**Response to the referees and the editor**

Thank you for the comments of the editor and the referees. We have revised the manuscript according to the advices, which are marked with red words in the track-changes file.

**Response to the editor**

Dear Prof. Yang and Co-authors,
thank you for re-submitting the revised version of your manuscript. The revised manuscript has been reviewed by two reviewers. Both reviewers raised some minor but also some major concerns which need to be addressed before publication of the manuscript. Please address carefully all concerns with special consideration of the points raised by Rev. #1 (report #1).

I am looking forward to your revised manuscript.

Yours Sincerely
Hermann Bange

Response: Thank you for the comments of the editor. According to the two referees' comments, we have revised the manuscript.

**Response to the referee #1**

The revised manuscript "Spatial and seasonal variability in volatile organic sulfur compounds in seawater and overlying atmosphere of the Bohai and Yellow Seas"" Yu et al., addresses most of the reviewers' comments and concerns. Back trajectories from different sides are implement which serve as basis to partly explain differences in atmospheric mixing ratios. Furthermore, additional information about the calibrations are given. Global climatologies and databases for DMS, OCS and $CS_2$ are now included in the discussion section which serve as a comparison for the presented dataset. However, there might be an issue with the flux calculations (mentioned below) which would influence the results. Additionally, some parts of the manuscript are still unclear and need revision as stated below.

General comments
Calibration: 1.) Calibrations, shown in the supplement, reveal very different slopes and y-intercepts in between the two seasons (spring, summer). Perhaps the authors could give some information on how this is possible, as for both seasons the same standards have been used. 2.) The y-intercept for COS and $CS_2$ calibrations is very high. A high (positive) blank could be induced by e.g. contamination. $CS_2$ and COS are known to easily contaminate the sample e.g. when using non-PTFE or silicon tubing. Did the

authors check if these high “blank” values are only related to the standard used or if these blank values are also visible when measuring a “blank sample”? It would be great to hear an explanation of this issue.

Response: 1) Although the same standard was used, GC-MS exhibited different status at different seasons, including the chromatographic column status and ion source status, which may explain the different slopes and y-intercepts in spring and summer.

2) The tubes and trap used in the examination were all PTFE tubes. The high y-intercept for COS and $CS_2$ calibrations may be due to the small blank values when measuring a blank sample because of slight existence of COS and $CS_2$ in the carrier gas. There is no DMS in the carrier gas. However, the values measured in the samples were 10 fold of blank standard deviation, so the results in our research were reliable.

Henry’s law constants: According to the reference in Table S2 for the calculation of the DMS Henry’s constant, the given constant C1 is wrong, which would result in a different H by ~1 order of magnitude. If Dacey et al. (1984) was used, C1 should be 0.56 mol $L^{-1}$ $atm^{-1}$ instead of 0.048 mol $L^{-1}$ $atm^{-1}$. Please check if it is just a typo in the manuscript. Otherwise fluxes have to be recalculated and values including their implications for the discussion have to be revised.

Response: Yes, it is a typo in Table S2. We have changed 0.048 mol $L^{-1}$ $atm^{-1}$ to 0.56 mol $L^{-1}$ $atm^{-1}$ in Table S2.

Flux calculation discussion (ll.392): It is not clear to me why the authors recalculate global DMS, COS and CS2 fluxes. What are the presented global numbers in l. 397 based on? Did the authors use mean fluxes from their study and extrapolated them to global numbers assuming the same flux also at different locations in the world? This paragraph needs a revision.

Response: We misunderstood the meanings of the advice of the referee for the last revision. The paragraph has been deleted and we have compared our data with the database. See the 1$^{st}$ paragraph in sections 4.1.1, 4.2, and 4.3.

Specific comments

l.56: Please introduce $^3CDOM^*$, $^1O_2$, $H_2O_2$ and $^{\bullet}OH$.

Response: $^3CDOM^*$, $^1O_2$, $H_2O_2$ and $^{\bullet}OH$ have been introduced as “excited triplet states of chromophoric dissolved organic matter ($^3CDOM^*$), singlet oxygen ($^1O_2$), hydrogen peroxide ($H_2O_2$), and hydroxyl radical ($^{\bullet}OH$)” in the introduction.

ll.128: According to the answers of the reviewers, the authors calculated the flux of DMS assuming the atmospheric mixing ratio to be zero. This should be mention here in the method section. Additionally, the authors should mention in the discussion section, that their calculated DMS fluxes should be seen as upper limits (due to setting the atm mix ratio to zero). Why did the authors not use atm mix ratios for the flux calculation as they are available?

Response: The $C_g$ of DMS is assumed to be zero in this study. This is based on the fact that atmospheric mixing ratio of DMS are typically several orders of magnitude lower

than concentrations in seawater (Turner et al., 1996). The sentences have been added in section 2.3.
We have mentioned that the calculated DMS fluxes should be seen as upper limits due to setting the atm mix ratio to zero in the discussion section 4.3.

ll.132: The sentence “This method has been internationally accepted” can be deleted.
Response: The sentence “This method has been internationally accepted” has been deleted.

l. 189: surface water is referred to 4m depth. Later on (e.g. l.204) surface water is referred to 3m depths. Please be consistent.
Response: The surface sampling depths for spring and summer are different, which is ~4 m and ~3 m, respectively. See the values in Fig. 4 and Fig. 5 in data at https://doi.org/10.6084/m9.figshare.14971644. In order to be consistent (3−5 m), the sentence of “Surface seawater was sampled at a depth of 3−5 m.” has been added in section 2.1.

ll. 206: To my understand, a x-fold increase between A and B is defined as the ratio between A and B. Somehow, throughout the whole manuscript, it seems the authors define a x-fold increase as the ratio between (A-B) and B. Please check the correct definition or use other wording. In order to easier compare two values, I suggest to use the ratio A/B.
Response: According to the advices of the referee, x-fold increase has been changed to the ratio A/B in the revised manuscript.

ll.219: I do not understand the revised sentences about differences in atmospheric values of DMS between B47 and B49. According to the provided back trajectories for B47 and B49 (which are single runs and no ensemble or similar) I think the authors can not say that they are different. Therefore, the conclusion why B47 and B49 strongly differ in atm mix ratio is lacking.
Response: In order to see the differences, we have added the 12 h and 24 h back trajectories. The differences in the 72 h back trajectories can not be seen, while they can be observed in 12 h or 24 h back trajectories. The 12 h and 24 h back trajectories for station B47 showed that the air mass over station B47 differed slightly from that over station B49 as it traversed the land of Liaoning province. See Fig. S3. The sentence of “The air mass over station B47 differed slightly from that over station B49 as it traversed the land of Liaoning province (12 h and 24 h backward trajectories in Fig. S3).” has been added in 3.3.1.

ll.222: “high oceanic DMS concentrations at or near stations where air masses were passing through…may be the reason”. I do not understand this sentence.
Response: This sentence is our speculation according to the data and the air masses pathway without confirmation. Therefore, this sentence has been removed.

ll.320: The introduction of $^3CDOM^*$ should happen when using the abbreviation for the first time.
Response: The introduction of $^3CDOM^*$ has been added when using the abbreviation for the first time in the $4^{th}$ paragraph in the introduction in the revised manuscript.

ll.341: The paragraph about diurnal COS variations does not state or show any data from the actual study. It is not clear to me what exactly the authors want to highlight with this paragraph with respect to their dataset. If they did not investigate the atmospheric concentrations with respect to the light intensities or sampling time they should at least state that the sampling time could influence the measured COS atm mix ratios (besides wind speed, wind direction and oceanic concentration). Or did the authors sample the station always at the same time of the day?
Response: The paragraph about diurnal COS variations were added according to the other reviewer's suggestion. Yes, we did not evaluate the diurnal COS variations. We have simplified the discussion about diurnal variations, and the sentences "The maximum COS concentration occurred 3 h after the maximal global radiation intensity (COS: 15:00; global radiation intensity: 12:00) due to the balance between COS production and removal (Xu et al., 2001)." have been deleted, and pointed that "Therefore, the sampling time can influence the measured COS concentrations in the seawater.".

Figure 6: Please increase the resolution of the figure (higher quality). I was hard to see small dots (low concentrations) on the map.
Response: The small dots for low concentrations in Figure 6 have been enlarged and increased the resolution of the figure. We can provide the TIF formation for the figure to increase the resolution, if needed.

**Response to the referee #2**

The authors addressed most of my comments and a good number of the other reviewer's. However, I still have some issues with the manuscript. My specific comments are below (line numbers refer to track changes manuscript):

Introduction - this whole section is still a bit unclear. There is a whole paragraph dedicated to COS (2nd) and one for DMS (3rd), but $CS_2$ does not have the same. In the 4th paragraph (and on), all the compounds are mixed together, which makes it (them) somewhat confusing. There is no introduction sentence in the paragraphs that helps to set the main ideas.
Response: The sentences in the 4th paragraph have changed or added as "The photochemical reaction of DOM generates excited triplet states of chromophoric dissolved organic matter ($^3CDOM^*$), singlet oxygen ($^1O_2$), hydrogen peroxide ($H_2O_2$), and hydroxyl radical ($^{\bullet}OH$). These reactive species subsequently interact with DMS, resulting in the production of $CS_2$ (Modiri Gharehveran and Shah, 2021). The oxidation

reaction involving the OH radicals and $CS_2$ is a substantial contributor to the generation of $SO_2$, which subsequently leads to the production of acid rain (Logan et al., 1979).". The sentence "Production and loss processes of COS, DMS, and $CS_2$ have been documented by many researchers in the following manners." has been added in the 1st paragraph to help to set the main ideas of the 2-4 paragraphs.

Lines 43-44 - This sentence is repetitive (following the sentence before). Either take out this idea from the previous sentence or remove this sentence.
Response: The referee is right and the sentence is repetitive. The sentence of "COS production rates increase with increasing nitrate concentration (Li et al., 2022)." has been removed.

Lines 56-57 - The English is not clear here.
Response: The sentence "$^3CDOM^*$, $^1O_2$, $H_2O_2$, and $^{\bullet}OH$ produced by the photochemical reaction of DOM react with DMS and produce COS and $CS_2$ (Modiri Gharehveran and Shah, 2021)." has been changed into "The photochemical reaction of DOM generates excited triplet states of chromophoric dissolved organic matter ($^3CDOM^*$), singlet oxygen ($^1O_2$), hydrogen peroxide ($H_2O_2$), and hydroxyl radical ($^{\bullet}OH$). These reactive species subsequently interact with DMS, resulting in the production of $CS_2$ (Modiri Gharehveran and Shah, 2021).".

Lines 66-68 - This is repetitive in comparison to an earlier paragraph that talks about sources and sinks of COS. I am not sure why it is repeated here again. I think it should be removed as it doesn't add any additional information. In this paragraph, please just focus on what your study does and why it is relevant/important.
Response: The sentences of "Yu et al. (2022) investigated the distributions of COS, DMS, and $CS_2$ and sea-to-air flux in the Changjiang Estuary and the adjacent East China Sea, demonstrating that oceanic VSCs (COS, DMS, and $CS_2$) are sources of atmospheric VSCs. In contrast, Zhu et al. (2019) showed that the ocean was a COS sink." have been removed.

Lines 250-251 (261-262, 385) - I know the other reviewer asked for stats in relation to how the wind is a main controlling factor for the flux, but I think this is unnecessary. You use wind to compute the flux, often with a quadratic dependence. Therefore, if the wind is not correlated to the flux, there would be a problem. I think this should be removed - wind is an obvious controlling factor. The statement on line 249 -250 is appropriate.
Response: Yes. We use wind to compute the flux and wind is an obvious controlling factor. Therefore, lines 250-251 (261-262, 385) have been removed.

Section 4.1.1 - you mention pollution as a factor for $CS_2$ (also for DMS), in the context of seawater concentrations, but is there any evidence of invasion? You state that the ocean is a major source to the atmosphere for all gases, which means to me that the oceanic distribution controls what is in the atmosphere (not the other way around).

Response: No, there is no direct evidence of invasion, and this is a speculation. The sentence of “For example, rayon production is the main source of anthropogenic $CS_2$ (Campbell et al., 2015) in the northern cities of the BS.” in section 4.1.1 has been removed.
Thanks a lot for the advice. The atmospheric sources are not only oceanic distribution. Therefore, the sentences in the abstract and conclusion have changed “major” into “important”.

Lines 316-321 - There is much focus on sources of the gases, but could slower or less loss also be responsible here?
Response: The sentences of “In addition, the loss processed include exhalation, downward mixing, and hydrolysis. Among these processes, hydrolysis is the main sink (Xu et al., 2001). Slow hydrolysis rate may be another reason to explain the high COS concentrations in the surface seawater.” have been added.

Section 4.3 – I am a bit confused by the discussion and revision of this section. The idea was not necessarily to use the method of Lennartz et al. (2021) to compute the fluxes for COS, but to compare to the database. How do your calculations match other comparable areas (i.e., marginal seas). Then why is there a therefore on line 394 about how DMS was computed? Also, why are global fluxes extrapolated? I think these findings simply need to be compared to others to give context. Finally, again, I do not understand the argument about anthropogenic emissions. Are the anthropogenic emissions from runoff or so, so that they go directly into the water?
Response: We misunderstood the meanings of the advice of the referee for the last revision. The paragraph has been deleted and we have compared our data with the database. See the 1st paragraph in sections 4.1.1, 4.2, and 4.3.
No, the anthropogenic emissions were not meant from runoff. They mean coal combustion, industrial production, et al., which contribute to the VSCs mixing ratios in the atmosphere. The anthropogenic emissions discussion has been removed because of the revision in this paragraph.

A note on back trajectories – I want to make sure I am clear here…a back trajectory of 72 hours is not useful for DMS. It is fine if you say that only a few of the points are useful (the ones that represent the last 24 hours), however, you should state where those are. Is each dot 12 hours? If so, the trajectory, for example, over B49 does not pass over land within that time period. Therefore, land sources are not a good explanation for DMS.
Response: The 12 h and 24 h back trajectories for DMS has been added in Fig. S3. “The air mass over station B47 differed slightly from that over station B49 as it traversed the land of Liaoning province (12 h and 24 h backward trajectories in Fig. S3)” (added in section 3.3.1), where DMS may loss when passed over the land.” See Fig. S3.

A statement of the atmospheric lifetimes of the compounds of interest should be made and compared with the trajectories. For COS (long-lived), the trajectories are

meaningful. Land sources of COS should be stated clearly. What about $CS_2$? The lifetime is about 1 week, so the land sources are important for it too. You say that the spring back trajectories show anthropogenic influences, but how? Do you see reduced fluxes of COS and $CS_2$ when the back trajectories come from a certain area? Is there visible trend? All but one of the back trajectories show land influence. Was H09 in summer anomalous? I think the discussion of this information is not deep or always meaningful.
Response: A statement of the atmospheric lifetimes of the compounds has been added in section 4.2 to show the meaning of the trajectories.
Spring back trajectories show anthropogenic influences, including coal combustion, industrial production, et al. It is a speculation, because the spring back trajectories traversed the land.
No, we did not see. Not all back trajectories maps were drawn, and we only drew them for some stations.
No, H09 in summer was not anomalous, it is only an example for oceanic source.