

Response to the referee

This manuscript is the second from these exact set of cruises to the Yellow and Bohai Seas, by the same authors. Here we are shown the methods for observing COS, CS₂, and DMS (DMS is also in the other publication submitted to JGR) and their distributions (horizontal and vertical). Air and water values of the gases were measured and air-sea fluxes computed. Certain factors deemed relevant are correlated with the measured values to understand sources and sinks of these gases in the air and water. This manuscript requires a major overhaul before it can be published. The English needs to be thoroughly revised and the main ideas need to be clearer. What are the major findings from this work? Although the measurements are valuable, in order for them to be published in a scientific journal, there needs to be some insight or something new found. How does this contribution further our understanding? In addition, I am not sure if it is appropriate to publish the DMS values here without citing the other article that has been written about them (I was a reviewer of that article as well). Related to that point, other sections of the article should not be direct copies of the other manuscript submitted about this cruise (methods, etc.). Please check that.

Response: Our manuscript had been edited by a professional language editing service-EditorBar Language Editing. See the revised manuscript. The certificate of language editing is shown in the last page.

The major findings of this work are the seasonal variations in VSCs, distributions of VSCs and the impact factors, and discussion about the sources of atmospheric VSCs based on the 72 h back trajectories and the contribution to the global scale.

Yes, this manuscript had been rejected by the journal of JGR-Oceans before, and we have revised the manuscript according the comments of the reviewers. The spatial and depth distribution values have been cited from the Zhang et al. (2023, JGR-Oceans, in press). Other sections (methods, etc.) of the article have been checked and is not directly copied from the other manuscript submitted about this cruise. The figures about DMS are drawn by ourselves.

Specific comments:

General – Did the authors measure dissolved O₂ concentrations? This would be useful information to show, especially for the depth profiles. Also, when discussing the atmospheric values, it would be more proper to call them mixing ratios and not concentrations.

Response: No, we have not measured the dissolved O₂ concentrations. Although dissolved O₂ concentrations is useful information, unfortunately, it is not design in that cruise. We will measure it in the future research.

Thank you for your advice. The atmospheric values have been changed to call them mixing ratios.

Lines 54-55 - Citation formatting is awkward.

Response: The citation formatting has been revised as “Two different approaches (ice core and isotope measurements) were used to evaluate anthropogenic COS emissions (Aydin et al., 2020; Hattori et al., 2020).”.

Lines 83-85 – These two sentences can be merged into one.

Response: These two sentences have been merged into one “The stability of VSCs in fused silica-lined canisters has been verified during storage for 16 d at room temperature (Brown et al., 2015).”.

Section 2.2 – Why were different instruments used for the air and water measurements? The description of the atmospheric calibration is not clear, specifically regarding the primary standard. It seems like the primary standard was bought and it contained a 1 ppt mixing ratio for all three gases. Is this 1 part per trillion or part per thousand. I understand ppt = part per trillion. If so, this is a very low standard. It would also be nice to see some of the data from the calibrations, and perhaps some schematics of how the instruments were set up, in the supplemental material.

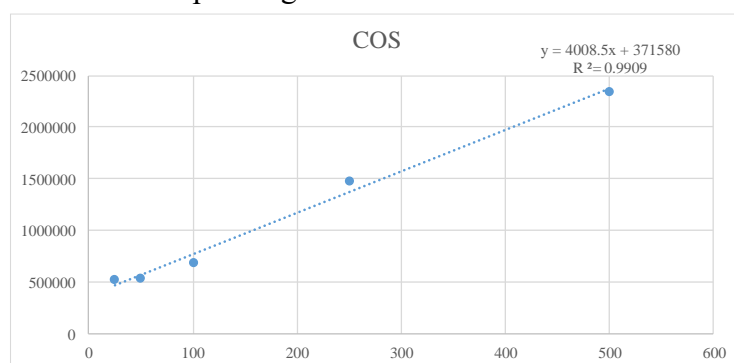
Response: A gas chromatograph (GC) can be used to measure oceanic VSCs. In comparison, the VSCs concentrations in the atmosphere is too low that they can not be measured by a GC, therefore, we used a gas chromatograph-mass spectrometer (GC-MS) to measure atmospheric VSCs.

Standard gases were bought and it contained a 1ppmv mixing ratio for all three gases, and they were diluted to 1 ppbv and 5 ppbv with Nutech gas diluter. The 1 ppbv and 5 ppbv standard gases were used as using standard gases mixing ratios, and then the VSCs standard curves were obtained as follows: (Fig. S1).

1. VSCs standard curves in spring

(1) COS standard curve:

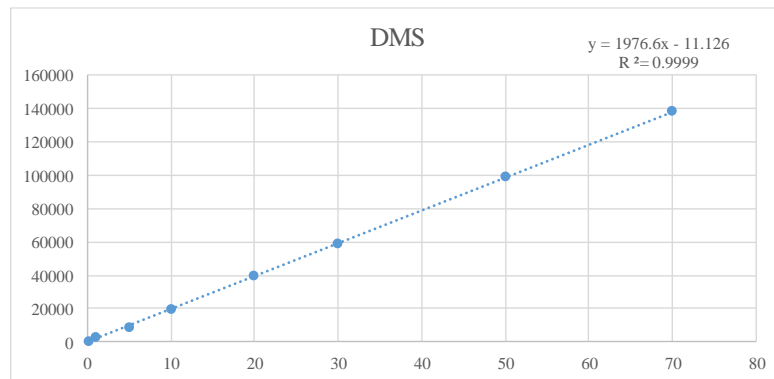
The 5 ppbv standard gas was used, the injection volumes were set as 5, 10, 20, 50, 100 mL. We use the standard gas mixing ratio * injection volume (25, 50, 100, 250, 500) as the X-axis, and the peak area detected as Y-axis. The mixing ratios of COS were calculated according to the peak area and correlative equation ($y = 4008.5x + 371580$). The injection volume of atmospheric gas is 200 mL.



(2) DMS standard curve:

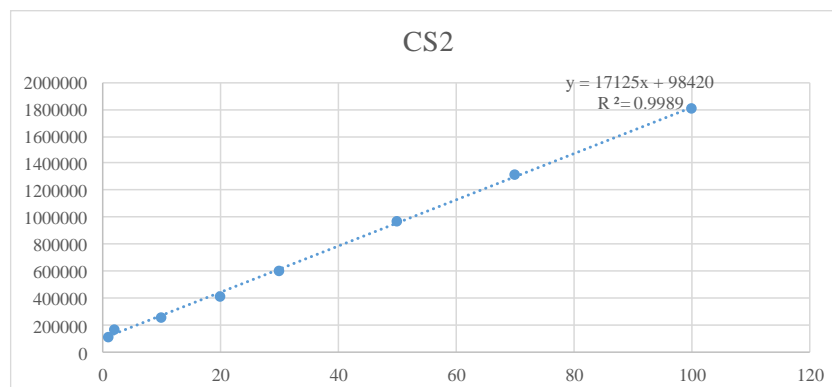
The 1 ppbv standard gas was used, the injection volumes were set as 0.2, 10, 20, 30, 50, 70 mL. We use the standard gas mixing ratio * injection volume (0.2, 10, 20, 30, 50,

70) as the X-axis, and the peak area detected as Y-axis. The mixing ratios of DMS were calculated according to the peak area and correlative equation ($y = 1976.6x - 11.126$).



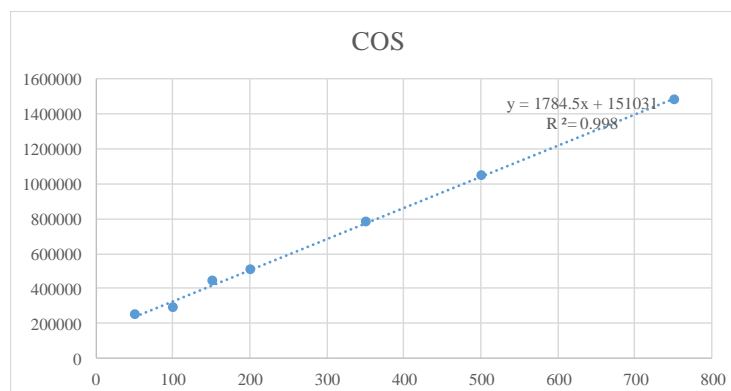
(3) CS₂ standard curve:

The 1 ppbv standard gas was used, the injection volumes were set as 1, 2, 10, 20, 30, 50, 100 mL. We use the standard gas mixing ratio * injection volume (0.2, 10, 20, 30, 50, 70) as the X-axis, and the peak area detected as Y-axis. The mixing ratios of CS₂ were calculated according to the peak area and correlative equation ($y = 17125x + 98420$).

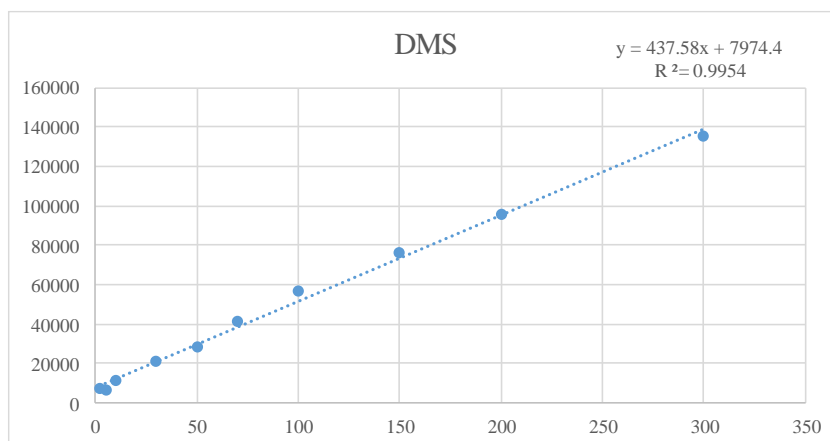


2. VSCs standard curves in summer

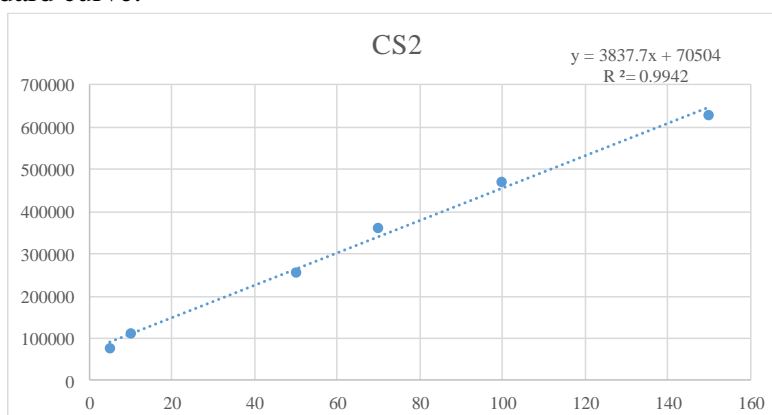
(1) COS standard curve:



(2) DMS standard curve:



(3) CS₂ standard curve:



3. The schematics of how the instruments were set up are as follows:

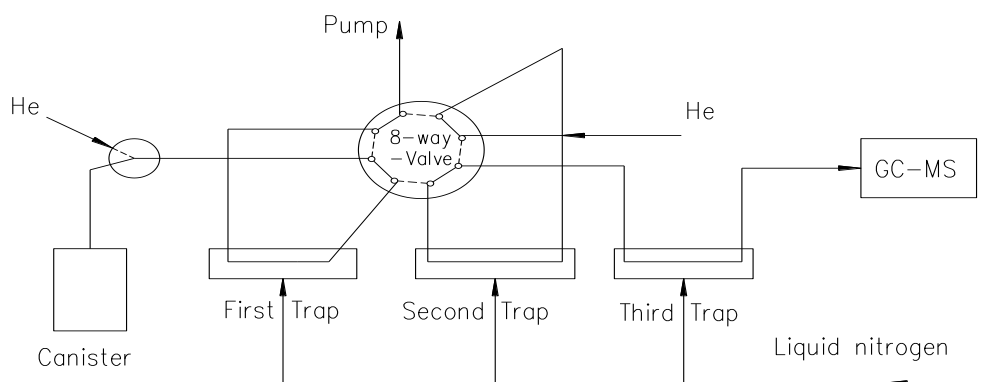
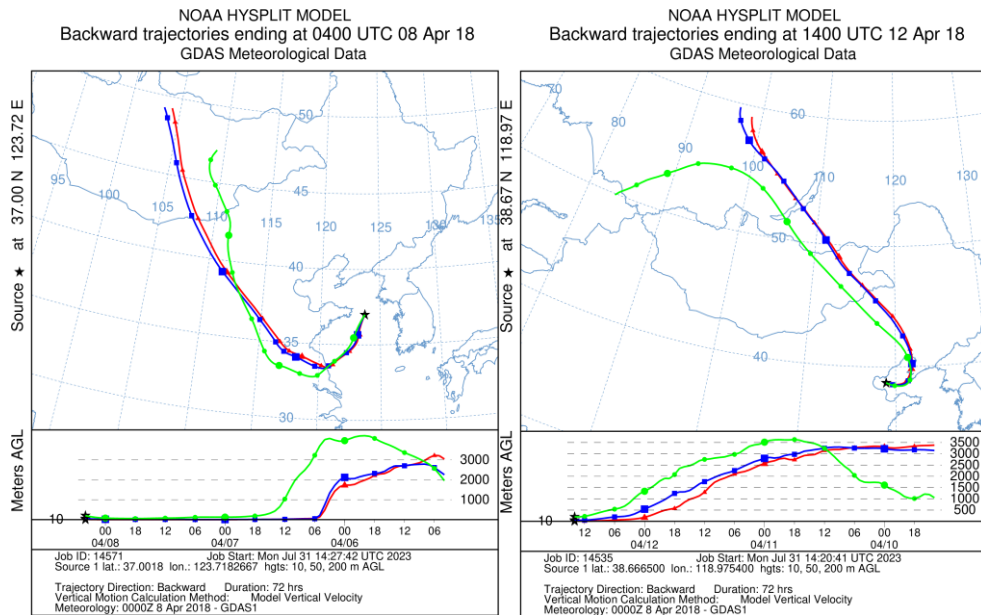


Fig. S1 The VSCs standard curves and the apparatus diagram used for analysis of VSCs in atmosphere

Section 3.3.3 – There is discussion of atmospheric sources here and some use of back trajectories (supplemental material), but I do not understand why only one station was examined in this way. I think back trajectories from various parts of the cruise track would be extremely useful. The atmospheric lifetimes of the gases are very different, so the back trajectories over multiple timescales for the various regions could tell a different story for each gas.

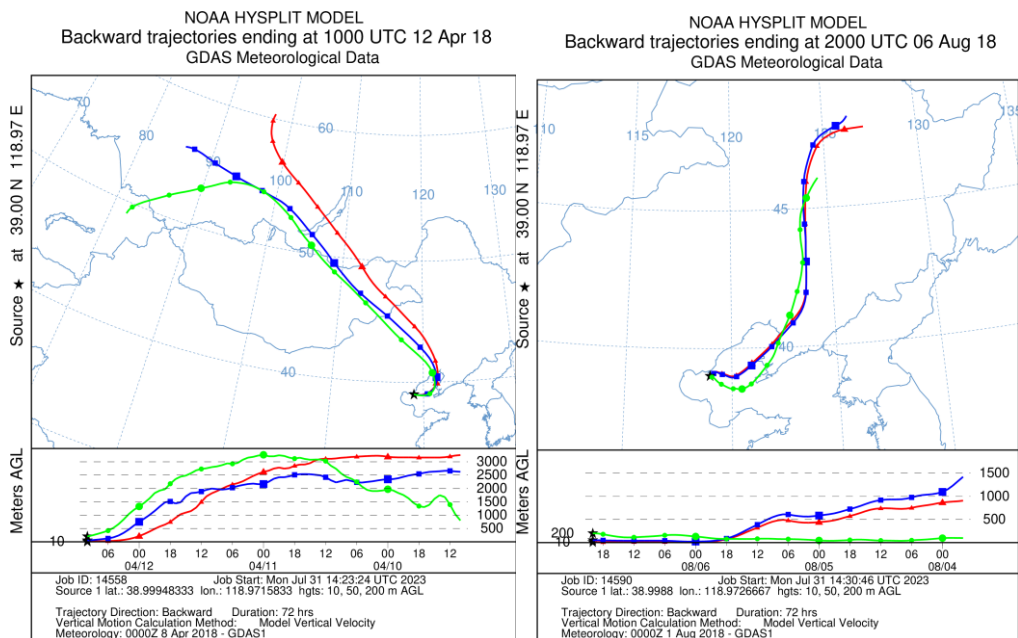
Response: Backward trajectory of stations B49, B47, B08 in spring and B49, B64, H09 in summer have been redrawn to find the sources and the reasons of different VSCs concentrations. See Figure S3.

72-hour back trajectories mean trajectories from 72 h to 0 h before sampling, therefore, which include 24 h and 48 h (1/3 and 2/3 of the line near the sampling station).



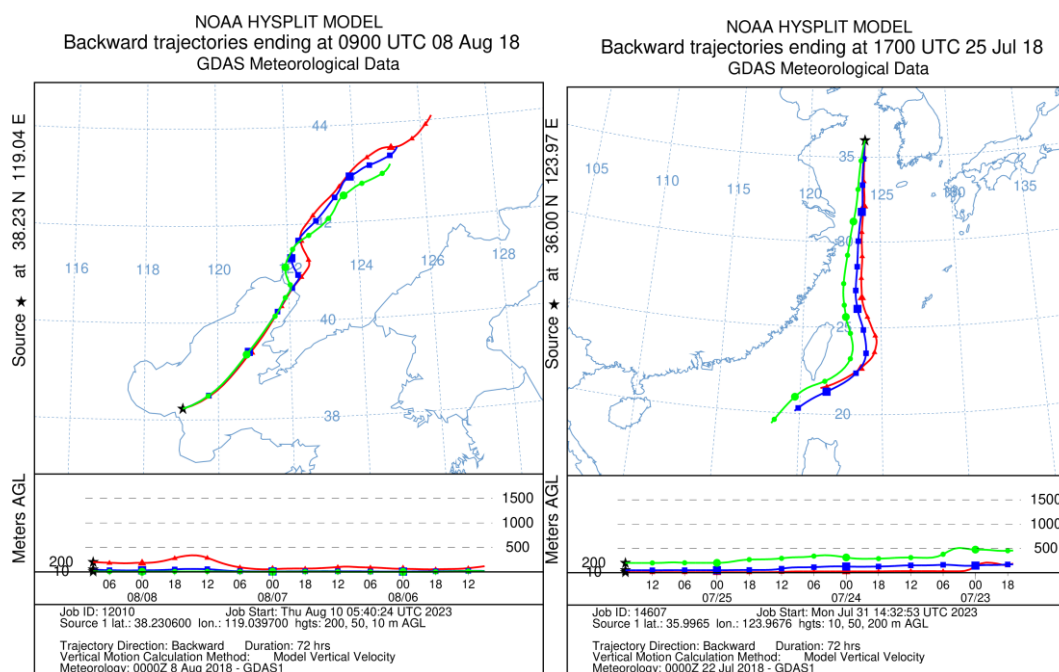
B08-spring

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.

Section 3.4 and supplemental tables – There is no good explanation in the subsequent discussion (section 4) about why the correlations between the different factors change so much, especially between variables such as COS and DOC in seawater.

Response: The original first sentence in section 3.4 is wrong according to table S4, and it has been changed into “A positive correlation occurred between the COS and DOC concentrations in seawater ($P < 0.05$) and between the CS₂ and Chl a concentrations in seawater ($P < 0.05$) during summer (Table 1).”. The discussion about COS and DOC in seawater has been added in the second paragraph of section 4.1.1.

Section 4.1.1 – This seems like a random assortment of statements. What are the main ideas of each paragraph? I had a hard time finding the clear points here.

Response: The Section 4.1.1 mainly stated spatial distributions of VSCs and compared with the other sea areas and the impact factors. The first paragraph states the spatial distributions of VSCs in this study and analysis the results in others’ research. The second paragraph states the reasons, the impact factors, and production or consumption which resulted in the spatial distributions. We have modified the structure and put the photochemical mechanisms of CDOM together and delete the wordy sentence “High COS concentrations in spring may be due to the influence of the sediment input from the Yellow River into the BS, which was more turbid and not conducive to the photochemical production of COS.”. The third paragraph stating seasonal variations of VSCs has been moved to section “4.1.3 Seasonal and diurnal variations of VSCs in

seawater”. Discussions about seasonal and diurnal variations of VSCs have been added according to the advices of the other referee.

Section 4.1.2 – I again do not understand the point of this section. What is new? The information cited is very old. Yes, COS and CS₂ processes depend on light. What is added here? Also, the statements at the end of the paragraph about sulfur in the deeper sea cannot be substantiated, as no dissolved oxygen measurements are presented. Finally, the Lennartz et al. ESSD database paper is cited, but was it used in any way to put the measurements in some context? The data presented in this manuscript should also be submitted to that database. This would be a wonderful way to use this data (for COS, CS₂, air and water). There was a follow-on paper in ESSD (Lennartz et al., 2021) that looked more deeply into modelling gas exchange and a separate Lennartz et al. (2019) publication on oceanic processes. These might be useful to consider as well.

Response: The point of section 4.1.2 is the depth distributions characters and their impact factors and reasons.

The vertical distributions presented the character at 35 °N transect. Vertical distributions were related to the solar radiation. Unfortunately, CDOM and solar radiation were not measured in this study, we will set up these parameters in the future research to confirm the distribution driving factors.

Yes, the cited information is old, the citation in the last two sentences were deleted and some new information were cited.

“The addition of photosensitizers-natural dissolved organic matter (DOM) and commercial humic acid (HA) photo-catalyzed glutathione (GSH) and cysteine, and enhanced the COS formation (Flöck et al., 1997). An excited triplet state CDOM (³CDOM*) is produced by COS in the presence of ultraviolet light (Li et al., 2022).” has been added after “The high COS concentrations in the surface seawater in spring in this study may be attributed to the photochemical production reactions of CS₂ and COS in the euphotic zone because they are dependent on light (Flöck et al., 1997; Xie et al., 1998).”.

The statements at the end of the paragraph about sulfur in the deeper sea (“It has been shown that CS₂ can be produced by anaerobic fermentation by bacteria and by reactions between H₂S and organic matter in pore water (and anoxic basins) (Andreae, 1986). This hypothesis agreed with the results of Wakeham et al. (1987), where the concentration of CS₂ peaked (at about 20 nmol L⁻¹) near the sediment-water interface. Jørgensen and Okholm-Hansen (1985) found that the release rate of VSCs (such as CS₂) in surface seawater was usually 10 to 100 times lower than that in underlying sediments in a Danish estuary, indicating that release from sediments is an important source of CS₂.”) have been deleted. The sentences of “Consistent with our CS₂ results, Xie et al. (1998) showed that the CS₂ concentrations decreased with the depth, coinciding with solar radiation changes. Decreased photochemical reaction due to decreasing solar radiation with water depth may explain the vertical distribution of CS₂ (Xie et al., 1998). Similar to the results of Xie et al. (1998), the high CS₂ concentrations in the bottom seawater at station H15 in spring may be attributable to a sedimentary source.” have been added.

Global sea-air fluxes have been added in Section 4.3. “The model of Lennartz et al. (2021) was not used to evaluate the global sea-air fluxes of DMS, OCS, CS₂ 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 CS₂ were evaluated by the mean sea-air fluxes of OCS or CS₂ multiplied by the ocean area and the time. The global sea-air fluxes of DMS, OCS, and CS₂ 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 CS₂ 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 CS₂. The another reason may be the high sea-air fluxes of OCS or CS₂ in the BS and YS because marginal seas are significantly influenced by anthropogenic emissions (Watts, 2000). The sea-air fluxes of DMS, OCS, and CS₂ 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.”

Section 4.2 – Every possible explanation is given for the atmospheric distributions. Again, what are the findings here and the main idea of each paragraph? The discussion of the DMS values in the air need more explanation (especially related to the anthropogenic source). First of all, the atmospheric lifetime of DMS is on the order of 1 day. Therefore, 72-hour back trajectories are not appropriate. If there is a relevant anthropogenic DMS source, it needs to be stated and cited.

Response: The first paragraph discussed the results of VSCs mixing ratios in this study and previous studies. The new reference (Xu et al., 2023) has been added. The second paragraph discussed the differences in the atmospheric VSC mixing ratios between spring and summer and the reasons, i.e., anthropogenic VSCs emissions and VSCs concentrations in seawater. The third paragraph discussed the mechanisms, resources, or reasons of several high VSCs mixing ratios. The fourth paragraph discussed the wind direction and air masses of the back trajectories of several stations to find the sources or reasons of the high or low VSCs mixing ratios. The wind direction of air mass and the back trajectories of Miyakojima, Yokohama, and Otaru in Japan in winter of Hattori et al. (2020) have been discussed.

Explanation about the discussion of the DMS values in the air (especially related to the anthropogenic source) has been added. 72-hour back trajectories mean trajectories from 72 h to 0 h before sampling, therefore, which include 24 h~0 h (1/3 of the line near the sampling station).

The anthropogenic DMS source has been stated and cited “The wind direction is from continental Asia to the Pacific in spring. The backward trajectories of B49, B47, and B08 showed that anthropogenic and oceanic DMS emissions accounted for the atmospheric DMS sources. The wind direction of the air mass from the back trajectories of Miyakojima, Yokohama, and Otaru in Japan in winter (January to March) observed

by Hattori et al. (2020) was similar to ours in spring (March to April). Hattori et al. (2020) reported that the anthropogenic COS originated primarily from the Chinese industry and was transported by air to southern Japan. The backward trajectory of H09 showed that the wind direction changed from the southeast in summer, and oceanic sources accounted for the atmospheric DMS.”.

Supplemental material – The figures are cited out of order in the main text. Using a compromise to provide the same scale for the two plots in figure S1 might make the information more attainable. Table S2 should have references to the work providing the constants. Why are tables S3 and S4 in the supplements and not the main text? They seem like key components of the discussion.

Response: The same scales have been used in the nutrient figures in Fig. S2.

The reference (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).) for the constants in Table S2 are provided. Tables S3 and S4 in the supplements have been merged into Table 1 and moved to the main text.

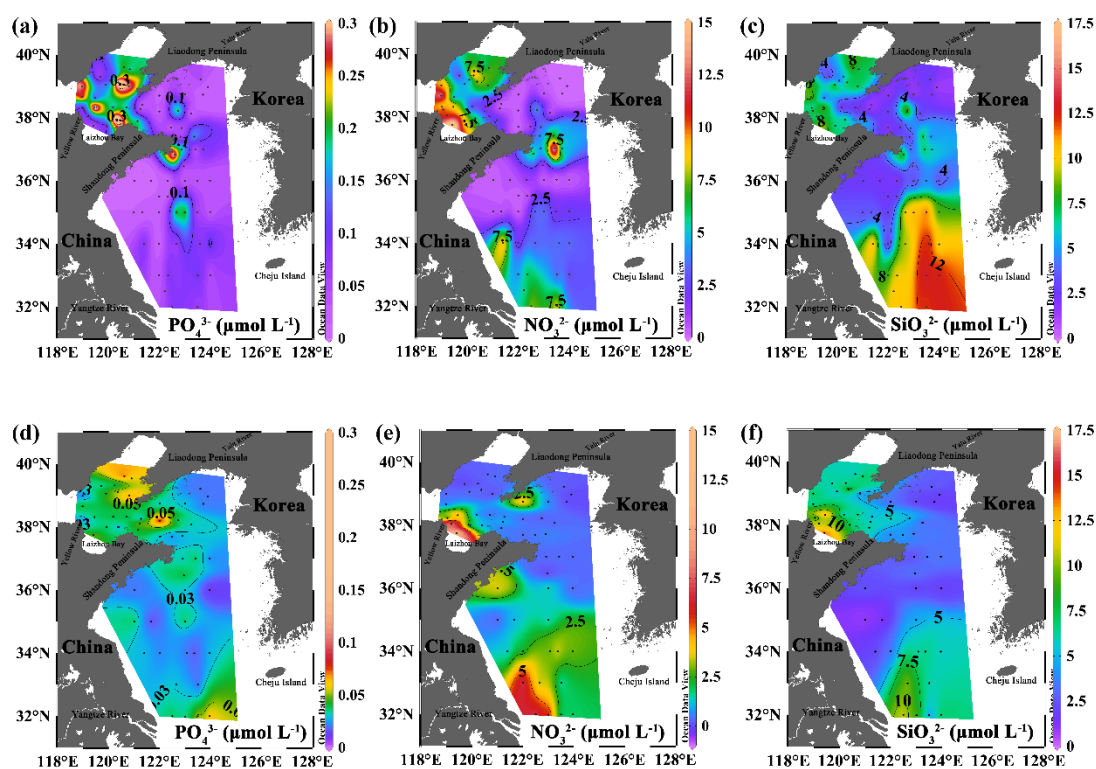


Figure S2. Spatial distributions of nitrate, phosphate, and silicate in the surface water of the BS and YS in spring (a)-(c) and summer (d)-(f).

Table 1 Correlation analyses of the three VSCs and environmental factors in the BS and YS in spring and summer.

Spring	COS (seawater)	DMS (seawater)	CS ₂ (seawater)	COS (atmosphere)	DMS (atmosphere)	CS ₂ (atmosphere)
COS (seawater)	1					
DMS (seawater)	0.021	1				
CS ₂ (seawater)	0.193	0.281*	1			
COS (atmosphere)	-0.246	-0.355	-0.182	1		
DMS (atmosphere)	0.296	0.04	0.274	0.117	1	
CS ₂ (atmosphere)	-0.201	-0.264	-0.213	0.554**	-0.013	1
Chl <i>a</i>	0.132	0.044	-0.095	0.033	0.179	-0.141
Temperature	0.286*	0.082	0.319**	-0.257	0.179	-0.372
Salinity	0.11	-0.009	-0.109	0.24	0.019	0.236
Silicate	-0.103	-0.252*	-0.029	0.351	-0.008	0.54
Phosphate	-0.084	-0.205	-0.353**	0.621	-0.128	0.36
Nitrate	-0.299*	-0.293*	-0.226	0.075	-0.096	0.044
DOC	-0.146	-0.153	-0.073	0.037	-0.122	0.008
Summer	COS (seawater)	DMS (seawater)	CS ₂ (seawater)	COS (atmosphere)	DMS (atmosphere)	CS ₂ (atmosphere)
COS (seawater)	1					
DMS (seawater)	0.009	1				
CS ₂ (seawater)	-0.007	0.424**	1			
COS (atmosphere)	0.358	0.472	0.184	1		
DMS (atmosphere)	-0.266	0.404	0.31	0.451	1	
CS ₂ (atmosphere)	0.452	0.229	0.424	0.855**	0.251	1
Chl <i>a</i>	-0.059	0.25	0.274*	0.461	-0.294	0.565
Temperature	0.088	-0.076	-0.143	-0.097	-0.349	0.072
Salinity	0.128	-0.172	-0.143	-0.12	-0.352	-0.044
Silicate	0.114	0.122	0.276*	0.312	-0.548	0.377
Phosphate	0.104	-0.169	-0.245	-0.49	-0.539	-0.482
Nitrate	-0.095	0.145	0.057	-0.008	0.224	-0.155
DOC	0.342*	-0.015	0.012	0.02	0.924	0.319

* indicates $P < 0.05$, ** indicates $P < 0.01$.

CERTIFICATE OF LANGUAGE EDITING

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

Manuscript Information

ID	LE202308120181
Editing date	2023-08-15
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	<input type="checkbox"/> Very poor <input type="checkbox"/> Poor <input type="checkbox"/> Fair <input checked="" type="checkbox"/> Good <input type="checkbox"/> Very good <input type="checkbox"/> Excellent
Recommendation after language editing	<input type="checkbox"/> Submitting to target journal directly <input checked="" type="checkbox"/> Submitting to target journal after minor revision <input type="checkbox"/> Re-editing required after major revision <input type="checkbox"/> 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.

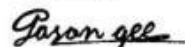
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