An itemized response (blue words) to reviewers' comments and suggestions

We thank the reviewer for their constructive comments on our manuscript "Influence of oceanic ventilation and terrestrial transport on the atmospheric volatile chlorinated hydrocarbons over the Western Pacific" [EGUSPHERE-2025-251]. These suggestions are valuable for improving the paper and guiding our future work. In response, we have made every effort to revise and enhance the manuscript, with all changes highlighted in blue. The main corrections and our detailed responses are provided below.

General comments

Ocean plays an important role on the biogeochemical cycle of volatile chlorinated hydrocarbons. However, due to scarce data, our understanding is limited about Western Pacific. Quantitative analysis of marine CCl_4 emission or sink help us to narrow or even close the gap of CCl_4, thus the topic of this paper is very important.

This manuscript presents an interesting data set of measurements of $CHCl_3$, C_2HCl_3, CCl_4 and CH_3CCl_3 from both atmosphere and sea water, and present sea-to-air fluxes in globally important region where in addition very little data has been published previously. The manuscript is well structured and generally appropriately written. However, there are a number of problems mainly connected to the data analysis and interpretation that should be addressed first.

Reply: Thank you for your positive assessment of the importance of our study and the value of the data presented. We truly appreciate the constructive comments provided, which have greatly helped improve the quality of our manuscript. Revisions have been made accordingly, with all changes highlighted in blue in the revised manuscript. Below is our point-by-point response.

1. **First of all,** the variation of atmospheric CCl_4 is with 10% and the atmospheric concentration of CH_3CCl_3 and C_2HCl_3 in only several pptv, so that the quality of sample analysis is essential for this study. The authors presented precision for CCl_4 as 4% and precision of C_2HCl_3 is 3% in the supplement material. Generally, the precision will be much worse if the concentration is smaller. In this study, the concentration of C_2HCl_3 is almost two orders of magnitude lower compared to CCl_4 concentration. I doubt why the precision of C_2HCl_3 are even better than CCl_4. Can the author provide more detailed information how these precisions were achieved?

Reply: We sincerely appreciate the reviewer's careful assessment and insightful comments.

(1) Enhancement of precision evaluation

During the experimental stage, the precision of the target compounds was evaluated using two approaches (a uniform halocarbon concentration and ambient concentration levels). In the original manuscript, considering that most VCHCs occur at relatively low ambient concentrations, a uniform low concentration (5 pptv, prepared by dynamically diluting a 100 ppbv VCHCs standard gas with the Nutech 2202A system) was selected to characterize measurement precision. This approach led to the situation where the reported precision of CCl_4 did not fully correspond to its higher ambient concentration (approximately 80 pptv). However, this discrepancy does not affect data quality, and the ambient concentrations reported in this study remain reliable.

Following the reviewer's suggestion, to more accurately reflect method performance at environmentally relevant levels, we have included precision results for the target compounds at ambient concentrations in the revised manuscript. The updated Table S1 presents these data in full, confirming that at ambient levels the precision of C_2HCl_3 is indeed lower than that of CCl_4, consistent with the reviewer's expectation. In addition, detailed descriptions of the precision testing and calculation methods have been added to the Supplementary Materials. **We wish to emphasize that this correction pertains solely to the presentation of values in Table S1; it does not affect any original sample measurements or the overarching conclusions of our study.**

The updated Table S1 is provided below:

Table S1 The method detection limits (MDL), measurement precision, and atmospheric lifetimes of the selected VCHCs in air.

Compound	**Typical ambient concentration**	**Precision at ambient-relevant concentration**[1]	**MDL**[2]	**Lifetime** [3]
	pptv	**%RSD, n=7**	**pptv**	
$CHCl_3$	10-15	3.2	0.50	178 days
C_2HCl_3	2-5	4.9	0.10	5.6 days
CH_3CCl_3	2-3	6.3	0.20	5 years
CCl_4	80-100	1.1	1.00	30 years

Notes: [1] Precision at ambient-relevant concentrations is expressed as RSD (%), based on seven replicate measurements of mixed standard gases: 2 pptv (C_2HCl_3, CH_3CCl_3), 10 pptv ($CHCl_3$), and 100 pptv (CCl_4).

[2] MDL refers to the method detection limit as determined in accordance with the US EPA procedure (2019).

[3] Atmospheric lifetimes are taken from WMO (2022).

We have added a detailed description of the precision calculation and measurement procedures in the supplementary information, as follows:

"**Text S3 Evaluation indexes in atmospheric VCHCs measurements**

Precision was evaluated in accordance with US EPA (2019) guidelines by conducting seven replicate analyses of standard gas samples prepared at environmentally relevant concentrations. These concentrations reflect typical atmospheric levels of the target VCHCs ($CHCl_3$, C_2HCl_3, CH_3CCl_3, and CCl_4), as referenced in WMO (2022). The test standards were generated by dynamically diluting a 100 ppbv primary mixture (Spectra Gases, USA) with ultra-high-purity nitrogen using a dynamic dilution system (Nutech 2202A). Under consistent analytical conditions, each standard gas sample was measured seven times. Precision was expressed as the relative standard deviation (RSD, %) calculated using Eq. (1).

$$\mathrm{Precision}=\mathrm{RSD}_{(X)}=\frac{\sqrt{\frac{\sum_{i=1}^{n}(X_{(i)}-\bar{X})^2}{n-1}}}{\bar{X}}\times 100\% \qquad (1)$$

where $X_{(i)}$ denotes the measured concentration of the target compound in the sample gas derived from a multipoint external calibration curve. $\bar{X}$ is the arithmetic mean of seven replicate measurements, and n=7. The precision results at ambient-relevant concentrations are summarized in Table S1." **(supplementary information Texts S3)**

(2) Supplementary improvements in the methods section:

To enhance the transparency of this study, more detailed descriptions of the experimental procedures have been added in the revised manuscript. In addition, chromatograms of VCHCs at ambient concentration levels (Figures A and B) and a schematic diagram of the sample analysis workflow (Figure C) are provided to clearly demonstrate the applicability and operability of the method. Furthermore, the analytical approach employed in this study is a well-established and reliable technique and widely applied in numerous peer-reviewed studies (Li et al., 1999; Kurihara et al., 2010; Liu et al., 2011; Yokouchi et al., 2013; Cao et al., 2023).

We have added detailed methodological descriptions to the "2.2 Analysis of VCHCs in air" section in the revised manuscript, as follows:

"2.2 Analysis of VCHCs in air

Atmospheric samples were collected by 3 L pre-evacuated stainless-steel Summa polished canisters (SilconCan, Restek Co., Ltd) that were pre-cleaned to measure VCHCs concentrations at ambient pressure. These sampling canisters underwent an intensive cleaning process using an automated cleaning system (Nutech 2010 DS) prior to sample collection. Previous studies have shown that VCHCs remain stable in rigorously pre-cleaned Summa canisters for more than 3 months (Yokouchi et al., 1999; Yokouchi et al., 2013; Yu et al., 2020). To avoid ship exhaust contamination, sampling was conducted upwind on the ship's top deck during low-speed transit. All atmospheric samples were analyzed within 3 months after the collection. Meteorological parameters such as wind speed and direction were recorded by shipboard sensors at a height of 10 m above the sea surface.

The atmospheric concentrations of VCHCs were determined using a three-stage cold-trap preconcentrator (Nutech 8900DS) coupled with a gas chromatography–mass spectrometry (GC–MS) system (Agilent 7890A/5975C). Chromatographic separation was achieved on a DB-624 capillary column (60 m × 0.25 mm × 1.4 µm film thickness) in selective ion monitoring (SIM) mode. Prior to injection, samples (400 mL) were preconcentrated using the three-stage cold-trap system, effectively removing interfering components such as H_2O and CO_2. Target compounds were quantified using a multipoint external calibration method. Calibration curves were established with a 100 ppbv mixed standard gas (Spectra Gases, USA), which was dynamically diluted with ultra-high-purity nitrogen using a mass-flow–controlled dilution system (Nutech 2202A; accuracy ±1%) to achieve ppt–low ppbv concentration levels. Six concentration gradients were prepared, and the diluted standards were analyzed following the same procedures as the field samples. The calibration results showed correlation coefficients for all target compounds are ≥ 0.996. According to the US EPA (2019) procedure, the method detection limits (MDL) of the target compounds ranged from 0.10 to 1.0 pptv. MDL is defined as 3.143 × standard deviation of seven replicates of the low concentration standard gases (5× the expected MDL), where 3.143 represents the t-value at 99% confidence level. Precision in this study was assessed from seven replicate measurements of standard gas samples prepared at environmentally relevant concentrations, with relative standard deviations (RSD) consistently below 7% for all target compounds (Table S1). This method has been validated through comparisons with the China Meteorological Administration Meteorological Observation Centre for independent analysis using an AGAGE-traceable Medusa-GC/MS system (Zhang et al., 2017; Yu et al., 2020; An et al., 2021). Furthermore, it is consistent with previously published methodologies (Zhang et al., 2010; Zheng et al., 2013; Yi et al., 2021; Li et al., 2024). Detailed analytical procedures and data processing methods are provided in the supplementary information (Texts S2 and S3)." **(Lines 197-237)**

We have provided representative GC–MS chromatograms (Fig. A, B), which show clear peak separation and ambient-level detection of all target VCHCs, as follows:

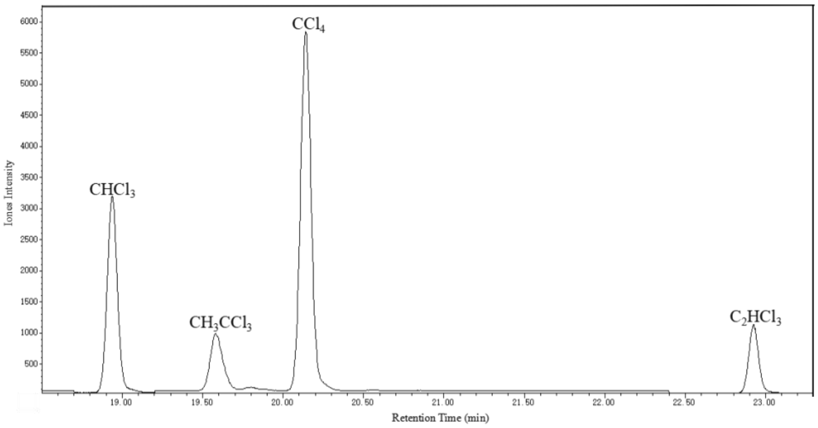


Fig. A. Chromatogram from an ambient air sample obtained. X axis: retention time (min). Y axis: ion intensity value without unit.

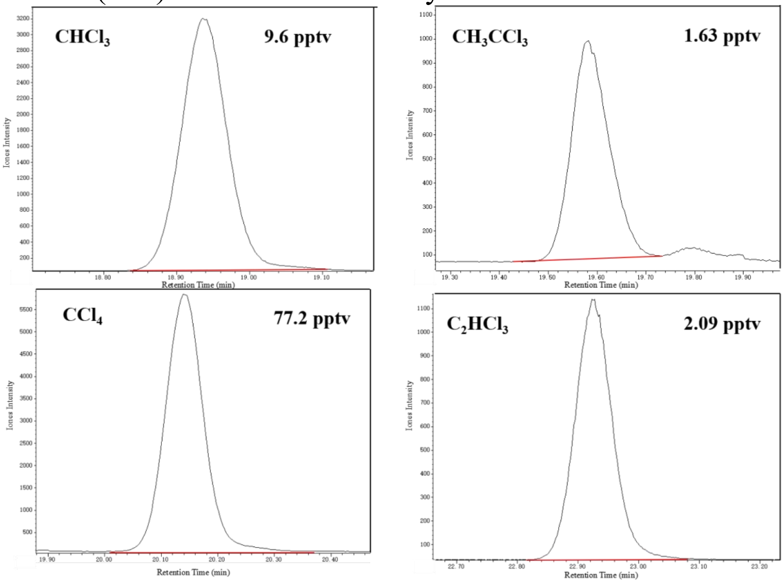
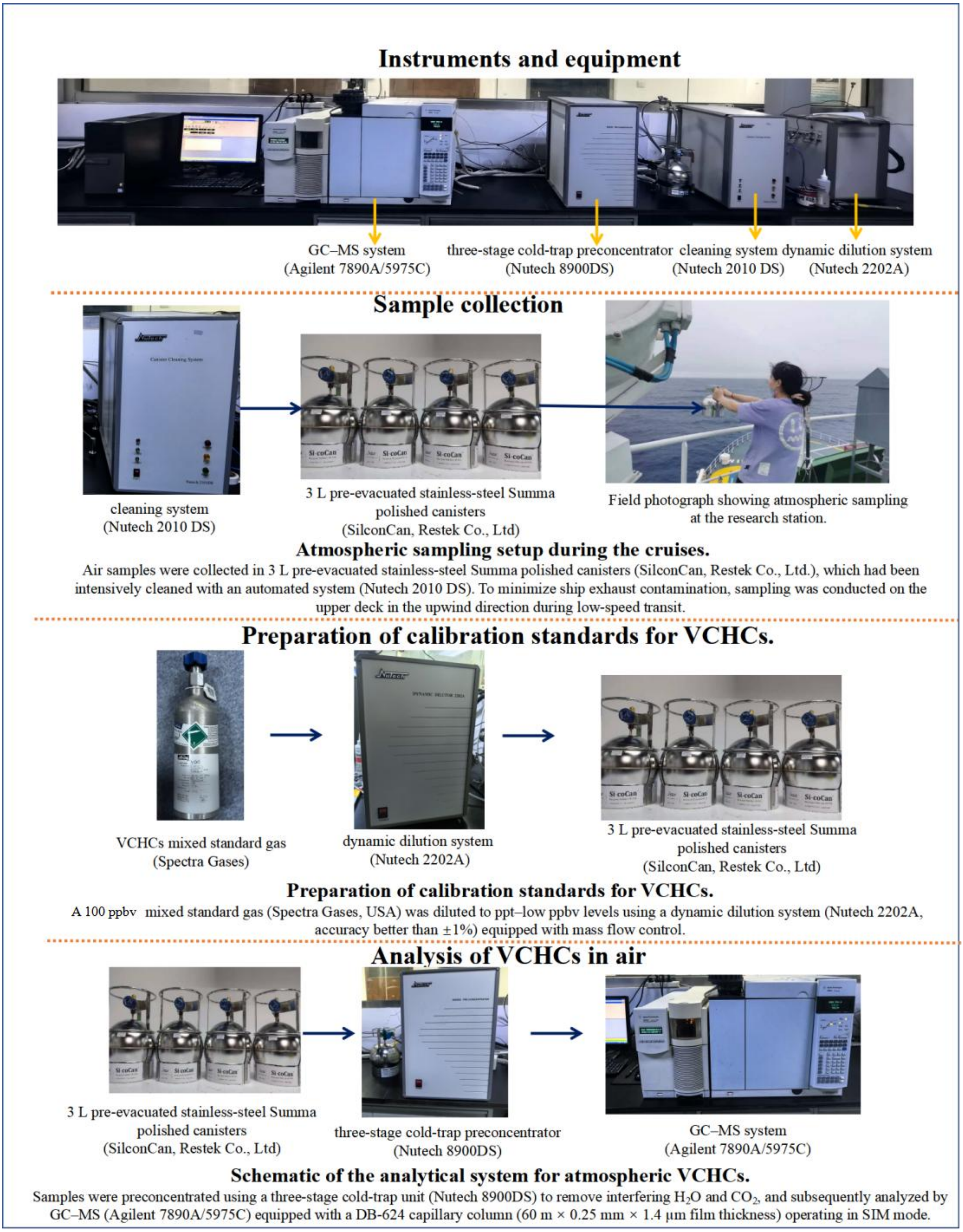


Fig. B. Chromatograms of VCHCs with ambient mole fractions.

A schematic diagram of the sample analysis workflow, as follows:



Air samples were collected in 3 L pre-evacuated stainless-steel Summa polished canisters (SilconCan, Restek Co., Ltd.), which had been intensively cleaned with an automated system (Nutech 2010 DS). To minimize ship exhaust contamination, sampling was conducted on the upper deck in the upwind direction during low-speed transit.

A 100 ppbv mixed standard gas (Spectra Gases, USA) was diluted to ppt–low ppbv levels using a dynamic dilution system (Nutech 2202A, accuracy better than ±1%) equipped with mass flow control.

Samples were preconcentrated using a three-stage cold-trap unit (Nutech 8900DS) to remove interfering H_2O and CO_2, and subsequently analyzed by GC–MS (Agilent 7890A/5975C) equipped with a DB-624 capillary column (60 m × 0.25 mm × 1.4 μm film thickness) operating in SIM mode.

Fig. C Sample-analysis workflow diagram.

The related literatures are listed below:

An, M., Western, L. M., Say, D., Chen, L., Claxton, T., Ganesan, A. L., et al. (2021). Rapid increase in dichloromethane emissions from China inferred through atmospheric observations. Nature Communications, 12(1), 7279.

Cao, X., Gu, D., Li, X., Leung, K. F., Sun, H., Mai, Y., et al. (2023). Characteristics and source origin analysis of halogenated hydrocarbons in Hong Kong. Science of the Total Environment, 862, 160504.

Kurihara, M. K., Kimura, M., Iwamoto, Y., Narita, Y., Oohi, A., et al. (2010). Distributions of short-lived iodocarbons and biogenic trace gases in the Open Ocean and atmosphere in the western North Pacific. Marine Chemistry, 118 (3-4), 156-170.

Liu, Y. N., Yvon-Levis, S. A., Hu, L., Salisbury, J. E., O'Hern, J. E. (2011). $CHBr_3$, CH_2Br_2, and $CHClBr_2$ in the U.S. coastal waters during the Gulf of Mexico and East Coast Carbon cruise. Journal of Geophysical Research: Oceans, 116, 1440-1450.

Li, B., Huang, J., Hu, X., Zhang, L., Ma, M., Hu, L., et al. (2024). CCl_4 emissions in eastern China during 2021–2022 and exploration of potential new sources. Nature Communications, 15(1), 1725.

Li, H. J., Yokouchi, Y., Akimoto, H. (1999). Measurement of methyl halides in the marine atmosphere. Atmospheric Environment. 33.12: 1881-1887.

Miller, B. R., Weiss, R. F., Salameh, P. K., Tanhua, T., Greally, B. R., Mühle, J., & Simmonds, P. G. (2008). Medusa: A sample preconcentration and GC/MS detector system for in situ measurements of atmospheric trace halocarbons, hydrocarbons, and sulfur compounds. Analytical Chemistry, 80(5), 1536-1545.

Prinn, R. G., Weiss, R. F., Fraser, P. J., Simmonds, P. G., Cunnold, D. M., Alyea, F. N., et al. (2000). A history of chemically and radiatively important gases in air deduced from ALE/GAGE/AGAGE. Journal of Geophysical Research: Atmospheres, 105(D14), 17751-17792.

Yokouchi, Y., Li, H. J., Machida, T., Aoki, S., & Akimoto, H. (1999). Isoprene in the marine boundary layer (Southeast Asian Sea, eastern Indian Ocean, and Southern Ocean): Comparison with dimethyl sulfide and bromoform. Journal of Geophysical Research: Atmospheres, 104(D7), 8067-8076.

Yokouchi, Y., Inoue, J., & Toom-Sauntry, D. 2013. Distribution of natural halocarbons in marine boundary air over the Arctic Ocean. Geophysical Research Letters, 40, 4086–4091.

Yu, D., Yao, B., Lin, W., Vollmer, M. K, Ge, B., Zhang, G., et al. (2020). Atmospheric CH_3CCl_3 observations in China: historical trends and implications. Atmospheric Research, 231, 104658.

Zhang, G., Yao, B., Vollmer, M. K., Montzka, S. A., Mühle, J., Weiss, R. F., et al. (2017). Ambient mixing ratios of atmospheric halogenated compounds at five background stations in China. Atmospheric Environment, 160, 55–69.

2. For the second, the calibration method described in lines 199-204. How much uncertainties introduced by the dilution? For VCHCs at pptv level, normally calibration scales are applied in calibration to minimize the inconsistent between standards. I also doubt the comparison between this study and AGAGE background is misleading due to the calibration method.

Reply: We thank the reviewer for the insightful comments on calibration uncertainties and the comparability of our data with AGAGE. We have added detailed information demonstrating the reliability of our dilution and calibration procedures, including internal validation and external verification by the China Meteorological Administration laboratory. Moreover, the methodology employed in this study is a

well-established technique that has been widely applied in comparisons with AGAGE data. Therefore, we are confident that the comparisons with AGAGE background data presented in this study are both reliable and valid. Our detailed responses are provided below:

Methodology validation for VCHCs measurements:

(1) Dilution independent verification:

To ensure the accuracy of our calibration procedures, we employed a two-tiered validation approach:

(a) Primary calibration: In this study, a 100 ppbv VCHCs standard gas (Spectra Gases, USA) was dynamically diluted to pptv levels using a mass-flow–controlled dilution system (Nutech 2202A; accuracy ±1%). Six concentration gradients were prepared and analyzed using the same procedures as the field samples. The results showed that the correlation coefficients of the calibration curves for all target compounds are $\geq$ 0.996, indicating excellent linearity and methodological reliability.

(b) Secondary verification: More importantly, to ensure calibration accuracy and international comparability, aliquots of the diluted standard gases were sent to theChina Meteorological Administration Meteorological Observation Centre (CMA-MOC) for independent blind analysis using an AGAGE-traceable Medusa-GC/MS system (Zhang et al., 2017; Yu et al., 2020; An et al., 2021). The results showed the differences between our measurements and CMA-MOC analyses within ±5%, confirming the reliability of our dilution and calibration procedures.

(2) Robustness and comparability with AGAGE data:

The methodology adopted in this study—using a commercial standard gas with dynamic dilution and multipoint calibration—is a well-established and reliable approach for ppt-level VCHCs measurements. It has been widely validated and applied in numerous studies directly comparing with AGAGE background data (e.g., Zhang et al., 2010; Zheng et al., 2013; Yi et al., 2021; Li et al., 2024).

To ensure calibration accuracy and international comparability, aliquots of the diluted standard gases were sent to the CMA-MOC for independent analysis using an AGAGE-traceable Medusa-GC/MS system (Zhang et al., 2017; Yu et al., 2020; An et al., 2021). The CMA-MOC results were reported as dry-air mole fractions on calibration scales established and maintained by the Scripps Institution of Oceanography (SIO) (Prinn et al., 2000; Miller et al., 2008). The intercomparison indicated that the differences between the measurements and those from CMA-MOC for target compounds were within ±5%: $CHCl_3$ (−4.3% to −1.2%), CH_3CCl_3 (+1.5% to +4.6%), and CCl_4 (+1.1% to +3.8%). The independent intercomparison with the CMA laboratory further demonstrated that the concentration differences are small.

These results reinforce the methodological robustness of our approach, ensuring that comparisons with AGAGE background data are both technically sound and scientifically valid.

(3) Supplementary revisions:

In the revised manuscript, we have added the related content of the dilution in the Supplementary Information (Text S2), as follows:

"Text S2 Accuracy of the dynamic dilution system

Calibration curves were established using a 100 ppbv standard mixture gas (Spectra Gases, USA), which was dynamically diluted with ultra-high-purity nitrogen via a mass-flow–controlled dilution system (Nutech 2202A; accuracy ±1%) to achieve ppt–low ppbv concentration levels. Six concentration gradients were prepared, and the diluted standards were analyzed following the same procedures as the field samples. The results showed that the correlation coefficients of the calibration curves for all target compounds are ≥ 0.996, indicating excellent linearity and methodological reliability.” **(supplementary information Texts S2)**

The related literatures are listed below:

An, M., Western, L. M., Say, D., Chen, L., Claxton, T., Ganesan, A. L., et al. (2021). Rapid increase in dichloromethane emissions from China inferred through atmospheric observations. Nature Communications, 12(1), 7279.

Li, B., Huang, J., Hu, X., Zhang, L., Ma, M., Hu, L., et al. (2024). CCl_4 emissions in eastern China during 2021–2022 and exploration of potential new sources. Nature Communications, 15(1), 1725.

Miller, B. R., Weiss, R. F., Salameh, P. K., Tanhua, T., Greally, B. R., Mühle, J., & Simmonds, P. G. (2008). Medusa: A sample preconcentration and GC/MS detector system for in situ measurements of atmospheric trace halocarbons, hydrocarbons, and sulfur compounds. Analytical Chemistry, 80(5), 1536-1545.

Prinn, R. G., Weiss, R. F., Fraser, P. J., Simmonds, P. G., Cunnold, D. M., Alyea, F. N., et al. (2000). A history of chemically and radiatively important gases in air deduced from ALE/GAGE/AGAGE. Journal of Geophysical Research: Atmospheres, 105(D14), 17751-17792.

Yi, L., Wu, J., An, M., Xu, W., Fang, X., Yao, B., et al. (2021). The atmospheric concentrations and emissions of major halocarbons in China during 2009–2019. Environmental Pollution, 284, 117190.

Yu, D., Yao, B., Lin, W., Vollmer, M. K., Ge, B., Zhang, G., et al. (2020). Atmospheric CH_3CCl_3 observations in China: historical trends and implications. Atmospheric Research, 231, 104658.

Zheng, J., Yu, Y., Mo, Z., Zhang, Z., Wang, X., Yin, S., et al. (2013). Industrial sector-based volatile organic compound (VOC) source profiles measured in manufacturing facilities in the Pearl River Delta, China. Science of the Total Environment, 456, 127-136.

Zhang, Y. L., Guo, H., Wang, X.M., Simpson, I. J., Barletta, B., Blake, D. R., et al. (2010). Emission patterns and spatiotemporal variations of halocarbons in the Pearl River Delta region, southern China. Journal of Geophysical Research: Atmospheres, 115(D15).

Zhang, G., Yao, B., Vollmer, M. K., Montzka, S. A., Mühle, J., Weiss, R. F., et al. (2017). Ambient mixing ratios of atmospheric halogenated compounds at five background stations in China. Atmospheric Environment, 160, 55–69.

3. **For the third,** the variation of atmospheric CCl_4 in this study is similar to its analysis precision. So the authors need to prove the changes of atmospheric CCl_4 is not caused by the measurement uncertainties and be aware not to over-interpretation the concentration differences.

Reply: We thank the reviewer for emphasizing the importance of distinguishing real atmospheric signals from analytical uncertainties. We fully agree that this consideration is critical, particularly when the observed variations in trace gas concentrations are close to the analytical precision of the method. It is equally essential to demonstrate that the observed changes in the atmospheric concentration of CCl_4 are not driven by measurement uncertainties. In accordance with the reviewers' suggestions, we have systematically refined our interpretation of concentration variations through the following improvements to avoid over-explaining the differences in concentration: (1) CCl_4 differences exceeding the 3σ analytical precision threshold (3.3%, Table S1) are considered reliable, therefore, only variations greater than 4% above the regional mean were retained for discussion in this study; (2) the observed enhancements show clear spatial consistency, systematically concentrated at nearshore stations influenced by continental air masses rather than random noise; and

(3) these enhancements coincide with elevated concentrations of SF_6, an independent anthropogenic tracer (Ni et al., 2023). Taken together, these multiple lines of evidence indicate that the CCl_4 features reported in the revised manuscript represent real atmospheric signals.

In addition, we have carefully interpreted the concentration differences in the revised manuscript to avoid overstatement.

The corresponding revisions in the revised manuscript are as follows:

"Figure 3 shows the spatial distributions of atmospheric mixing ratios of CCl_4 and CH_3CCl_3. Significant enhancements were observed at nearshore stations (e.g., P1-4, P1-5, P1-7, P1-8 and EQ12). CCl_4 concentrations in these stations were 4–6% higher than the regional average, exceeding its analytical precision threshold of 3σ (3.3%, Table S1). These enhancements exhibited clear spatial consistency, systematically concentrated in nearshore areas influenced by continental air masses (Fig. 1 and Fig. 3). Moreover, the elevated levels coincided with enhanced concentrations of the independent anthropogenic tracer SF_6 (Ni et al., 2023), further corroborating their origin from continental pollution outflows. This result is also consistent with previous studies reporting that elevated CCl_4 and CH_3CCl_3 levels are primarily concentrated in coastal regions (Blake et al., 2003; Zhang et al., 2010)." **(Lines 365-376)**

The related literature is listed below:

Blake, N. J., Blake, D. R., Simpson, I. J., Meinardi, S., Swanson, A. L., Lopez, J. P., et al. (2003). NMHCs and halocarbons in Asian continental outflow during the Transport and Chemical Evolution over the Pacific (TRACE-P) Field Campaign: Comparison with PEM-West B. Journal of Geophysical Research: Atmospheres, 108(D20).

Ni, J., Liu, S. S., Lang, X. P., He, Z., & Yang, G. P. (2023). Sulfur hexafluoride in the marine atmosphere and surface seawater of the Western Pacific and Eastern Indian Ocean. Environmental Pollution, 335, 122266.

Zhang, Y. L., Guo, H., Wang, X. M., Simpson, I. J., Barletta, B., Blake, D. R., et al. (2010). Emission patterns and spatiotemporal variations of halocarbons in the Pearl River Delta region, southern China. Journal of Geophysical Research: Atmospheres, 115(D15).

4. For the fourth, in section 3.1, the authors ascribe the elevated concentrations of VCHCs to the influence of polluted air mass from mainland or east China may not be correct. From Fig. 3 and Fig. 4, the trajectories of air mass with high observed concentration are generally from Siberia and Northeast China, and then pass Japan. However, from the recent publications, these regions are not the major source of VCHCs and there is no report of fluorine/chlorine chemistry located in these regions. It should be noted most of the trajectories in figure 4 did not cover North China or East China mentioned in the references.

Reply: We thank the reviewer for the valuable comment. We acknowledge that our previous attribution of the elevated concentrations to polluted air masses from northern or eastern China was inaccurate. In the revised manuscript, we have adjusted the wording in Section 3.1 to more cautiously describe the possible transport pathways, emphasizing the potential for long-range transport and mixing with polluted air masses from industrialized regions along the route, rather than directly attributing the high concentrations to specific source areas without direct evidence.

The corresponding revisions in the revised manuscript are as follows:

"In this study, the highest atmospheric concentrations of CCl_4 and CH_3CCl_3 were recorded at station P1-4 near Japan. Backward trajectory cluster analysis (Fig. 1c and Fig. S4) indicated that approximately 13% of the air masses originated from short-range transport along the eastern coast of Japan, while the remaining 87% were associated with long-range transport from Siberia and Northeast China and subsequently passed over the eastern coast of Japan. Although Siberia and Northeast China are not typical source regions for halocarbons, previous studies have shown that air masses from these regions may mix with pollution plumes from East Asian industrial areas during long-range transport (Stohl et al., 2002; Blake et al., 2003; Liang et al., 2004; Chang et al., 2022), which could lead to elevated VCHCs concentrations at downwind observation stations. This is corroborated by reports of CCl_4 pollution events at the Shangdianzi (SDZ) regional background station in northern China (117.17°E, 40.65°N, Fig. 1a), with peak mixing ratios reaching 151 pptv (Yi et al., 2023). As anthropogenic compounds, CCl_4 and CH_3CCl_3 are primarily emitted from industrial activities, including chloromethane and perchloroethylene production (Liang et al., 2016; Sherry et al., 2018), as well as unreported releases from chlorine and bleaching processes (estimated up to 10 Gg yr^{-1}). Furthermore, Zheng et al. (2019) and Ou-Yang et al. (2017) reported elevated CCl_4 and CH_3CCl_3 levels in China and Japan, respectively. Lunt et al. (2018) identified continued CCl_4 emissions from Eastern Asia. Collectively, these findings suggested that continental air mass transport was likely the dominant factor driving the elevated CCl_4 and CH_3CCl_3 levels observed at stations P1–4 during the study period."

(Lines 377-399)

"The 96-h backward trajectory cluster analysis revealed that the KEO region is affected by air masses originating from Siberia, Northeast China, Korea, and Japan (Fig. 1c). While Siberia and Northeast China themselves are not recognized as major VCHCs source regions, the long-range air masses from these areas may have entrained polluted plumes during their transport (Section 3.1.1). Indeed, surrounding regions are known to be significant emitters: Feng et al. (2019) reported a marked rise in China's $CHCl_3$ emissions in recent years, and An et al. (2023) further showed that emissions peaked at

193 Gg yr^{-1} in 2017 before declining to 147 Gg yr^{-1} in 2018 and remaining stable thereafter, with eastern China consistently identified as a major contributor. Moreover, Ou-Yang et al. (2017) observed high atmospheric $CHCl_3$ mixing ratios (39 ± 11 pptv) at the Mt. Fuji research station, Japan, in 2015; and in 2017, the highest annual mean $CHCl_3$ (43 ± 18 pptv) was recorded at the Gosan station (GSN, 127.17°E, 33.28°N, 72 m above sea level, a regional baseline station; Fig. 1a) on Jeju Island, South Korea. Furthermore, industrial activities are known sources of $CHCl_3$ and C_2HCl_3 (Montzka et al., 2011; Oram et al., 2017; Zheng et al., 2019). Thus, the elevated $CHCl_3$ and C_2HCl_3 observed in the KEO region were likely related to terrestrial air mass transport and subsequent mixing with polluted plumes during their transit." **(Lines 428-445)**

The following related literature has been added:

Blake, N. J., Blake, D. R., Simpson, I. J., Meinardi, S., Swanson, A. L., Lopez, J. P., et al. (2003). NMHCs and halocarbons in Asian continental outflow during the Transport and Chemical Evolution over the Pacific (TRACE-P) Field Campaign: Comparison with PEM-West B. Journal of Geophysical Research: Atmospheres, 108(D20).

Chang, C. Y., Wang, J. L, Chen, Y. C., Pan, X. X., Chen, W. N., Lin, M. R., et al. (2022). A study of the vertical homogeneity of trace gases in East Asian continental outflow. Chemosphere, 297, 134165.

Liang, Q., Jaeglé, L., Jaffe, D. A., Weiss-Penzias, P., Heckman, A., & Snow, J. A. (2004). Long-range transport of Asian pollution to the northeast Pacific: Seasonal variations and transport pathways of carbon monoxide. Journal of Geophysical Research: Atmospheres, 109(D23).

Stohl, A., Eckhardt, S., Forster, C., James, P., & Spichtinger, N. (2002). On the pathways and timescales of intercontinental air pollution transport. Journal of Geophysical Research: Atmospheres, 107(D23), ACH-6.

Yi, L., An, M., Yu, H., Ma, Z., Xu, L., O'Doherty, S., et al. (2023). In Situ Observations of Halogenated Gases at the Shangdianzi Background Station and Emission Estimates for Northern China. Environmental Science & Technology, 57(18), 7217-7229.

5. Last but not least, both two surveys were conducted in autumn and winter. Concerning the seasonal variation of marine microalgae and seawater temperature, wind speed, the sea-to-air flux obtained by this study might be bias from yearly average.

Reply: We thank the reviewer for this comment. According to your suggestion, we have added the discussion of potential effects of seasonal variations in marine microalgae, seawater temperature, and wind speed on the annual flux estimates in the revised manuscript.

The corresponding revisions in the revised manuscript are as follows:

"It should be noted that the sea–air flux estimates of $CHCl_3$, C_2HCl_3, CCl_4, and CH_3CCl_3 presented in this study are derived exclusively from autumn and winter cruise observations, and thus may not fully represent their annual averages due to seasonal variability. The spatiotemporal patterns of sea–air fluxes are primarily governed by the concentrations of VCHCs in the atmosphere and seawater, SST, and wind speed. In particular, seawater concentrations of $CHCl_3$ and C_2HCl_3 are strongly modulated by biological activity (Section 3.3), while both SST and wind speed exhibit pronounced seasonal variations (Fig. 7a). Moreover, according to AGAGE data (https://agage.mit.edu/), the interannual variability of global atmospheric CCl_4 in 2019 was 2%, with variability at GSN sites of 3% (Fig. 7b), whereas global atmospheric CH_3CCl_3 varied by 13% (Fig. 7c). During the observation period of this study, mean atmospheric concentrations of CCl_4 at GSN sites and globally were 1% lower than their respective annual averages, while the global atmospheric CH_3CCl_3 concentration was

3% lower, indicating that their levels in the western Pacific were generally close to annual averages. In addition, while SST and Chl-*a* in the study region during the observation period were largely consistent with their annual averages, wind speeds were on average 10% higher (Fig. 7a; Tang et al., 2022). Consequently, the flux values reported in this study are likely somewhat higher than the annual mean, primarily due to the elevated wind speeds during the cruises.” **(Lines 683–702)**

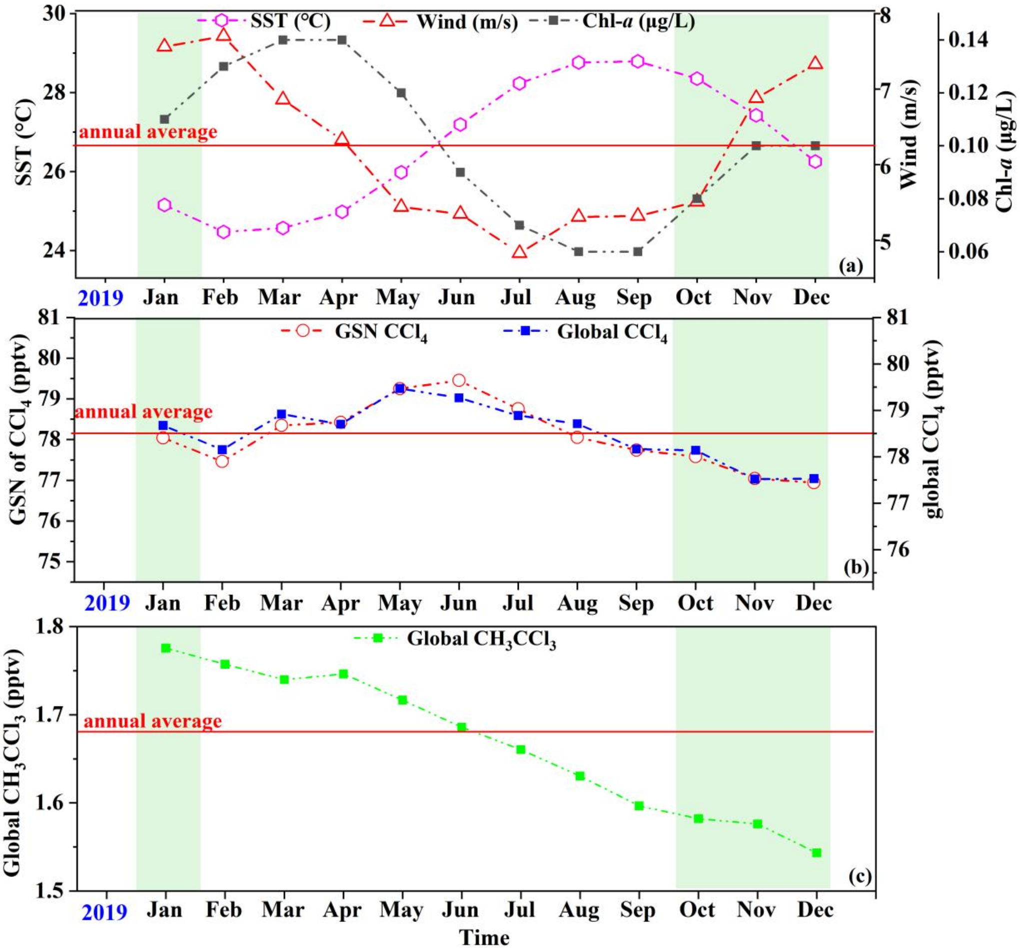


Fig. 7. (a) Interannual variations of wind speed, SST, and Chl-*a* concentration in the study area during 2019. (b) Interannual variability of CCl_4 at the GSN site in 2019 compared with the global mean. (c) Interannual variability of CH_3CCl_3 at the global scale in 2019. Wind speed data were obtained from the ERA5 reanalysis (Hersbach et al., 2020), SST data from the ECCO2 cube92 dataset (Menemenlis et al., 2008), and Chl-*a* data from the NASA Ocean Biology Processing Group (2022). CCl_4 data for the GSN site were retrieved from the AGAGE network (https://agage.mit.edu/). Shaded areas indicate the cruise sampling periods.

In short, we have carefully considered the reviewer's comments and suggestions and conducted the revision seriously. We are very thankful to the reviewer for all the valuable comments and helpful suggestions to improve this manuscript.