Response to the referee

In the manuscript "Spatial and seasonal variability in volatile organic sulfur compounds in seawater and overlying atmosphere of the Bohai and Yellow Seas" Yu et al., compare surface measurements and depth profiles of marine OCS, DMS and CS2 in two different seasons (spring and summer). Accompanied by ancillary data (ocean temperature, salinity, chla, nitrate, DOC) the authors try to interpret their data related to production and loss processes of each sulfur compound. Finally, using also atmospheric OCS, DMS and CS2 measurements they calculate the sea-to-air-flux of the described sulfur compounds.

Measurements of sulfur compounds in the ocean and atmosphere are scarce (especially CS2 and OCS in comparison to DMS), but they are urgently needed to investigate their influence on a global scale. Therefore, this dataset is a valuable contribution to increase the number of measurements during different seasons in this specific marginal sea area. However, the scientific content of the manuscript remains pretty descriptive. The discussion part seems very comprehensive but at the same time stays superficial. The introduction part ends with "…we investigate…variability of COS, DMS, and CS2…to better understand production and loss processes of VSCs". Here, I strongly disagree. The authors know and also mention in the introduction the different parameters (e.g. CDOM, DMSP, bacteria) which influence (photochemical or biological) production and loss of the presented sulfur compounds but this ancillary data is not presented here. I suggest to revise the manuscript following the main comments below, also with respect to the English language, before publication.

Response: The influences on a global flux have been evaluated.

The methods have been added some description, and some deep discussion has been added. The sentence in the introduction part has been changed.

The English language has been edited by a professional language editing service-EditorBar Language Editing. The certificate of language editing is shown in the last page.

General comments

Introduction

The introduction should be clearly structured. Presentation of different production and loss processes is mixed for COS, DMS and CS_2. It would help the flow to clearly distinguish between these three compounds and their production/loss processes.

Response: The production and loss processes COS, DMS, and CS_2 have been shown in different paragraph and clearly distinguish between these three compounds.

Material and Methods

The sampling/measurement procedure of the ancillary data (section 2.4) should be presented in a bit more detail. Also, phosphate and silicate measurements are missing in this section, although data is presented in Table S3 and Table S4.

Response: The sampling/measurement procedure of the ancillary data (section 2.4) has been presented in a bit more detail. Also, phosphate and silicate measurements have been added in this section.

Discussion

The authors explain parts of their results and also relate their results to other findings. However, some parts should go in to the introduction part as this is state-of-the-art knowledge. This would also give the introduction a more detailed content, also with respect to the findings of this study.

Response: Some parts have been added in the introduction part. See also, "COS production is dependent on UV radiation, chromophoric dissolved organic matter (CDOM), cysteine, and nitrate concentration (Lennartz et al., 2021; Li et al., 2022). COS production rates increase with increasing nitrate concentration (Li et al., 2022).", "$^{3}CDOM^{*}$, $^{1}O_2$, H_2O_2, and $^{\bullet}OH$ produced by the photochemical reaction of DOM react with DMS and produce COS and CS_2 (Modiri Gharehveran and Shah, 2021).".

Oceanic COS is known to have a distinct seasonal, but also diurnal cycle due to the photochemical production. This is not at all mentioned or discussed in the manuscript, especially with respect to the different times of samplings (spring to summer but also potentially on a diurnal basis.

Response: Seasonal and diurnal variations of COS discussion has been added in section 4.1.3.

I was missing the main story in the discussion part. The authors relate their findings to some other studies in the same area also with respect to different seasons which is good and valuable. However, what is about the bigger picture or how can the results from the YS and BS be referred to other marginal seas? The authors highlight the influence of oceanic sulfur emissions on the atmospheric chemistry. How strong are emissions of those compounds compared to other regions and on global scale? The authors state in the conclusion "marginal seas…make a considerable contribution to the global sulfur budget" but miss to discuss and prove this with actual numbers. The DMS climatology from Hulswar et al. (2022) (not even cited) or a compilation of CS_2 and COS measurements by Lennartz et al. (2020) could help as a start to discuss the findings in a global context.

Response: Hulswar et al. (2022) has been cited. The following sentences about global fluxes have been added in the discussion section 4.3.

The model of Lennartz et al. (2021) was not used to evaluate the global sea-air fluxes of DMS, OCS, CS_2 in this study due to a lack of parameters, i.e., the absorption coefficient of CDOM at 350 nm (a350), global radiation (converted to UV radiation), and sea surface pressure. Therefore, the global sea-air fluxes of DMS were calculated following Hulswar et al. (2022) with minor modifications. The global sea-air fluxes of OCS or CS_2 were evaluated by the mean sea-air fluxes of OCS or CS_2 multiplied by the ocean area and the time. The global sea-air fluxes of DMS, OCS, and CS_2 were 21.3, 2.3, and 2.0 TgS $year^{-1}$, respectively. The global sea-air flux of DMS was similar to the

results of Hulswar et al. (2022) (27.1 TgS $year^{-1}$). In comparison, the global sea-air fluxes of OCS and CS_2 were 15.9- and 9.9-fold higher than the results of Lennartz et al. (2021). The different calculation method we used may overestimate the global sea-air fluxes of OCS and CS_2. The another reason may be the high sea-air fluxes of OCS or CS_2 in the BS and YS because marginal seas are significantly influenced by anthropogenic emissions (Watts, 2000). The sea-air fluxes of DMS, OCS, and CS_2 in the BS and YS were 28.2, 3.1, and 2.7 GgS $year^{-1}$, accounting for 0.10%, 2.23%, and 1.44% of global sea-air fluxes. The BS and YS comprise 0.13% of the global sea area; therefore, they contribute considerably to global sea-air fluxes.

Specific comments

ll.39: “Some researches indicates that the ocean is the source of VSCs. Opposite results also were reported that the ocean is the sink of VSCs.” I do not think that this is true for DMS and CS2. In case the authors relate this to COS (as the citation suggests), please revise this sentence to make it COS specific.
Response: The sentence has been changed into “Some studies have indicated that the ocean is a COS source (Chin and Davis, 1993; Yu et al., 2022), whereas others have shown that the ocean is a COS sink (Zhu et al., 2019).”.

ll.57: “The production and loss of VSCs involves in phytoplankton and bacteria synthesis, zooplankton grazing, bacterial degradation, sea-air diffusion, photo-oxidation and/or photochemical reaction”. This is a very general sentence. Please be more precise with respect to the different compounds presented in the manuscript.
Response: The sentence has been changed into “The production and loss of DMS involve phytoplankton and bacteria synthesis, zooplankton grazing, bacterial degradation, and sea-air diffusion (Schäfer et al., 2010). COS and CS_2 production are related to photo-oxidation and/or photochemical reactions (Lennartz et al., 2020; Xie et al., 1998).”.

ll.68: “In this study, we investigate… the effects of YSCWM on VSCs distributions to better understand the production and loss processes of VSCs.” As already mentioned I think this sentence is too ambitious with respect to the dataset.
Response: The sentence has been changed into “…and the effects of the YSCWM (the 35 °N transect) on the VSC distributions to better understand the distributions and impact factors of VSCs in Chinese marginal seas.”.

l.98: “Based on the similarities…” I guess the authors want to say that they calculated the concentrations with help of a calibration using standard gases?
Response: Yes, the reviewer is right. The sentence has been changed into “The VSC concentrations were calculated after calibration using standard gases (Fig. S1).”.

l.110: “The detection limit of the method for VSCs was 2.5-3.5 ng…” According to section 2.2 the authors used 30mL of sample to measure COS, DMS and CS_2 in seawater. Using this volume and a detection limit of 2.5ng would result in a detection limit concentration of ~1.3nmol/L. However, most of the presented DMS data and all of the presented CS_2 and COS data falls below this threshold. Please check.
Response: The original detection limit is wrong. We have checked the data and the sentence has been changed into “The detection limits of the method for COS, DMS, and CS_2 were 33 pg, 387 pg, and 22 pg and the measurement precision was 5.59%-11.70% (Tian et al., 2005).”.

ll.120: “...and selected ion monitoring mode (SIM).” What masses did the authors use for qualification and quantification of the different compounds?
Response: “The mass-to-charge ratios (*m*/*z*) for COS, DMS, and CS_2 were 60, 62, and 76, respectively”, which has been added in 2.2.

ll.161: “The distribution of CS_2…(Fig. 2)…was similar with that of DOC.” I do not see that.
Response: “, which was similar with that of DOC” has been deleted.

l.169: ”...which may have been due to the abundance of nutrients…” Please also show nitrate in both summer and spring figures and not only in the supplement.
Response: The nutrient data were provided by the cruise, see 2.4, therefore, it is unsuitable to show the figures in the main text. We show the figures in the supplement to avoid repeating presentation in the main text from others. Figures of phosphate and silicate have been added in Figure S2, and the sentence has been changed into “which may have been due to the abundance of nutrients (nitrate and silicate)”.

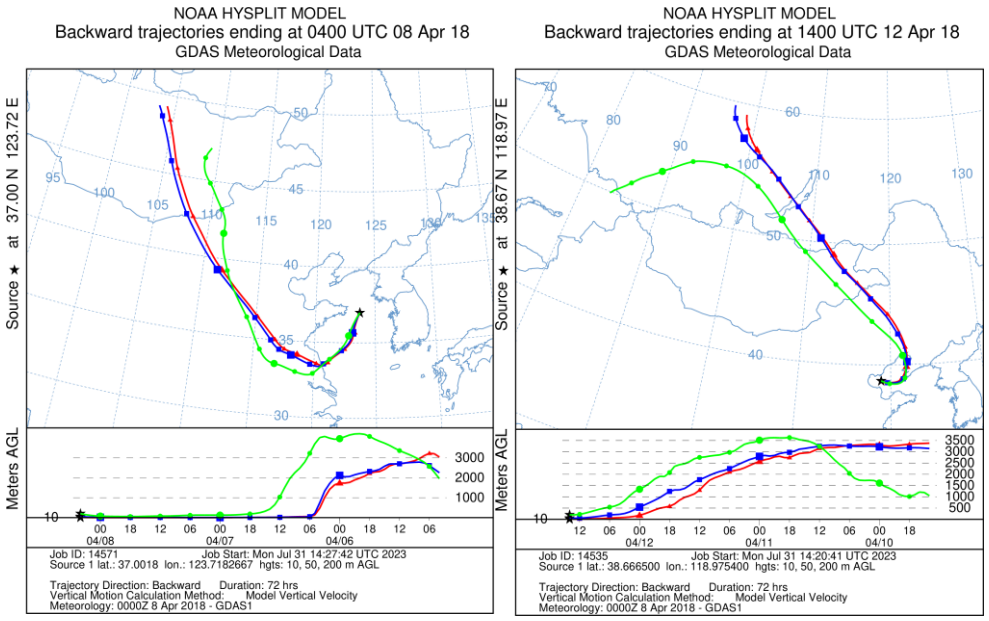
ll.201: “However, in the bottom waters of station H16, COS had a relatively high concentration (Fig. 5).” What means relatively? Please be precise with respect to the actual concentration or with respect to the sampling location the authors compare to.
Response: Thank you for your advice, the word “relatively” used here was Chinese English expression, and it has been deleted and the actual concentration was shown as “the COS concentration was high in the bottom waters of station H16 (0.465 nmol L^{-1})”.

ll.202: “The mean concentrations of Chl a, COS, DMS, and CS_2 at different depths were … higher in summer than spring.” It is not clear by “different depths” what numbers are related to each other.
Response: The mean concentrations of Chl a, COS, DMS, and CS_2 of the whole values at different depths were calculated and shown in the data, the original calculated data were wrong, they have been revised as “The mean concentrations of Chl *a*, COS, DMS, and CS_2 of all samples at different depths were 1.2-, 0.0-, 4.6-, and 1.0-fold higher or equal to those in summer (1.34 μg L^{-1}, 0.20 nmol L^{-1}, 4.38 nmol L^{-1}, and 0.158 nmol L^{-}

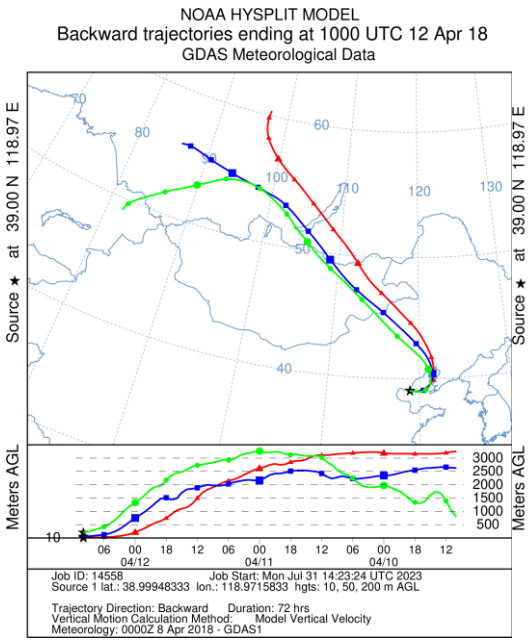
^{1}, respectively) than in spring (0.61 μg L^{-1}, 0.20 nmol L^{-1}, 0.78 nmol L^{-1}, and 0.080 nmol L^{-1}, respectively).".

Section 3.3.3: The title is misleading and results shown in this section should be moved to section 3.3.1 and section 3.3.2 to add more content to the respective sections.
Response: The title of 3.3.3 has been deleted and the results related to spring and summer shown in this section have been moved to section 3.3.1 and section 3.3.2 respectively to add more content to the respective sections.

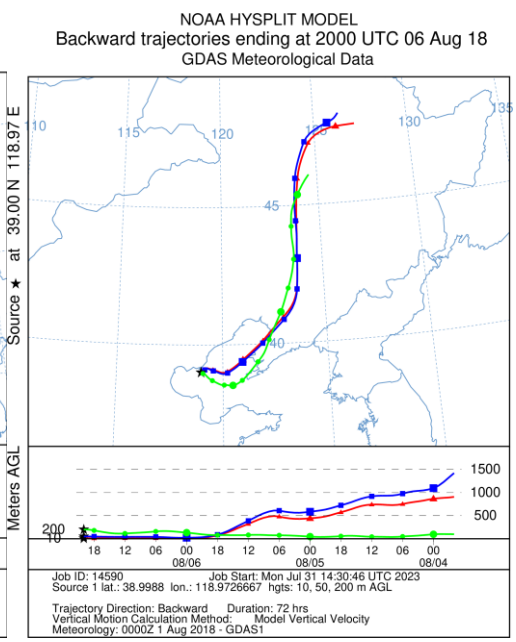
l.219 and Fig S2: "According to 72h backward trajectory…". Is there a reason why the authors started the trajectories at 500m, 1000m, and 1500m height? Do the authors have information about the marine boundary layer height? Otherwise I would suggest to start these trajectories at a much lower height in relation to the height of the actual measurements.
Response: Thanks for the suggestion of the reviewer. The original 72h backward trajectory is indeed at too high heights. The trajectories have been redrawn and with a much lower height (10 m, 50 m, and 200 m) in relation to the height of the actual measurements. See Fig. S3.



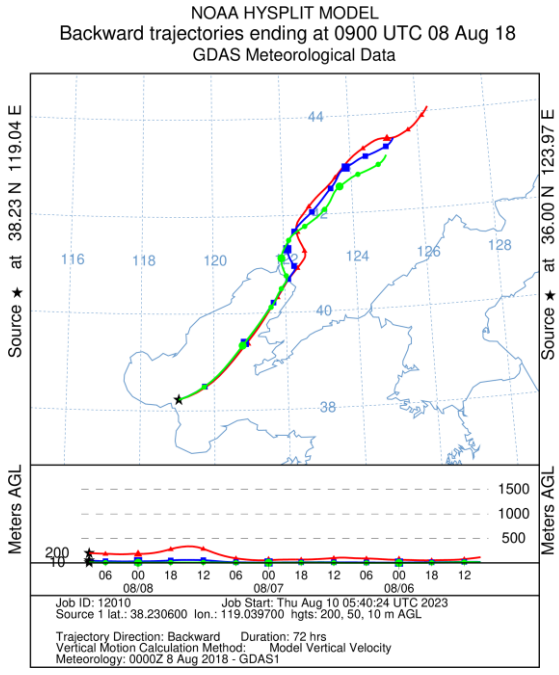
B08-spring B47-spring



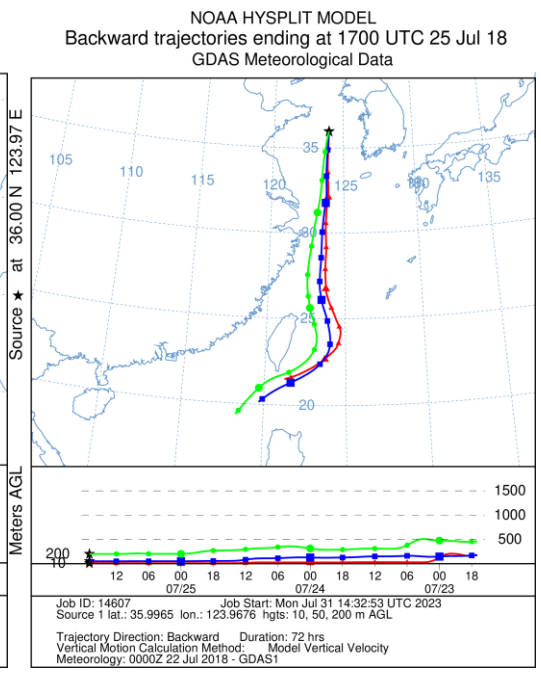
B49-spring



B49-summer



B64-summer



H09-summer

Figure S3. 72 h backward trajectory of the air mass above stations B08, B47, B49 in the BS in spring and stations B49, B64, H09 in summer of 2018.

ll.220: “The lowest atmospheric DMS concentration appeared at station B47 (Fig. 6a), probably due to the low DMS concentration in seawater (0.5 nmol L-1).” I was

wondering, why the authors only check the backward trajectories once for a single station and not for the whole area? Especially as B49 (backward trajectory provided, high atm DMS) and B47 (no backward trajectory provided, low atm DMS) are very close to each other.
Response: Backward trajectory of stations B49, B47, B08 in spring and B49, B64, H09 in summer have been redrawn to find the sources and the reasons of different VSCs mixing ratios. See Fig. S3.

l.230: "P>0.05" should be "P<0.05".
Response: Yes, the reviewer is right. "P>0.05" has been changed into "P<0.05" in the section 3.4.

section 3.4: Please structure this section logically.

Response: Section 3.4 has been structured logically as follows "A significant correlation was found between the DMS and CS_2 concentrations in the surface seawater in spring ($P < 0.05$, Table 1) and summer ($P < 0.01$, Table 1). A positive correlation occurred between the COS and DOC concentrations in seawater ($P < 0.05$) and between the CS_2 and Chl *a* concentrations in seawater ($P < 0.05$) during summer (Table 1). There was a significant correlation between the atmospheric COS and CS_2 mixing ratios in spring and summer ($P < 0.01$, Table 1).".

ll.300: "In this study, the concentrations of the three VSCs in seawater during summer were higher than those in spring, which may be due to the higher Chl a in summer than in spring." As already outlined in the manuscript, the three VSCs have different sources. Therefore, high chla as a general reason, seems a bit misleading.
Response: According to the comments, the sentence "In this study, the concentrations of the three VSCs in seawater during summer were higher than those in spring, which may be due to the higher Chl *a* in summer (mean: 1.60 μg L^{-1}) than in spring (mean: 1.19 μg L^{-1})." has been changed into "The significant positive correlations between the CS_2 and Chl *a* concentrations during summer may explain the higher CS_2 concentration in seawater during summer than during spring in this study.".

ll.370: "Wind speed was the main influencing factor…" Did the authors do any statistical analysis?
Response: According to the formula $F = k_w(c_w\text{-}c_g/H)$, where F is the sea-to-air flux of VSCs (μmol m^{-2} d^{-1}); k_w is the VSCs transfer velocity (m d^{-1}); k_w was calculated from wind speed and sea-surface temperature by the N2000 method (Nightingale et al., 2000), Therefore, "wind speed was the main influencing factor…". Statistical analysis has been done, and showed that "A significant correlation was found between the sea-to-air fluxes of COS, DMS, and CS_2 and the wind speed in spring or summer ($P < 0.05$).".

Figure 1: Only YSCWM is mentioned in the manuscript. To increase readability of the figure please delete all other current names.

Response: The other currents names have been deleted from Figure 1.

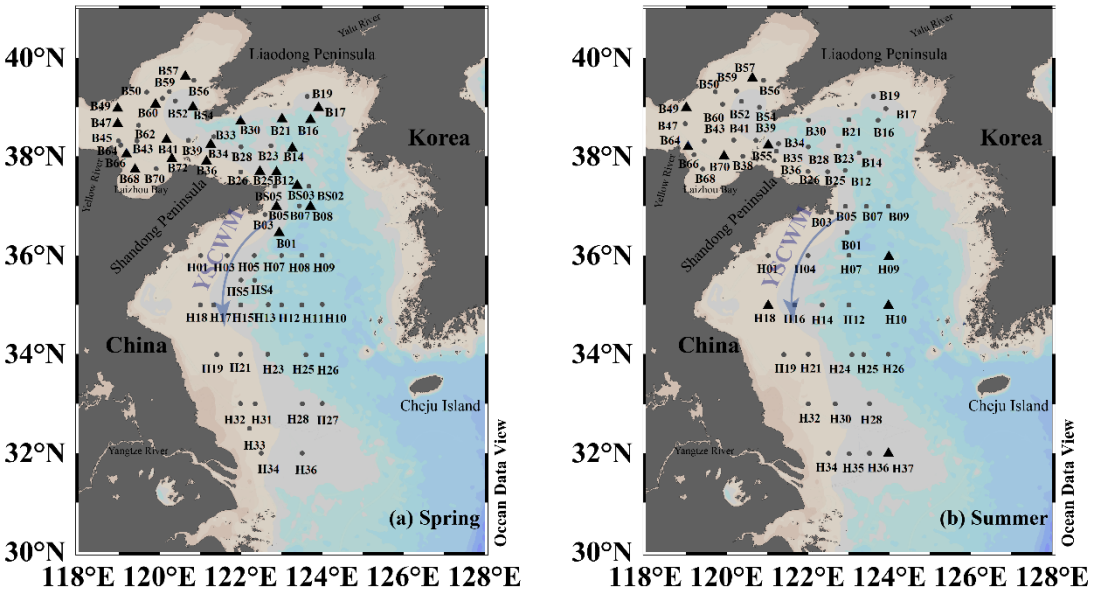


Fig. 1. Sampling stations in the Yellow Sea and Bohai Sea during (a) spring and (b) summer (▲ indicates stations where atmospheric samples were collected). Yellow Sea Cold Water Mass: YSCWM. The maps were plotted with Ocean Data View (ODV software) (Schlitzer, 2023).

Figure 6: Stations are presented in alphabetical order. However, in the manuscript, atmospheric measurements are often related to inshore or offshore locations. It would be great if this information could also be part of this figure for a better comparison and interpretation of the data. Both subplots next to each other and on the same y scale would improve comparability between spring and summer.

Response: To improve comparability, the atmospheric data have been drawn in ODV figures with black circles showing the values, and the inshore or offshore locations can be seen clearly. See Fig. 6.

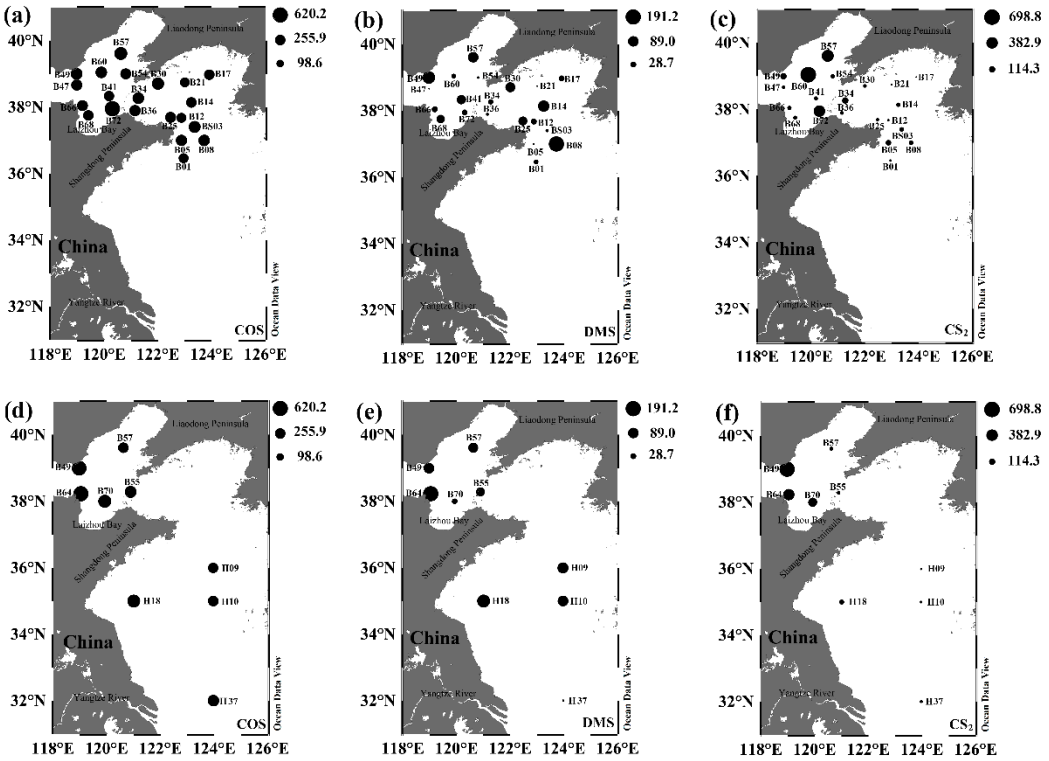
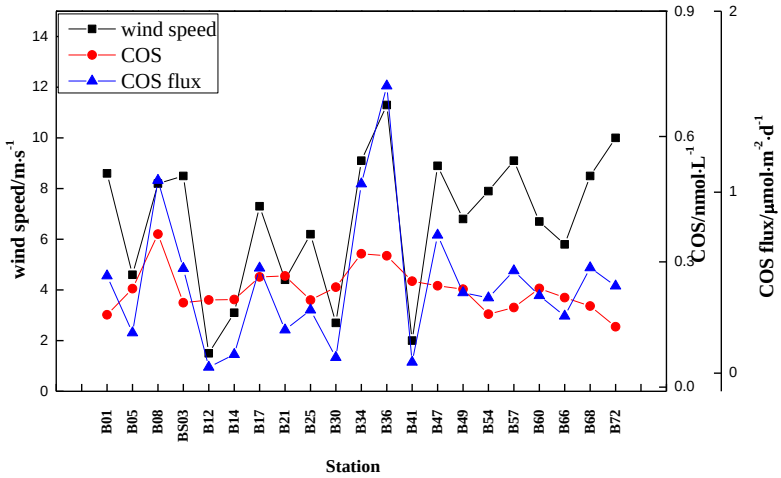
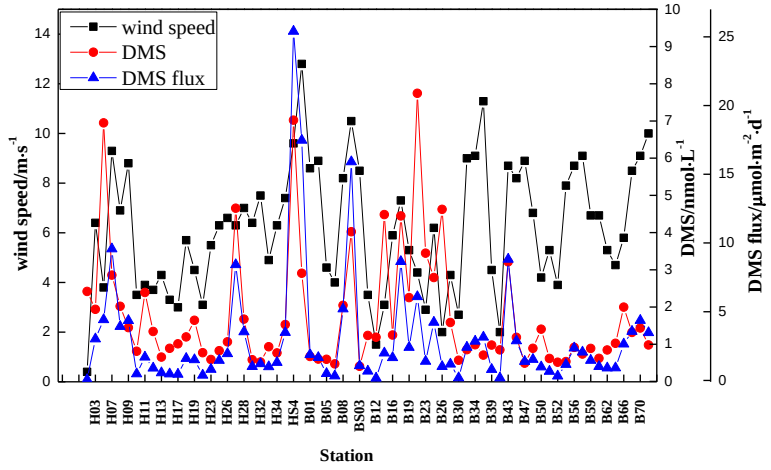


Fig. 6. Spatial distributions of COS, DMS, and CS_2 in the atmosphere over the BS and YS in (a)-(c) spring and (d)-(f) summer.

Figure 7 and 8: There are much more datapoints for the fluxes than atmospheric measurements? How is this possible? Are there atmospheric measurements missing in Figure 6?

Response: The original fluxes of COS and CS_2 were calculated using the mean atmospheric concentration, and DMS fluxed were calculated with DMS in ocean because the DMS concentrations in the atmosphere are much lower than those in the seawater. The DMS concentrations in the atmosphere can be considered as 0. Therefore, the DMS fluxes are not changed. The fluxes of COS and CS_2 have been revised and calculated using the formula $F = k_w(c_w\text{-}c_g/H)$ in section 2.3, and the Figure 7 and 8 have been redrawn as follows.





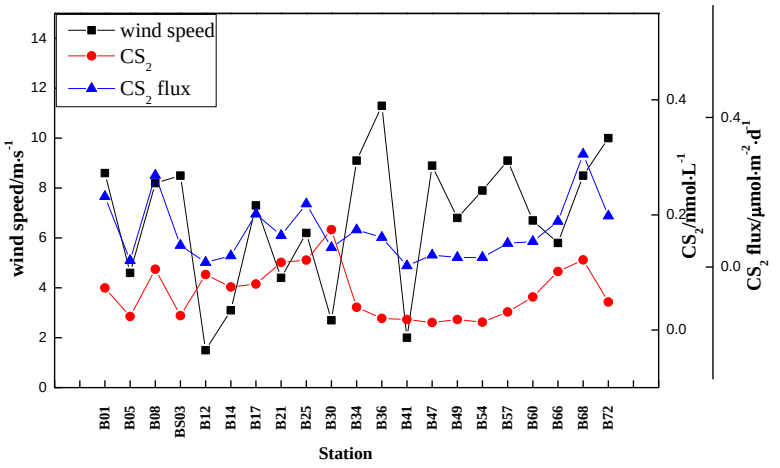


Fig. 7. Variations in sea-to-air fluxes of VSCs, VSCs concentrations in seawater, and wind speeds in the BS and YS in spring 2018.

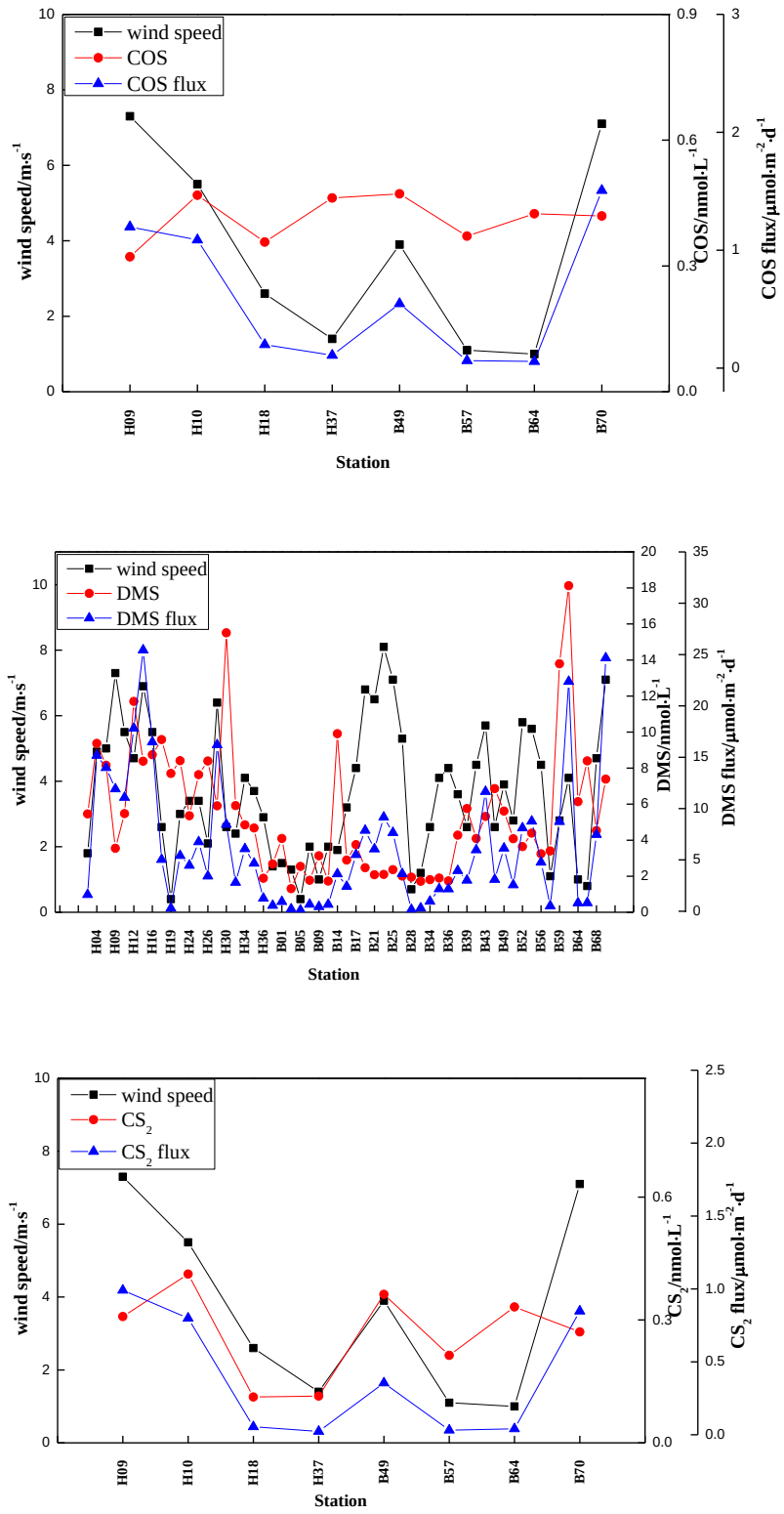


Fig. 8. Variations in sea-to-air fluxes of VSCs, VSCs concentrations in seawater, and wind speeds in the BS and YS

TableS2: Please add references to temperature dependent Henry constants.

Response: The reference to temperature dependent Henry constants (Tian, X.:

Determination of volatile s-compounds in the atmosphere and surface seawater of Chinese coastal areas, Peking University Master Thesis, pp 65, 2004, (in Chinese with English abstract).) has been added.

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CERTIFICATE OF LANGUAGE EDITING

The English writing of the following manuscript was carefully edited by a native English speaker.

Manuscript Information

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Title	Spatial and seasonal variability in volatile organic sulfur compounds in seawater and overlying atmosphere of the Bohai and Yellow Seas
Corresponding author	Gui-Peng Yang
Language writing before editing	□ Very poor □ Poor □ Fair ■ Good □ Very good □ Excellent
Recommendation after language editing	□ Submitting to target journal directly ■ Submitting to target journal after minor revision □ Re-editing required after major revision □ Not suitable for publication
Overview comments	This paper required edits with regard to wording, sentence structure, punctuation, language, tense, and grammar. There were some sections where your meaning was unclear. I added comments and offered alternate wordings for these sections or asked you to clarify if I could not interpret your meaning from the context. You should check those sections carefully to ensure that I did not change your intended meaning with my edits.

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