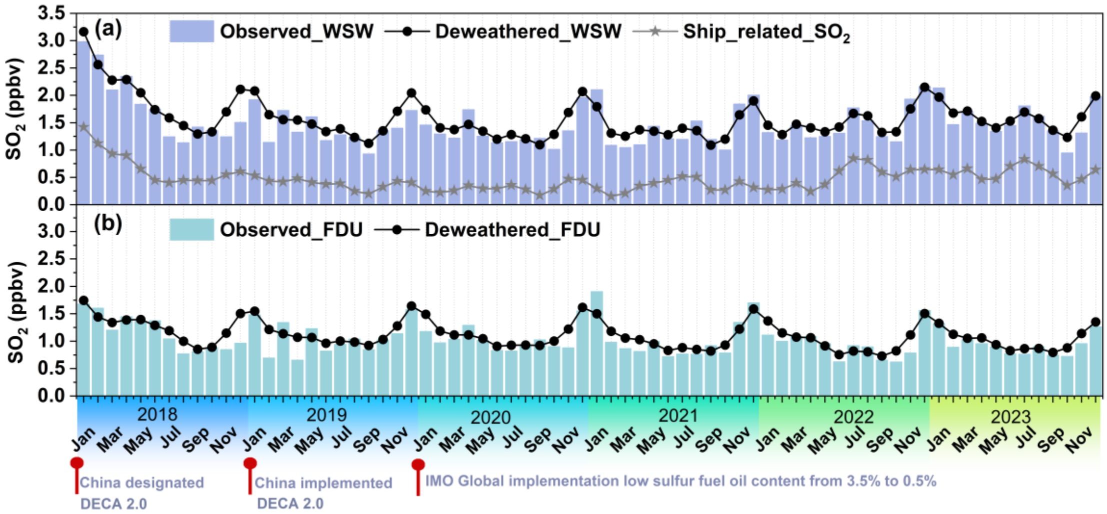

Figure 4: *Monthly observed_SO_2 concentrations based on DOAS and deweathered_SO_2 after weather normalization in WSW and FDU, and Ship_related_ SO_2 contributions during 2018-2023. (a) The light purple bars represent the monthly average Observed_SO_2 concentration at WSW; The solid black circles represent 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 210-215.*

2. The experimental setup is poorly described. In section 2, it is mentioned that over a thousand vessels pass daily the confluence of the two rivers. However, more information about the ships are needed such as the used engine (main or auxiliary), speed and age as they all affect the SO_2 emissions in addition of FSC and meteorological effects. It is important to know the stack heights and how well the DOAS system could capture the smoke plumes. At which heights the light emitter and the retroreflector located? More discussion is needed of these topics.

Response: Thank you for your comment. In response, we have made substantial additions to the manuscript and supplementary materials to clarify the local ship traffic conditions and the vessel characteristics relevant to SO_2 emissions. Including a description of the number of ships and types of composition, as well as statistics on the main engine, auxiliary engine, speed. In addition, we have also added explanations regarding the height of the optical path. Specifically, we have included two new figures (Figure S1, Figure S2) and a text (Text S1) in the Supporting information:

Figure S1 shows that the WSW channel experienced a generally increasing trend in ship traffic over the study period, with a recurring seasonal decline around the Chinese New Year holidays each year. Cargo ships and passenger boats consistently dominated vessel types.

To further address your point regarding engine characteristics, we have added Figure S2 that illustrates the temporal statistics of daily mean main engine (ME) and auxiliary engine (AE) power. Figure S2a shows the daily mean ME power with standard deviation and the 25th, 50th, and 75th percentiles, while Figure S2b provides the same statistics for AE power.

As expected, ME power is consistently and substantially higher than AE power, underscoring the dominant contribution of propulsion engines to total energy consumption and, consequently, SO_2 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). Moreover, the upward trend in the 50th and 75th percentiles of both ME and AE power since 2021 suggests a shift toward higher-powered vessels in recent years.

Speed is another important factor influencing emissions. However, unlike engine power (which is a fixed vessel attribute), speed is highly dynamic—even a single ship may shift between stationary, acceleration, and deceleration phases within a short time frame. Therefore, it is not meaningful to calculate a simple average speed across vessels or time. Nonetheless, AIS data reveal that the maximum vessel speed in this area can reach up to 52.62 knots, while many ships either remain stationary near the shoreline or navigate slowly (typically at 5–6 knots) within the channel.

The distance of the optical path from the water surface is not always fixed, as tidal water levels and ship cargo capacity affect the position of the optical path relative to the ship's stack. We add the objective altitude parameters of the observation site (including the altitude of the observation site, the height of the DOAS optical path from the ground, and the local tidal altitude) in the manuscript, and analyze the impact of altitude uncertainty on the research conclusions when answer Main comments #5.

We would like to emphasize that no single parameter—be it ship number, type composition, engine power, or speed—can independently and accurately represent SO_2 emissions. Unfortunately, AIS data do not provide information on vessel age, which is another potentially relevant parameter. However, as elaborated in our response to your main comment #7, we have used a bottom-up ship emission inventory (detailed in Text S5) to estimate SO_2 emissions based on AIS-derived parameters and ship characteristics, and to validate the observed variation in ship_related_SO_2 concentrations.

To address your concern directly, we have revised both the main text and the supplementary materials to include these new figures and additional contextual information, providing a more comprehensive and transparent description of ship activity in the WSW channel relevant to our experimental setup.

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 WSW, the light was emitted from a laboratory on the third floor (approximately 10 meters above ground level) of the Wusong Maritime Safety Administration building (ground elevation ~6 m above sea level) and reflected across the channel by an array of retroreflectors located on the opposite bank (which is also about 10 meters above ground level), forming a light path of 1,540 m. Given the

local tidal range of approximately 1-4 meters, the vertical height of the light path above the water surface varied between roughly 12 and 15 meters.” Please refer to Line 87-92.

In supplementary materials:

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.”

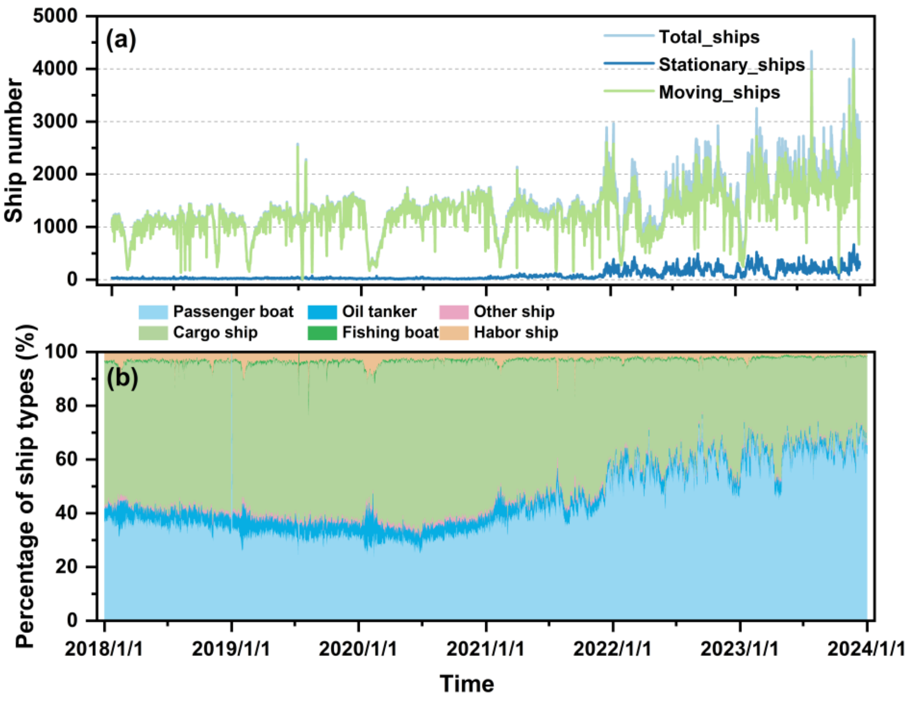

Figure S1. Temporal dynamics of daily ship traffic and ship type composition in the WSW channel (2018–2023). (a) Daily number of total ships, moving ships, and stationary ships detected from AIS records. (b) Percentage composition of different ship types over time, including passenger boats, cargo ships,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

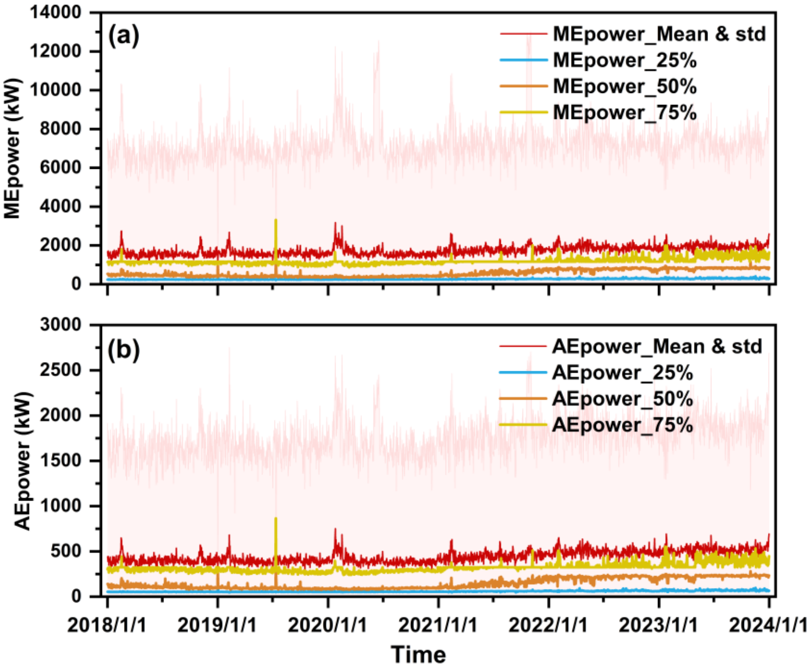

Figure S2. Temporal statistics of main engine and auxiliary engine power of vessels in the WSW channel (2018–2023). (a) Time series of main engine (ME) power, showing the mean ± standard deviation (shaded area) and the 25th, 50th, and 75th percentiles of power (kW). (b) Time series of auxiliary engine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

3. The Deweathered models used seven meteorological factors and time-related variables to capture the SO_2 pattern. Which was the most important variable for SO_2 explanation in WSW and in FDU. I suggest that you produce figures depicting the variable name as a function of variable importance similar as in Fig. 2 of Grange and Carslaw, 2019.

Response: Thank you for your comment. In response, we have produced figures depicting the variable importance for the Deweathered models at both the WSW and FDU sites, following the methodology of Grange and Carslaw (2019). Specifically, we trained 50 Extra-Trees Regression (ETR) models on bootstrap samples of the training data for each site and computed the permutation importance (with 95% confidence intervals) for each predictor variable.

Our analysis revealed that "wind direction" was the most important variable for explaining SO_2 variability at both sites, which aligns with the findings of Grange and Carslaw at the port city of Dover in England. This result is physically consistent because when winds originate from sectors with intensive ship activities (i.e., the direction towards the main shipping lanes), they transport emissions directly to the monitoring sites, leading to elevated SO_2 concentrations.

The resulting variable importance plots are now included as Figure S11 for WSW and FDU in the Support information. We have also added relevant instructions in the manuscript:

In the manuscript:

"We trained 50 ETR models on bootstrap samples of the training data for each site and computed the permutation importance (with 95% confidence intervals) for each predictor variable. The result shows that "wind direction" became the most important variable for explaining SO_2 variability at both sites (Fig. S11), which aligns with the findings of Grange and Carslaw (2019) at the port city of Dover in England." *Please refer to Line 199-203.*

In supplementary materials:

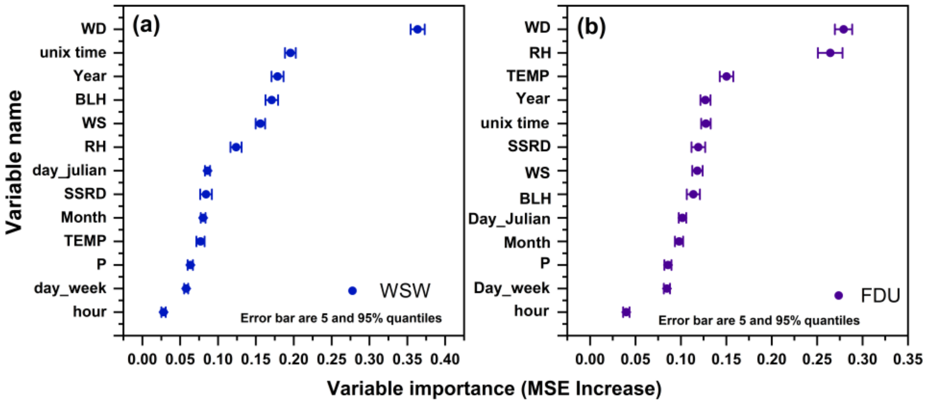

Figure S11. Variable importance plot for SO_2 at (a)WSW and (b)FDU between 2018 and 2023 calculated by 50 ETR models. The Mean Squared Error (MSE) increase quantifies how much predictive accuracy depends on each variable; a higher value denotes greater importance.

4. Is this the first time when the ETR learning model have been applied in deweathering ship SO_2 data. If not, add the references found in literature.

Response: Thank you for your comment. According to our investigation, while tree-based ensemble learning models have been widely used for deweathering air quality data, the application of the ExtraTreesRegressor (ETR) specifically for deweathering ship_related_SO_2 data has not been reported in the existing literature. Therefore, to the best of our knowledge, this is the first study to employ ETR for this purpose.

We have clarified this point in the manuscript:

"To the best of our knowledge, this study is the first to apply the ETR specifically for deweathering ship-related SO_2 data." *Please refer to Line 134-135.*

5. I suggest to present residual error plots between the actual and predicted SO_2 concentrations in both regions. Please include a discussion about the limitations of your study and about the uncertainties.

Response: Thank you for your comment. As recommended, we have added plots of the residual errors between the actual and predicted SO_2 concentrations for both regions in the revised supplementary material (Text S3, Figsure S4, S5). Specifically, the updated section now includes: Residual error plots (predicted minus observed SO_2) (Figsure S4a, b); histograms of residual frequency distribution (Figure S4c, d) and scatter plots illustrating the correlation between predicted and observed SO_2 (Fig. S5).

In supplementary materials:

"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"

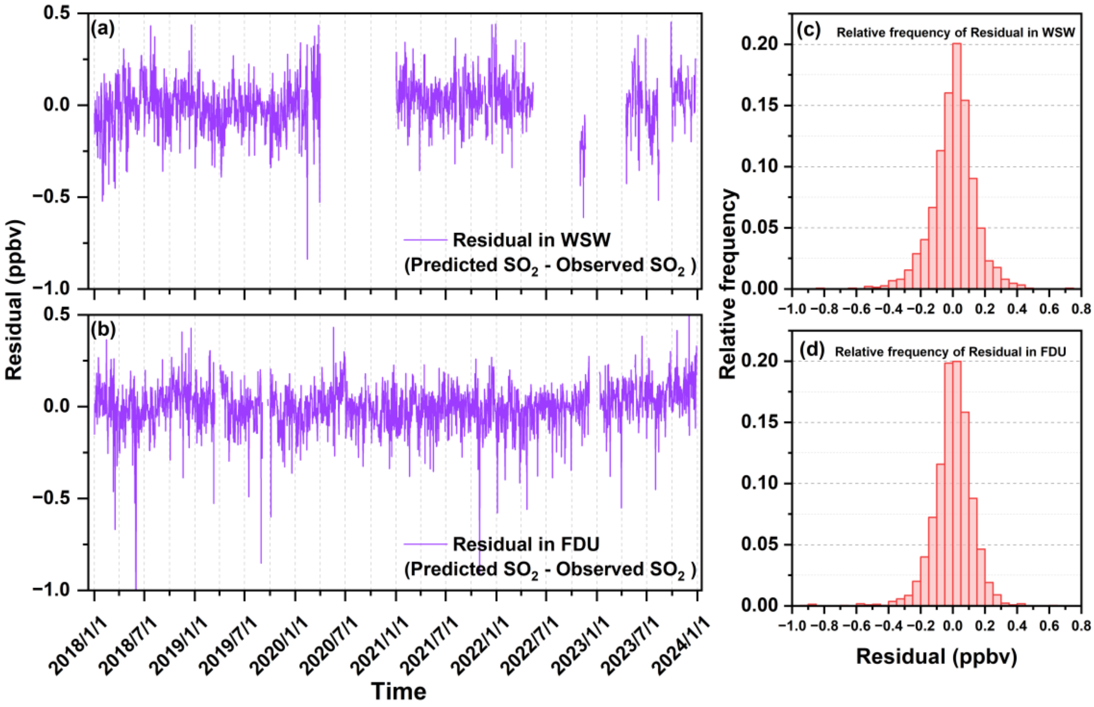

Figure S4. Time series and frequency distribution of residuals (Predicted SO_2 minus Observed SO_2) at the daily mean scale for (a, c) WSW and (b, d) FDU during 2018–2023.

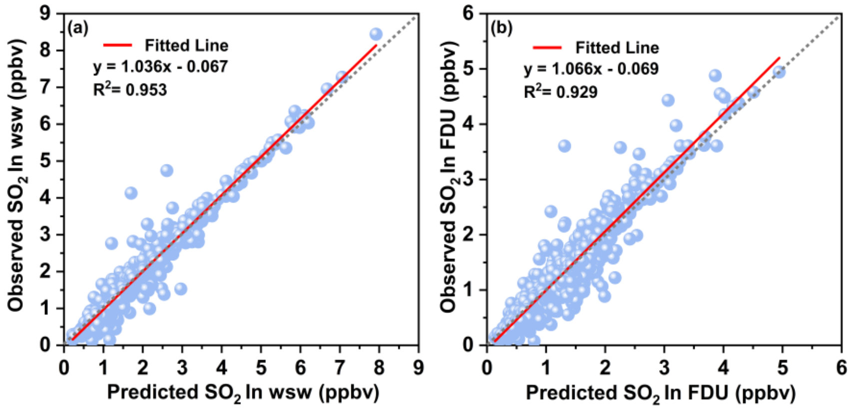

Figure S5. Scatter plots between predicted and observed SO_2 concentrations at the daily mean scale for (a) WSW and (b) FDU.

We have supplemented the discussions regarding uncertainties and limitations in both the manuscript and the supplementary material. In the Conclusion section of the manuscript, we have addressed the shortcomings of this study in terms of data, modeling, and experimental design in the form of future perspectives, offering suggestions for potential future developments. Furthermore, a detailed explanation of the causes of these limitations and their possible impacts on the results of this study has been provided in the supplementary materials:

In the manuscript:

"However, when expanding this framework to other regions with varying maritime traffic densities and regulatory contexts, or when applying it to monitor additional pollutants such as NO_X *and* $PM_{2.5}$*, it is imperative to acknowledge several methodological limitations. These include potential biases from the single-site background subtraction method, dependencies on meteorological reanalysis data in the Deweathered model, and uncertainties arising from vertical sampling geometry due to tidal variations and stack heights (detailed in Text S7). Although these systematic uncertainties do not substantially impact the conclusions supported by the large-sample data, they indicate that more precise data—such as using image recognition to determine specific ship activity and stack characteristics—would be necessary for finer-scale studies, such as quantifying emissions from individual ships. These factors should be carefully considered in future applications." Please refer to Line 359-357.*

In supplementary materials:

Text S7. Limitations and Uncertainties

"Although this study provides valuable insights into the contribution of maritime shipping to ambient SO_2 *in Shanghai, several limitations and uncertainties should be acknowledged.*

From a data perspective, an additional source of uncertainty lies in the background subtraction method, which assumes that the FDU site accurately represents the urban land-based SO_2 *level. In China, stringent emission control policies have led to a substantial reduction in land-based* SO_2*, and our long-term meteorology-adjusted analysis at FDU confirms that its background concentrations have already declined to relatively low levels with only minor interannual variability. Nevertheless, some degree of spatial heterogeneity in urban* SO_2 *emissions is unavoidable. As a result, the land-based contributions at FDU and WSW may still differ slightly, introducing potential bias in the background subtraction. However, such uncertainties are unlikely to affect the robustness of our analysis at broader temporal scales (e.g., monthly averages).*

From a model perspective, the Deweathered approach relies on the choice of input variables and on the assumption that meteorological impacts can be fully captured by the ERA5 parameters and time-related covariates. Other relevant factors, such as local-scale turbulence or unmeasured meteorological drivers, may not be fully represented.

From the experimental design perspective, an important source of uncertainty in this study arises from the vertical sampling geometry of the DOAS system. The light path was located approximately 10 m above ground level, with the observation site itself about 6 m above mean sea level. Tidal variation (1–4 m) and vessel stack heights mean that the intercepted section of the SO_2 *plume could vary between individual events—capturing different segments of the vertical plume profile depending on stack height and tidal level.*

However, the DOAS setup and tidal conditions remained broadly consistent during the entire 2018–2023 period, and vessel types and traffic patterns did not experience abrupt structural changes. Therefore, this geometric uncertainty is systematic and comparable across years, and is unlikely to bias the interannual patterns observed in the plume concentration distributions. Our analysis focuses on the relative frequency of plumes within specific concentration ranges and their temporal trends, rather than on deriving absolute emission rates for individual vessels.

If a quantitative estimation of individual vessel emissions were to be conducted, obtaining the actual stack height of ships would be crucial. Unfortunately, such information is not contained in the AIS system. A feasible solution would be to integrate camera-based observations to capture photographs of vessels passing through the light path at moments of elevated SO_2 signals, allowing stack height and plume geometry to be determined more accurately. This is a direction our group intends to pursue in future work to further reduce the uncertainties associated with vertical sampling geometry."

6. Not clear why the authors say that "After normalizing the meteorological influences, the (Deweathered) SO_2 concentrations in WSW and FDU showed an overall decrease during the observation period, while Table 1 shows that in WSW the SO_2 concentrations increased in 2021 2023 (lines 198-199).

Response: Thank you for your comment, which highlights an ambiguity in our original phrasing. We confirm that the Deweathered concentration time series at WSW indeed shows an increase in 2021-2023 after an initial decrease, as clearly presented in Table 1. Our intended meaning was that the process of Deweathered produces a data series that is overall lower in magnitude than the original observed data, However, in terms of the absolute value of concentration, it is true that it has increased. We have revised the relevant statements.

In the manuscript:

"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.

7. The time development of monthly (or annual) number of vessels or ship types in WSW would help to interpret the results in Figs. 3 and 4.

Response: Thank you for your valuable comment. We fully understand the interest in interpreting the results presented in Figs. 3 and 4 using ship activity data. In response to your suggestion, we have added a new figure (Fig. S12) in the supplementary material, which illustrates the monthly number of ships in the WSW channel from 2018 to 2023. Additionally, we have revised the manuscript to direct interested readers to the relevant supplementary material sections.

In the manuscript:

"The higher degree of fluctuation at WSW compared to FDU can be attributed to the more irregular ship emissions at WSW. 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 203-205.

In supplementary materials:

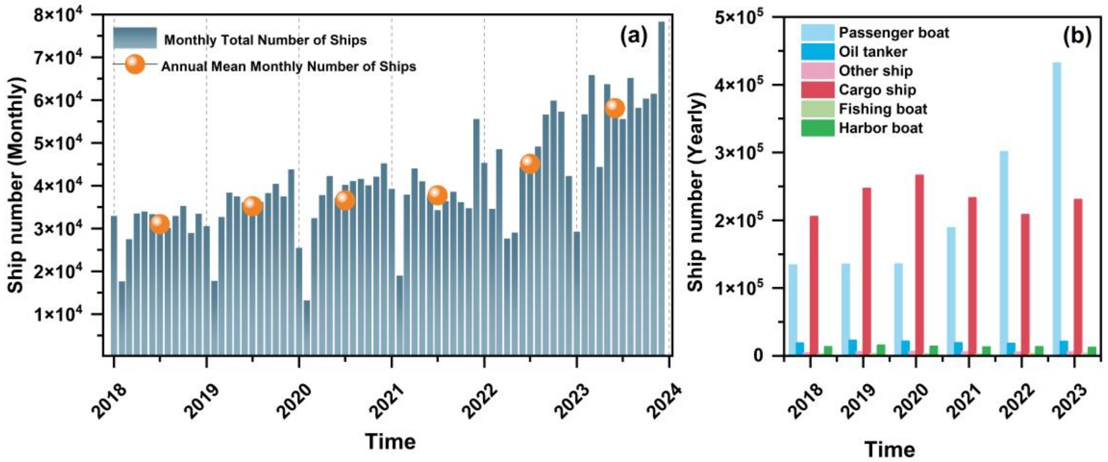

Figure S12. Annual variation of shipping activity in the channel from 2018 to 2023. (a) Monthly total number of ships and annual mean values. (b) Yearly ship number by ship type (cargo, oil tanker, passenger boat, fishing boat, and harbor boat). (For a more robust parameter of activity, a ship emission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

We would also like to take this opportunity to highlight that, beyond simple ship numbers, we have employed a more advanced indicator—a ship emission inventory derived from AIS data—to better interpret variations in Ship_related_SO_2 within the channel. Furthermore, the conclusions drawn from our study may provide valuable insights for refining future ship emission inventories.

Our approach and reason are detailed in the revised supplementary materials (Text S5, S6; Figure. 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 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. "*

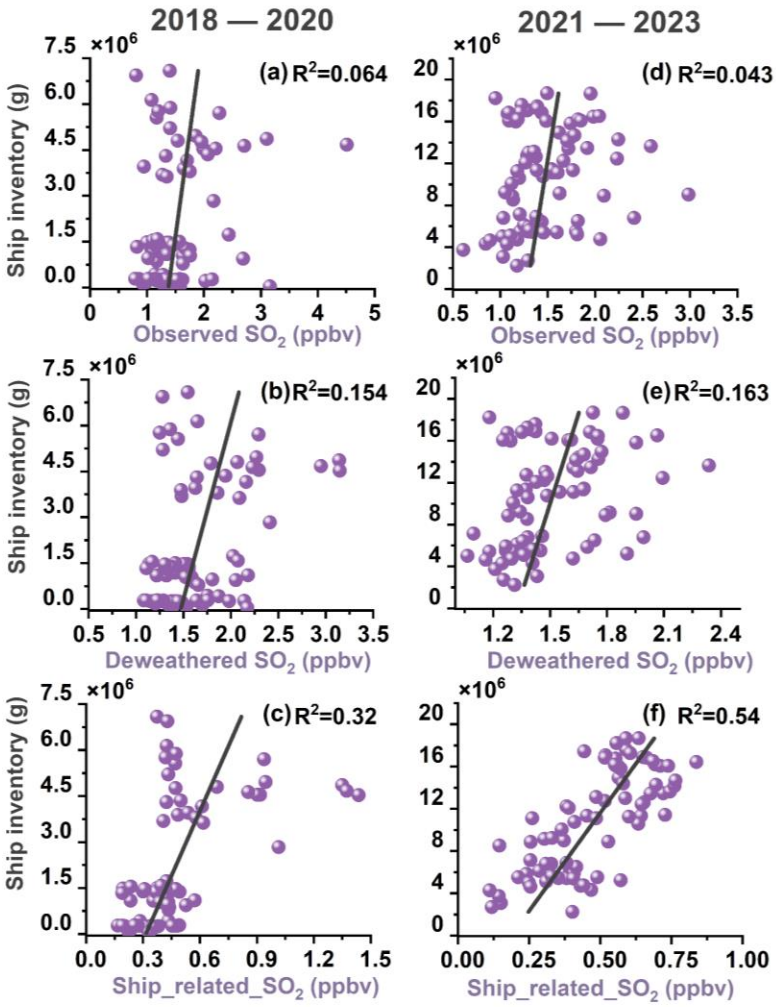

Figure S15. Correlations between 14-day mean SO_2 concentrations (x-axis) at WSW site and ship SO_2 inventory (y-axis), divided into three categories: (a, d) Observed_SO_2 concentrations, (b, e) Deweathered_SO_2 concentrations, and (c, f) Ship_related_SO_2 concentrations. (a–c) correspond to the policy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

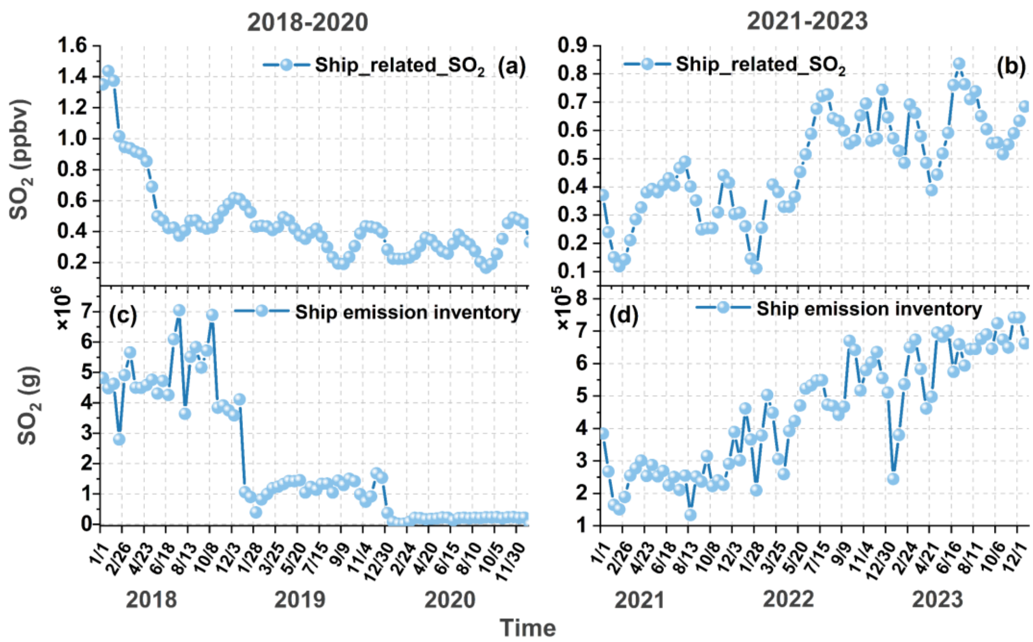

Figure S16. 14-day mean variations of Ship_related_SO$_2$ concentrations and emission inventory in the Wusong channel from 2018 to 2023. (a) and (b) represent the 14-day mean Ship_related_SO$_2$ derived from observations for 2018–2020 and 2021–2023, respectively. (c) and (d) show the corresponding 14-day mean SO$_2$ emissions from the ship emission inventory during the same periods.

8. Figure 6 shows that low SO$_2$ emission plume [4,6) ppbv started to increase from 2018 and to decrease just after 2020 while the lower SO$_2$ emission plume [2,4) started to increase (Fig. S6). The authors suppose that this reflects the transitional effect of policy implementation, and some ships have started to use lower sulfur fuels during the restriction period. Do you have any empirical evidence of this assumption? What about the alternative fuels such as liquid natural gas (LNG) and biofuels? Another reason might be the use of better scrubbers that efficiently clean the exhaust gas, particularly sulfur oxides. Numerous cargo ships are moving in the channel. I would like to see the annual development of the number of different ship types regarding Fig. 6a. More discussion is needed about this topic.

Response: Thank you for your comment. As suggested, we analyzed the annual numerical trends of different ship types (Figure S12b). The results show that the absolute numbers of major emission sources (such as cargo ships and Passenger boat) exhibited a stable or growing trend from 2018 to 2023. In other words, the patterns observed in Figure 6 of the main text—namely, the sharp decline in the frequency of high-concentration SO$_2$ plumes (>10 ppbv) alongside the systematic variations in low- and medium-concentration plumes (e.g., the overall increase of [2,4) ppbv and the initial rise of [4,6) ppbv)—occurred against a backdrop of increasing vessel numbers. This strongly demonstrates that the observed trends in SO$_2$ emissions were not driven by changes in the scale or composition of the ship fleet (since emission sources were actually increasing), but rather by changes in the emission behavior of individual ships.

Besides, based on our literature review, there are indeed multiple technical pathways to reduce SO$_2$ emissions from ships, including the use of fuels with lower sulfur content, LNG, biofuels, and exhaust gas cleaning systems (scrubbers). Among these, switching to low-sulfur fuels has been the most common choice, as it requires little or no modification of existing engine systems. Although LNG offers price advantages, its adoption has been limited by the high retrofitting costs of ship engine systems, and in practice, LNG-powered ships mainly operate in certain regions of Western Europe. Other fuel-switching options, like biofuels, generally involve substantial costs for engine retrofitting, which has restricted their widespread application. Scrubbers, while allowing the continued use of high-sulfur fuels, may cause secondary environmental problems due to wastewater discharges, and their uptake remains low, with less than 5% of the global ships reported to be equipped with such systems. In the revised manuscript, we have expanded the discussion to provide supporting explanations and references.

In supplementary materials:

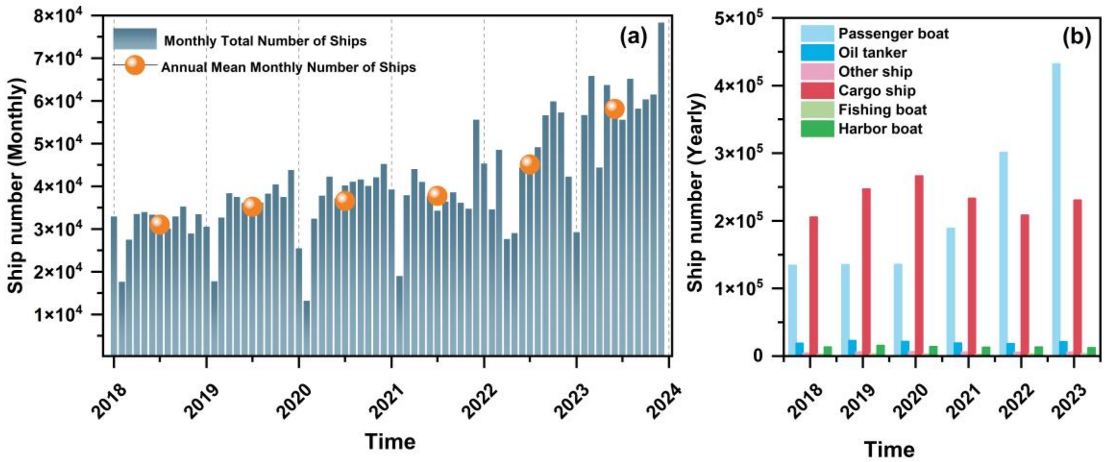

Figure S12. Annual variation of shipping activity in the channel from 2018 to 2023. (a) Monthly total number of ships and annual mean values. (b) Yearly ship number by ship type (cargo, oil tanker, passenger boat, fishing boat, and harbor boat). (For a more robust parameter of activity, a ship emission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

In the manuscript:

"The peak frequency of SO_2-rich plumes within the [6,30) ppbv range exhibits a general declining trend year by year, while the numbers of major emission sources in the channel (cargo ships and passenger boats) exhibited a stable or growing trend from 2018 to 2023 (Fig. S12b). This demonstrates that the observed trends in SO_2 emissions were not driven by changes in the scale or composition of the ship fleet (since emission sources were actually increasing), but rather by changes in the emission behavior of individual ships." Please refer to Line 266-270.

"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.

Minor comments

9. Which value did you use for the absorption cross section of SO_2, I could not find any value in the references given in Supple.

Response: Thanks for your comment, we add the absorption cross section of SO_2 in Supplement file. In this study, the SO_2 absorption cross sections obtained by Vandaele et al. (2009) were used in DOAS retival. This cross section has also been successfully used by our research team in previous studies(Cheng et al., 2019; Zhu et al., 2022). Here, the DOAS fitting was performed in the 299–308 nm wavelength range, and we have now added this reference and the relevant information to the Supplementary Material to ensure clarity.

In supplementary materials:

Table S1. The detection limits of DOAS retrieval and the analytical residual.

Observed Station	Trace gas	Fitting window (nm)	absorption cross sections	Polynomial degree	Detection limits	Residuals
WSW	SO_2	299~308	SO_2(Vandaele et al., 2009),NO_2 (Voigt et al., 2002), HONO (Stutz et al., 2000), HCHO (Meller and Moortgat, 2000), and solar spectrum (Kurucz, 1984)	5	0.13 ppbv	0.00054
	NO_2	365.3-380.4	NO_2 (Voigt et al., 2002), HONO (Stutz et al., 2000), HCHO (Meller and Moortgat, 2000), and solar spectrum (Kurucz, 1984)	5	0.51 ppbv	0.00043
	O_3	280.6-290.6	O_3 (Voigt et al., 2001a; Voigt et al., 2001b) , SO_2(Vandaele et al., 1998), HCHO (Meller and Moortgat, 2000), and NO_2 (Voigt et al., 2002)	5	2.51 ppbv	0.00154

	HCHO	313~341	HCHO (Meller and Moortgat, 2000), NO_2 (Voigt et al., 2002), SO_2(Vandaele et al.,1998), O_3 (Voigt et al., 2001a), HONO(Stutz et al.,2000)	5	1.10 ppbv	0.00057
FDU	SO_2	299~308	SO_2 (Vandaele et al., 1998),NO_2 (Voigt et al., 2002), HONO (Stutz et al., 2000), HCHO (Meller and Moortgat, 2000), and solar spectrum (Kurucz, 1984)	5	0.11 ppbv	0.00045

The reference is:

Vandaele, A. C., Hermans, C., and Fally, S.: Fourier transform measurements of SO_2 absorption cross sections: II.: Temperature dependence in the 29000–44000cm−1 (227–345nm) region, Journal of Quantitative Spectroscopy and Radiative Transfer, 110, 2115-2126, https://doi.org/10.1016/j.jqsrt.2009.05.006, 2009.

10. line 250: check the year, should be 2021 instead of 2023.

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 348-349.

11. Abbreviations should be defined when they appear for the first time, at least XGBR (line 98), ERA5 (line 148) and BEAD (line 228).

Response: Thank you for your careful reading and helpful suggestion. We have revised the manuscript to define the abbreviations upon their first appearance as follows:

"Therefore, this study developed two data-processing models using extremeGradientBoostingRegressor (XGB) and ExtraTreesRegressor (ETR)." Please refer to Line 108-110.

"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 with the

spatial resolution of 0.25° × 0.25°(Marshall, 2000; Hersbach et al., 2020).” Please refer to Line 164-168.

“This analysis was conducted by separating high-time-resolution DOAS observations using the Baseline Estimation and Denoising using Sparsity (BEADs) algorithm (Ning et al., 2014), as illustrated in Fig. 5 (an example from January 12 to 13, 2018).” Please refer to Line 252-254.

12. Give reference for ERA5.

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 with a spatial resolution of 0.25° × 0.25° (Marshall, 2000; Hersbach et al., 2020).” Please refer to Line 164-168.

13. Explain the error bars Fig. S3.

Response: Thank you for your comment. In response, we have added an explanation of the error bars both within Fig. S3 (Figure S9 now in the revised Supplementary material) and in its caption. Specifically, the error bars represent the standard deviation of hourly mean values, calculated using the STDEVP function in Excel across all hourly averages.

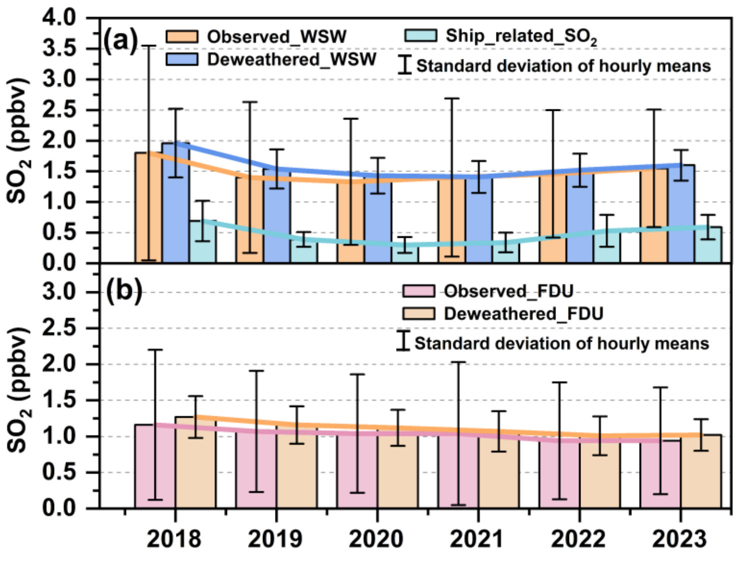

Figure S9. Yearly average SO_2 concentrations at two sites from 2018 to 2023. (a) Observed and deweathered SO_2 concentrations at the WSW site, with the contribution of ship-related SO_2. The orange bars represent observed SO_2 concentrations (Observed_WSW), the blue bars represent deweathered SO_2 (Deweathered_WSW), and the green line with stars shows ship-related SO_2. (b) Observed and deweathered SO_2 concentrations at the FDU site. The pink bars represent observed SO_2 (Observed_FDU), while the orange bars represent deweathered SO_2 (Deweathered_FDU). Error bars represent the standard deviation across hourly mean values.

14. The use of dots and commas should be checked in the main text as well as in supple.

Response: Thank you for your comment, we have checked the usage of punctuation marks in both the main text and the appendices. Our revisions are as follows:

"The first model was used to impute missing SO_2 concentration data (Fig 2a),"→ "The first model was used to impute missing SO_2 concentration data (Fig. 2a)," *Please refer to Line 109.*

"These models identify patterns between feature and target vectors in large datasets to make predictions or decisions, have been maturely applied to environmental research" → "These models identify patterns between feature and target vectors in large datasets to make predictions or decisions, and they have been maturely applied to environmental research," *Please refer to Line 114-115.*

"At the FDU site (Fig. 3a,b), the observed SO_2 concentrations display significant variability and weak inter-annual correlation, indicative of the influence of meteorological factors." → "At the FDU site (Fig. 3a, b), the observed SO_2 concentrations display significant variability and weak inter-annual correlation, indicative of the influence of meteorological factors." *Please refer to Line 171-172.*

"In contrast, at the WSW site (Fig. 3c,d), the Deweathered model also reduces variability and enhances the stability of the annual trends compared to the observed data." → "In contrast, at the WSW site (Fig. 3c, d), the Deweathered model also reduces variability and enhances the stability of the annual trends compared to the observed data." *Please refer to Line 175-176.*

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

15. lines 104-105: a subject is missing after comma

Response: Thank you for pointing this out. We have revised the sentence to correct the grammatical structure. The updated version now reads:

"These models identify patterns between feature and target vectors in large datasets to make predictions or decisions, and they have been widely applied in environmental research." *Please refer to Line114-115.*

16. line 108: "(Including…" should start with a small letter.

Response: Thank you for pointing this out. We have corrected the capitalization. "Including" has been changed to lowercase to read:

"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." *Please refer to Line 117-119.*

17. line 165: remove the dot after the word Figure

Response: Thank you for your comment, we already remove the dot after word Figure.

"Figure S9 displays their annual changes by a column chart." Please refer to Line 183-184.

18. line 37: a space is missing between the words "from" and "shipping"

Response: Thank you for your comment, we have corrected the spacing error by adding the missing space between "from" and "shipping".

"However, with the rapid expansion of maritime trade, SO_2 emissions from shipping are projected to keep increasing." Please refer to Line 37-38.

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.

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