

Chlorinated VSLs Surpass HCFCs in CFC-11-Equivalent Emissions for Ozone Layer Depletion in China

Corresponding Author: Professor Jianxin Hu

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

Comments on “China’s Chlorinated VSLs Surpass HCFCs in CFC-11-Equivalent Emissions for Ozone Layer Depletion”. This study provides critical insights into the historical and projected emissions of dichloromethane (CH₂Cl₂) and chloroform (CHCl₃) in China. It addresses a significant gap in the current literature, which predominantly relies on top-down atmospheric inversions for chlorinated very short-lived substances (Cl-VSLs), by constructing a detailed, sector-resolved bottom-up emission inventory. However, several concerns regarding the reliability of the emission estimates should be addressed to strengthen the manuscript’s conclusions.

Major comments:

1. The representativeness of the emission factor used should be enhanced. For example, the CH₂Cl₂ emissions from adhesive utilization and coating were calculated with emission factors (EF) in 2009 and 2012, respectively. The representativeness of such old EFs is doubtful. It is recommended to use a dynamic EF, which varies with the year.
2. Please provide a deeper discussion on the persistent gap between your bottom-up CHCl₃ estimates and top-down results post-2015. Consider if underestimation of unintentional industrial by-product emissions explains this discrepancy.
3. Explicitly link your findings on by-product emissions to recent studies on unintentional ODS release from coal combustion and iron & steel production. This highlights a broader regulatory gap beyond unregulated substances.
4. Figure S1 shows that the bottom-up CHCl₃ estimations during 2015–2020 are lower compared with the top-down estimations from An et al. (2023). Please consider adding a discussion about the reason for the gap between different methods.
5. Compared with previous literature (Feng et al., 2018, Bottom-up anthropogenic dichloromethane emission estimates from China for the period 2005–2016 and predictions of future emissions., Atmos. Environ.), the ratio of production leakage to CH₂Cl₂ emissions is significantly lower in the current work (Figure 1). What factors account for this?
6. The authors should ensure key quantitative findings are consistently presented alongside their uncertainty ranges, such as the projected crossover with HCFCs and future emission levels. This is crucial for robust conclusions.
7. Please briefly justify the linear phase-out assumption in your BPT scenario (e.g., policy drafts or analogous regulations). Additionally, the authors could emphasize the climate co-benefits of clean heating policies for a more integrated argument.
8. The authors should add the South China Sea region to Figure 2 to present a complete and accurate national map, aligning with standard cartographic practice.

Minor comments:

Line 75, 76, 77, etc.: CH₂Cl₂ emission -> CH₂Cl₂ emissions.

Line 239: “We set a phase-down schedule in the three sectors that the share CH₂Cl₂-free production or product will increase linearly from 20% in 2026 to 100% in 2030.” Is there any reference for the assumption of this linear increase?

Figure 2: The map of the South China Sea is recommended to be added.

Line 196: report -> reported.

Figure S1: “emisison” -> “emissions”

Reviewer #2

(Remarks to the Author)

The authors present some work on current Chinese emissions of CH₂Cl₂ and CHCl₃, based on emission factors of various

sources over the course of 25 years (part of which are projections). Given that the increasing global emissions of CH₂Cl₂ in the atmosphere are of growing relevance for stratospheric ozone depletion, this topic is undoubtedly of importance to multiple science communities and of interest to a wide readership.

While this is a well-written manuscript, there are some concerns, the main one being about the traceability of results as well as their uncertainties: The authors have commendably gathered information on activity data and emission factors from quite a wide range of sources. Some of their sources are however inaccessible (e.g., searching for CH₂Cl₂ or dichloromethane at <https://www.stats.gov.cn/sj/> only returns the message "Sorry! Failed to find the information you searched."; other similar examples are, e.g., the China Statistical Yearbook or the China Urban-Rural Construction Statistical Yearbook). Other sources are outdated (e.g., IPCC 2000: Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories, Lobert et al., 1999), or it is unclear why only a single number (no range) was chosen for the emission factor (also Lobert et al., 1999, feedstock use leakage, water treatment, biomass burning, coal combustion,...). As a more general comment, given the continuously evolving nature of industrial processes it is unclear why a single emission factor should even be considered adequate for a) an entire branch of industry, and b) over two decades or more. Even where a range is given, the rationale for this range is not made clear in many cases. Is it, e.g., arbitrarily 10 % for feedstock use leakage/adhesive utilization/coating? Most important is however that all of the above, when considered more adequately, would lead to a massive increase of the uncertainties of all emissions derived here. This increase (which is of course also partly due to the uncertain nature of bottom-up estimates) would likely render the main messages of this manuscript void (unless the authors can demonstrate otherwise). Comparisons of such results (for other trace gases) with top-down estimates based on atmospheric observations have demonstrated many differences in the past, and in most cases the bottom-up estimates were revised later on. The fact that the An et al. top-down estimates appear to agree very well with the results of this manuscript over several years gives a little bit of confidence (but only for this period). However, the biggest opportunity to add more credibility to this manuscript would be adding more top-down-based data (which exists for both CH₂Cl₂ and CHCl₃) for a more evidence-based assessment of Chinese emissions.

Nevertheless, if these concerns as well as the comments/questions below can be addressed appropriately, the manuscript could become suitable for publication in this journal.

Firstly, the English language is quite good for most of the manuscript but might benefit from another iteration.

69 Referring to Taiwan as a part of China might be commonplace in China but could be perceived as controversial internationally. Perhaps the Editorial Office wants to check whether this is in agreement with journal policy.

84 Calling this an "annual emission inventory" seems inappropriate since part of it is in the future.

169 The sudden injection of other Cl-VSLs into the manuscript seems out of place.

238-241 What is the rationale for these numbers (especially the BPT scenario)? Text S1 gives no indication as well.

Reviewer #3

(Remarks to the Author)

Zhang et al explore China's contributions to emissions of ozone-depleting chlorine from two short-lived chlorinated chemicals not controlled by the Montreal Protocol, and they project changes through 2035 considering two different scenarios. The results are derived from an activity-based analysis of China's production and use statistics for these chemicals in a variety of applications, and associated emission factors. The authors conclude that emissions of these "uncontrolled" chemicals from China are large relative to some chemicals controlled by the Montreal Protocol, and that those emissions are likely to increase in the future. The analysis is unique and contributes to the state of our understanding, primarily through the quantification of China's large contribution to emissions of these gases and by quantifying benefits associated with potential regulatory actions commensurate with those taken in other countries.

I would find the analysis appropriate for publication after further consideration is given to some key uncertainties associated with this analysis (ODP, IOD); if the authors could provide some additional ground-truth analysis to verify their inventory analysis; and if they are able to successfully address the minor comments provided below.

Estimates of ozone impacts from emissions of these short-lived gases are provided using the Integrated Ozone Depletion Index (IOD) (main text and SI). IOD relies on an accurate understanding of losses in the stratosphere relative to all losses, not the total lifetime, so the lifetime scaling to calculate the IOD for CHCl₃ (given Pyle's analysis of the IOD for CH₂Cl₂) isn't properly considered (re, line 67 of SI). Getting this loss ratio correct is rather important to the conclusions drawn in this paper with respect to ozone impacts, and a discussion is warranted, given that this ratio (strat loss/all loss) is a small number for these short-lived gases and highly uncertain. While Pyle's analysis suggests the ratio is about 8% (strat loss vs total loss), other estimates suggest (SPARC lifetime Report, 2013, table 3.7) that this ratio could be ~3% for chemicals (e.g., CH₃Cl) that have even longer OH lifetimes than CH₂Cl₂ and CHCl₃—suggesting that this ratio for VSLs could be even smaller than 3%. I also appreciate the discussion on lines 158-170 suggesting that the delivery of chlorine from VSLs could be even more efficient than the analysis of mean steady-state lifetimes might suggest. Such uncertainties need to be reflected in values for IOD and SODP-weighted emissions (CFC-11 eq) estimated for CH₂Cl₂ and CHCl₃ (lines 139-143 and elsewhere). Also, it isn't clear what the range quoted for the IOD for CFC11 emissions during 2012-2019 represents.

Inventory analyses of emissions associated with complex and multiple uses and applications, such as that provided in this

manuscript, are very useful, but they can provide answers that are quite different from top-down estimates based on atmospheric measurements. Strengthening a reader's confidence in the accuracy of the emission estimates and impacts estimated by the inventory analysis is a key component to strengthening the manuscript. The authors should consider how they might accomplish this independent of simply showing agreement with other inventory-based analyses. For example, why are no China-specific atmosphere-based estimates of emissions for these VSLS gases are provided in Figure 3? Kim et al (GRL, 2010) published values for 2008 and I'd be surprised if others have not been published for more recent years. Consider that such results are an essential component to establishing and confirming the reliability of the estimates you are providing here through your inventory analysis. Even the total "Asian estimates" provided in Figure 3 might be useful if a careful analysis of China's contribution to those totals over time was possible through some ancillary country-specific data.

Details:

Use of significant figures needs to be carefully considered throughout. In particular, quoting projected emissions with 4 or 5 significant figures given uncertainties that are larger than +/-20% is inappropriate.

Line 18, overstated? To say that these emissions threaten recovery means that recovery might not occur as a result, and that point is not made in the paper. More likely is that these emissions could or will delay recovery, perhaps substantially if they continue? Rephrasing is needed for accuracy.

Line 24, "would exceed"? Explanation needed

Line 25, international efforts, or domestic efforts in China? Explanation needed

Line 37, non-zero ODP, what's the point?

Line 41, citation lacking, what has increased, emission, concentration, both?

Line 67, report national emissions?

Line 86 consider clearer language "emissions reach xxx in the BAU scenario without additional controls."

Line 97 "from restrictions in those processes"? Also on that line, the wording isn't clear, is the potential ban being discussed directed toward PU foaming or on use of CH₂Cl₂ in PU foaming?

Line 105, future tense used in a way that suggests this reduction is likely to happen. Are there laws in place that will facilitate this? Or is instead are emission reductions possible if these practices were adopted?

Figure 4, it appears that the feedstock process for CH₂Cl₂ is incorrectly indicated as producing HCFC-22 as opposed to HFC-32 (the latter being mentioned in the text, lines 50 and 197). Mention of HCFC-22 on line 37 of the SI is also confusing, as HCFC-22 is not involved with CH₂Cl₂ and HFC-32 production, is it?

Line 173, unclear on meaning of text: "emissions from these sectors occur simultaneously" as it doesn't imply immediate benefits. Do you mean emissions occur shortly after use?

In paragraph S1 text, some background info is lacking, is it that production reporting for CH₂Cl₂ and CHCl₃ separately was considered less reliable than what you have done? Also lacking are sources or citations for HCFC-22 production amounts in the past that are projected to forward years.

SI line 33, I'm not clear on the point of "required average annual compound growth rate..."

Regarding N₂O and use of the term ODS. In the ozone science community, the term ODS is typically reserved for Montreal-Protocol controlled gases, and N₂O and VSLS are not controlled and therefore not labeled as such. Consider replacing with "chemical influencing ozone concentrations or ozone depletion" or something else.

Standard of English usage throughout the manuscript is quite good and typically doesn't limit understanding, though there are many small errors related to verb tense, use of definitive articles, or number agreement between subject and verb that could be addressed.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

My previous concerns were well addressed. The following two further comments from the response are provided as below:

1. The authors acknowledge in their response that the time-invariant treatment of natural CHCl₃ sources may explain part of the gap with top-down estimates. This is an important limitation. I recommend explicitly stating it in the main text (e.g., in the

Discussion or a dedicated Uncertainty section), rather than only in the response and the Supplementary Information.

2. The authors have clarified in the main text that a baseline uncertainty of $\pm 10\%$ was systematically applied to single-value emission factors. To improve reproducibility, it would be helpful to clearly indicate in Supplementary Text S6 which specific sectors adopt this default uncertainty and where it applies.

Reviewer #2

(Remarks to the Author)

I congratulate the authors for improving this manuscript, especially with regard to the transparency of their methodology. Almost all of the points raised in the original review have been addressed appropriately, with the exception of the comment on uncertainties. While the revised manuscript provides much better insight into how uncertainties are being considered, the main message that "findings reveal a sustained upward trend in historical emissions for both substances" is too strong given the large uncertainties shown in Figure 3. This could be improved by adopting a more careful wording, making it clearer to the reader that such trends are found for the best estimate only. Adding uncertainty ranges to the observed trends would also help to elucidate whether these are actually significantly different between 2010 and 2023. Once this rather small issue is addressed, I would recommend the publication the manuscript.

Reviewer #3

(Remarks to the Author)

The authors have done a thorough job of addressing the review comments. Only a few minor issues remain, see below.

Abstract: lines 25-27, "would exceed" is not very useful without a time indicated, I'd suggest you mention the approximate date or "by the end of that interval" or "by 2035".

Reconsider the point in the first part of the discussion section. I read the figure to show that total HCFC emissions (from Wu et al., were comparable to CH₂Cl₂ emissions ever since 2015, not just in 2020.

Line 60-61 on the relative contribution of CH₂Cl₂ anthropogenic emissions to total. The 90% contribution was cited from a 2006 report and anthropogenic emissions have increased 2 to 3 fold since potentially suggesting that the relative contribution of this potential natural source is 2 to 3 times smaller now compared to an estimate made then. Also, see the updated discussion in Claxton et al., 2020

Apologies for missing the top-down results that appeared in Figure S1. To ensure others don't do the same, I'd highly recommend that you call out figure S1 on line 85 in the Results section, and you could move forward that call-out for CHCl₃ from line 132 to line 130.

Consider rewording phrasing suggesting "scenario could achieve..." scenarios don't achieve things, but "In scenario xxx, we estimate that ..." makes clear the point you are making.

Citations, reconsider citing WMO reports. It is much more appropriate to cite lead authors names for the appropriate chapter. The only time a reference to WMO (2022) or WMO (2021) is appropriate, for example, is if you are referring to the content of the entire report.

The discussion of IOD and VSLS lifetimes in the stratosphere vs troposphere is improved, but is incomplete without a mention of the tau(strat)/tau(trop) value you used. This would allow a straightforward comparison and to the value suggested for CH₃Cl elsewhere—make it easy on the reader so they don't have to dig up other literature to know how the different values you are discussing relate to one another. I'd also suggest you include a call-out to the methods section (or SI) on line 180.

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Response to Editor and Reviewers

We extend our gratitude to the editor and reviewers for their meticulous review and insightful suggestions, which have undoubtedly enhanced the quality of this paper. We sincerely appreciate their valuable contributions. Our response can be found below, with the Lines in the response message referring to the Clear-version of the revised manuscript.

Reviewer #1 (Remarks to the Author):

Comments on “China’s Chlorinated VSLs Surpass HCFCs in CFC-11-Equivalent Emissions for Ozone Layer Depletion”. This study provides critical insights into the historical and projected emissions of dichloromethane (CH₂Cl₂) and chloroform (CHCl₃) in China. It addresses a significant gap in the current literature, which predominantly relies on top-down atmospheric inversions for chlorinated very short-lived substances (Cl-VSLs), by constructing a detailed, sector-resolved bottom-up emission inventory.

Response: Thanks for your careful review and summary of the content of the manuscript.

However, several concerns regarding the reliability of the emission estimates should be addressed to strengthen the manuscript's conclusions.

Major comments:

1. The representativeness of the emission factor used should be enhanced. For example, the CH₂Cl₂ emissions from adhesive utilization and coating were calculated with emission factors (EF) in 2009 and 2012, respectively. The representativeness of such old EFs is doubtful. It is recommended to use a dynamic EF, which varies with the year.

Response: We sincerely thank the reviewer for this insightful and constructive suggestion. We have discarded the static EFs and reconstructed the emission inventory using dynamic, annually varying EFs for both the adhesive and coating industries, to reflect the technological transitions in this sector in China. Furthermore, we have conducted a systematic review and update of the emission factors across all remaining sectors, including but not limited to the pulp and paper industry, coal production, coal combustion, and water treatment sectors. Details have been added to **Text S6 and Table**

S1-S3 in the Supplementary Information:

Line 155-303 (SI Text S6):

(1) Flexible PU Foaming sector

CH₂Cl₂ functions exclusively as a physical blowing agent (PBA) in the flexible polyurethane (PU) foam industry, facilitating cellular expansion through phase transition without participating in chemical synthesis. Consequently, the total mass of CH₂Cl₂ incorporated into the formulation is theoretically committed to eventual atmospheric release. Consequently, the entire quantity of CH₂Cl₂ added to the formulation is theoretically destined for eventual emission in the absence of capture measures²⁵. Following *Handbook of Polyurethane Products Manufacturing*²⁶, the baseline EF was parameterized at 0.11 g CH₂Cl₂/g of flexible foam produced.

While end-of-pipe control technologies, such as activated carbon adsorption, may be deployed, their effective mitigation potential is fundamentally constrained by operational requirements. However, it is important to note that foaming workshops necessitate exceptionally high ventilation rates to dilute highly toxic toluene diisocyanate (TDI) vapors. This high-volume dilution results in extremely low CH₂Cl₂ concentrations in the effluent gas, which significantly constrains the capture efficiency of adsorption systems. Previous research suggests that even when exhaust systems are optimized for vapor recovery, the net capture efficiency of CH₂Cl₂ typically ranges between 33% and 46%²⁷. This indicates that for facilities equipped with abatement technologies, actual emissions are approximately 30% to 50% lower than the theoretical values derived from the formulation. Industrial formulations exhibit substantial heterogeneity driven by target foam specifications, such as density and hardness. Notably, CH₂Cl₂ loading rates frequently exceed baseline values in the production of specialized low-density foams. Consequently, the 40% uncertainty parameter is employed as a robust bounding estimate. This interval simultaneously accounts for the potential attenuation of emissions through abatement systems and the inherent variability of raw material consumption across diverse and non-uniform production lines.

(2) Adhesive and coating industries

To achieve volatile organic compound (VOC) mitigation, the Chinese government has deployed a multifaceted regulatory framework targeting the adhesive and coating industries. Key interventions include the enforcement of mandatory national standards regarding VOC content limits and the introduction of a consumption tax on coatings in 2015^{18,28}. These mandates prioritize upstream source substitution, specifically incentivizing the transition toward water-borne formulations.

The content of halogenated hydrocarbons (CH_2Cl_2 , 1,2-dichloroethane, 1,1,1-trichloroethane, and 1,1,2-trichloroethane) in adhesive products must comply with the regulations set in GB 30982⁹ or GB 33372^{10,11}. Although relevant standards have not tightened the concentration limits for halogenated hydrocarbons (primarily CH_2Cl_2) in adhesives, the market penetration of these high-emission formulations has undergone a sustained decline. This shift has led to a decrease in the overall EF for the adhesive industry, which dropped from approximately 3.8 g/kg in 2010 to 2.8 g/kg in 2022. The calculation methodology is based on the consumption volumes of different adhesive types^{29–32} and their respective halogenated hydrocarbon limits, including GB 18583-2008³³, GB 30982-2014³⁴, and GB 19340-2014³⁵. Under these standards, for solvent-based adhesives, the concentration of halogenated hydrocarbons is restricted to 5 g/kg for neoprene-based types and 50 g/kg for all other varieties, while CH_2Cl_2 levels in SBS (Styrene-Butadiene-Styrene block copolymer) adhesives must not exceed 50 g/kg.

Within the coating sector, mitigation trajectories diverge between sub-sectors. In architectural coatings, where water-borne technologies had already achieved mainstream status by 2010, the potential for further CH_2Cl_2 attenuation is marginal. Conversely, industrial coatings maintain a lower adoption rate of low-VOC alternatives—accounting for less than 20% of the market as of 2019, according to the Ministry of Ecology and Environment¹⁸. Consequently, the aggregate CH_2Cl_2 EF for the coating industry has not exhibited a statistically significant downward trend. To capture the longitudinal impact of these disparate regulatory outcomes, this study adopts annually varying EFs. These factors were derived by integrating the consumption volumes of specific coating categories with their respective water-borne market shares, synthesized from the framework established by Qi et al.³⁶ and applicable mandatory national standards, including GB 18581-2009³⁷, GB 30981-2014³⁸, GB 38469-2019³⁹, GB 18581-2020⁴⁰, GB 18582-2020⁴¹, GB 24409-2020⁴², GB 30981-2020⁴³. Specifically, under these standards, the halogenated hydrocarbon content in solvent-based coatings is capped at 1% for industrial and marine coatings, and 0.1% for vehicle, architectural, and wood (furniture) coatings.

(3) Pharmaceutical industry

The EF of CH_2Cl_2 from the pharmaceutical industry was calculated based on the material balance using Eq.(S2)²:

$$EF_{pha} = b \times (1 - K) \times 10^3 \times [i \times (1 - \alpha) + (1 - i)] \quad (\text{S2})$$

Here, EF_{pha} is the CH_2Cl_2 emission required to produce a ton of penicillin or cephalosporin, kg/t; b represents the ratio of the solvent mass to the penicillin product or the cephalosporin mass needed for production (7-8); K is the solvent recovery

(85%-97%); α is the proportion of waste gas treated in the pharmaceutical industry as a whole 88%-95%; i is the proportion of waste gas treated at an individual plant. The proportion of waste gas undergoing treatment at the individual plant was parameterized at 50% before 2017². Following the implementation of the National Mandatory *Emission Standard of Air Pollutants for the Pharmaceutical Industry* (GB 37823-2019)⁴⁴ in late 2017, this ratio was linearly scaled to reach 100% by 2020. The mean value of EF_{pha} is 0.37 g/g when the proportion of waste gas treated at an individual plant is 50%.

(4) Pulp and paper-making industry

We validated our AOX (Adsorbable Organic Halogens) EFs for the pulp and paper industry using the *Guidelines for Advanced Applicable Technologies for Energy Saving and Emission Reduction in Light Industry*⁴⁵ from the Ministry of Industry and Information Technology of China (MIIT). These guidelines provide specific AOX emission ranges for the two dominant pulping processes in China: wheat straw chemical pulping (representing the majority of domestic virgin pulp) and deinked recovered paper pulping. To ensure consistency across varying production environments, all values are reported in Air-Dry Tonnes (adt), a standard industry measure representing pulp mass at a theoretical 10% moisture content.

To derive a representative national average, we performed a weighted analysis based on the technological distribution and raw material structure of the Chinese industry. According to the MIIT (2012) framework, ‘Advanced Levels’—representing the top-performing facilities—account for approximately 20% of virgin pulp production and 70% of recovered paper pulp production. For virgin pulp, emission intensities range from 0.25–0.72 kg/adt at the advanced level to 1.0–1.2 kg/adt at the general level. For recovered paper pulp, these values are significantly lower, ranging from 0.01–0.15 kg/adt (advanced) to 0.15–0.3 kg/adt (general). By integrating these technological splits with the industry’s raw material structure—where recovered paper pulp consistently accounted for 70–80% of total production⁴⁶—we determined that the composite AOX emission intensity for the Chinese industry ranges from 0.252 to 0.422 kg/adt for the early 2010s.

Based on the fundamental chemical coupling between liquid-phase AOX and gas-phase CHCl_3 , which are co-generated through identical chlorination and haloform pathways, we adopted the calculated AOX emission range as a scientifically justified proxy for the potential CHCl_3 emission levels. While CHCl_3 is more volatile, the AOX intensity serves as a reliable indicator of the total chlorination capacity of a given facility. This alignment is supported by the fact that the calculated AOX range (0.252–0.422 kg/adt) is fully encapsulated within the CHCl_3 EF (0.053–0.454 kg/adt) from previous

literature^{47–49}. This overlap demonstrates that our chosen parameters accurately capture the technological profile of the Chinese paper industry during the study period.

Regarding temporal dynamics, our assumption that the industry-wide average EF converged to the 2012 ‘Advanced Level’ by 2023 is anchored in the sector’s pioneering role in China’s environmental reforms. Specifically, the share of advanced production processes (as categorized in 2012) for virgin and recovered paper pulp increased linearly from 20% and 70% in 2012, respectively, to 100% by 2023. As mandated by the State Council, the pulp and paper industry was among the first to implement the Pollutant Discharge Permitting System, requiring mills to meet technology-based standards as early as 2017⁵⁰. Furthermore, the *Guiding Catalog for Industrial Structure Adjustment* (2019 Edition)¹⁷ officially designated Elemental Chlorine Bleaching (ECH) as an ‘Eliminated Category’, providing a statutory mandate for the decommissioning of chlorine-intensive legacy assets. Combined with the stringent decarbonization and ‘Green Manufacturing’ objectives of the 14th Five-Year Plan (2021–2025)⁵¹, the 2012 advanced benchmarks have effectively transitioned from elective performance targets into the minimum requisite standards for legal operation and market viability by 2023. Consequently, utilizing these parameters provides a robust and conservative framework that reflects the technological modernization of the industry and the substantial environmental dividends accrued from the universal adoption of Elemental Chlorine-Free (ECF) processes. The final calculated EFs are shown in Supplementary Table S3.

(5) Water treatment

For the drinking water treatment sector, the concentration of CHCl_3 in finished tap water typically does not exceed a regulatory or observed upper limit of 100 $\mu\text{g/L}$, with empirical mean values oscillating between 10 and 20 $\mu\text{g/L}$ ^{52,53}. Under a conservative assumption of a 100% volatilization-to-emission efficiency, the atmospheric EF for this sector was parameterized at a range of 1×10^{-2} to 2×10^{-2} g/m^3 water treated.

For the wastewater treatment sector, the total EF (EF_{Total}) for CHCl_3 was quantified by integrating primary influent volatilization (EF_{pri}) and secondary gaseous release during effluent chlorination (EF_{sec}), as defined in Eqs. (S3)-(S5).

$$EF_{Total} = EF_{pri} + EF_{sec} \quad (\text{S3})$$

$$E_{pri} = C_{inf} \times f_{aeration} \times 10^{-6} \quad (\text{S4})$$

$$E_{sec} = C_{eff,i} \times f_{escape} \times 10^{-6} \quad (\text{S5})$$

The aqueous concentrations of CHCl_3 in the influent ($C_{inf,i}$) and effluent ($C_{eff,i}$) were parameterized at 7.0 $\mu\text{g/L}$ and 14.6 $\mu\text{g/L}$, respectively, based on localized measurements

in Chinese wastewater treatment plants (Li et al.⁵⁴). To account for phase partitioning during biological treatment, an aeration stripping fraction ($f_{aeration}$) of 0.18 was adopted (Zhu et al.⁵⁵). Furthermore, given the high vapor pressure and biological recalcitrance of CHCl_3 in oxygenated surface waters, an ultimate environmental escape fraction (f_{escape}) of 1.0 was applied⁵⁶, assuming complete volatilization within the receiving water boundary based on low concentration in natural water. Based on these parameters, the integrated EF for the wastewater treatment sector was calculated to be $1.6 \times 10^{-2} \text{ g/m}^3$.

(6) Waste incineration

To ensure the reliability of our emission inventory, we utilized localized empirical data from a comprehensive field study of four municipal solid waste incineration plants in North China⁵⁷. To mitigate the impact of facility-specific outliers driven by heterogeneous waste streams, we adopted the mean mass fractions of specific halocarbons within the total VOC (TVOC) profile as representative parameters for the sector. Specifically, CH_2Cl_2 and CHCl_3 accounted for an average of 4.58% and 0.73% of the TVOC emissions, respectively. By applying these mean fractions to the observed TVOC emission baseline ($2.43 \pm 2.27 \text{ g/tonne-waste}$), we derived consolidated EFs of $0.111 \pm 0.104 \text{ g/tonne-waste}$ for CH_2Cl_2 and $0.018 \pm 0.017 \text{ g/tonne-waste}$ for CHCl_3 .

Additional summarized information can be found in **Table S1-S3** of the Supplementary Information.

2. Please provide a deeper discussion on the persistent gap between your bottom-up CHCl_3 estimates and top-down results post-2015. Consider if underestimation of unintentional industrial by-product emissions explains this discrepancy.

Response: We appreciate the reviewer's valuable suggestion. We have expanded our discussion in **Text S1**, to further explore the gap between our bottom-up CHCl_3 estimates and top-down results post-2015. Furthermore, the EFs for several industrial by-product CHCl_3 sources, including water treatment and waste disposal, have been updated. In particular, we refined the pulp and paper sector by implementing dynamic, time-dependent EFs, which capture the temporal shifts in industry-specific emission intensities more accurately.

Line 129-132 (Main text): Our inventory shows a gentle increase in annual total CHCl_3 emissions, whereas top-down estimates exhibit a rapid rise followed by a sharp decline¹³. This discrepancy may be partly attributed to our simplified, time-invariant treatment of natural sources, with further analysis provided in Text S1 and Figure S1.

Line 35-47 (SI): Our CHCl₃ emissions exhibit a gentle increasing trend, whereas the top-down estimates show a rapid rise prior to 2017, followed by a sharp decline thereafter. We derived annually varying CHCl₃ emission factors for China's pulp and paper industry, specifically accounting for the ongoing phase-out of elemental chlorine (EC) bleaching under national environmental mandates. This temporal refinement is consistent with global industrial trends; for instance, recent studies have documented a multi-fold decline in CHCl₃ emission intensities across the United States, Canada, and Japan over the past two decades as advanced bleaching technologies were universally adopted⁴. Owing to the presence of substantial natural sources of CHCl₃, this study quantifies only emissions from natural land surfaces. These emissions are treated as time-invariant, and therefore interannual variability in natural CHCl₃ emissions is not represented in our analysis. This simplified treatment may contribute to the discrepancies between the bottom-up inventory and top-down estimates, particularly in years when natural emissions are expected to vary in response to meteorological or biogeochemical conditions⁵.

Please refer to Supplementary Text S3 for the revised time-dependent EFs in the pulp and paper sector, as detailed in our response to Comment #1.

3. Explicitly link your findings on by-product emissions to recent studies on unintentional ODS release from coal combustion and iron & steel production. This highlights a broader regulatory gap beyond unregulated substances.

Response: We have expanded the Discussion regarding the unintentional ODSs during coal combustion and iron & steel production, and emphasized the gaps for compound-specific ODSs regulation in these industries. These revisions are in **main text lines 133-139**:

In our study, by-product emissions of CH₂Cl₂ and CHCl₃ from coal combustion and iron & steel production constitute only a minor fraction of total emissions compared to primary industrial sources. However, recent studies have identified unintentional ODS formation (e.g., CFC-11) during coal combustion and iron & steel processes, underscoring that combustion-related activities may generate ODSs beyond conventionally regulated sectors²⁵⁻²⁷. Collectively, these findings imply that the regulatory significance of by-product emissions is likely species-specific, necessitating rigorous, compound-by-compound evaluations to accurately assess their environmental relevance.

4. Figure S1 shows that the bottom-up CHCl_3 estimations during 2015–2020 are lower compared with the top-down estimations from An et al. (2023). Please consider adding a discussion about the reason for the gap between different methods.

Response: In the revised manuscript, we have added a detailed discussion in **Text S1** of the Supplementary Information to analyze the sources of this gap. One contributor to the discrepancy may be the reliance on static emission factors in previous inventories, which fail to capture the dynamic interannual variability driven by rapid technological transitions (e.g., 2015–2017). By implementing sector-specific, annually-varying parameters for the pulp and paper industry, we now better resolve these temporal shifts. Additionally, we acknowledge that our time-invariant treatment of natural CHCl_3 sources overlooks meteorologically driven fluctuations, which are inherently captured in top-down estimates (An et al., 2023). These additions provide a more comprehensive explanation for the observed gap and clarify the respective strengths and limitations of bottom-up and top-down approaches **as we detailed in our response to previous comment #2.**

Reference:

- An, M. *et al.* Anthropogenic Chloroform Emissions from China Drive Changes in Global Emissions. *Environ. Sci. Technol.* **57**, 13925–13936 (2023).

5. Compared with previous literature (Feng et al., 2018, Bottom-up anthropogenic dichloromethane emission estimates from China for the period 2005–2016 and predictions of future emissions., *Atmos. Environ.*), the ratio of production leakage to CH_2Cl_2 emissions is significantly lower in the current work (Figure 1). What factors account for this?

Response: When comparing the overlapping period of 2010–2016 (noting that numbers after 2017 from Feng et al. were projections), the absolute production leakage emissions are in very close agreement between the two studies. Feng et al. report an average CH_2Cl_2 production leakage of approximately 15.9 Gg yr^{-1} , while our estimate for the same period is 15.8 Gg yr^{-1} . These values are essentially consistent. The lower ratio of production leakage to total CH_2Cl_2 emissions in our study is not due to a reduction in the leakage emissions, but rather a more comprehensive characterization of total CH_2Cl_2 emissions.

This divergence stems from two primary structural refinements. First, we utilized reconstructed production data to resolve inconsistencies in official records (**Text S5**). By accurately subtracting HFC-32 feedstock use, our dataset allocates a larger volume

of CH₂Cl₂ to non-feedstock applications, establishing a more robust upper-bound constraint. Second, our study incorporates EFs that are better aligned with current industrial practices in China, particularly regarding the CH₂Cl₂ content in flexible PU foam formulations and coatings. In contrast, Feng et al. derived lower EFs by referencing voluntary environmental labeling standards that possess low market penetration among commercial products. Their methodology may have interpreted the absence of explicit halogenated hydrocarbon limits in certain incomplete standards as an indication of zero emissions, which could lead to highly conservative estimates of emission intensities. Together, these factors explain why the relative contribution of production leakage is lower in our study, even though absolute leakage emissions are comparable.

6. The authors should ensure key quantitative findings are consistently presented alongside their uncertainty ranges, such as the projected crossover with HCFCs and future emission levels. This is crucial for robust conclusions.

Response: We have carefully updated the manuscript to ensure that all key quantitative results, such as the 2035 emission projections and the projected crossover timing with HCFCs, are now consistently accompanied by their respective uncertainty ranges. These updates have been incorporated into both the main text and the summary tables to enhance the robustness of our conclusions.

7. Please briefly justify the linear phase-out assumption in your BPT scenario (e.g., policy drafts or analogous regulations). Additionally, the authors could emphasize the climate co-benefits of clean heating policies for a more integrated argument.

Response: We have extensively revised **Text S2** to provide these detailed international and domestic policy justifications. Specifically, the following text has been added:

Line 49-104 (SI): To evaluate future emission trajectories of CH₂Cl₂ and CHCl₃, two scenarios are defined: the Business-As-Usual (BAU) scenario and the Best Practicable Technology (BPT) scenario (as summarized in Table S4). Given that both substances are included in China's *List of Key Controlled Emerging Pollutants*⁶, accelerated emission mitigation driven by technological and regulatory interventions is anticipated. The BAU scenario projects future emissions based on currently implemented policies with emission factors across all industries remaining at historical levels, whereas the BPT scenario reflects enhanced mitigation informed by stringent international regulatory frameworks—such as the U.S. EPA's *Toxic Substances Control Act* (TSCA)⁷

and the EU's *Best Available Techniques (BAT) reference documents*⁸—as well as emerging policy signals in China.

(1) PU Foam, Coatings, and Adhesives Sectors

China has historically implemented a multifaceted regulatory framework targeting volatile organic compounds (VOCs), prioritizing upstream source substitution through mandatory national standards (e.g., GB 30982⁹, GB 33372^{10,11}). While these policies have catalyzed a transition towards water-borne and low-VOC formulations, the mitigation trajectories and current market penetrations of green alternatives diverge significantly across sub-sectors. The resulting evolution of EFs under historical trends is described in Text S6.

Under the BPT scenario, a full phase-out of the remaining high-emission applications is assumed. Internationally, the U.S. issued the *Final Risk Management Rule for Methylene Chloride*⁷ under TSCA in April 2024 that mandated the phase-out of the vast majority of consumer and industrial uses of CH₂Cl₂, with compliance dates mostly set before 2027. Furthermore, the U.S. EPA's *Significant New Alternatives Policy* (SNAP) effectively prohibited its use in flexible polyurethane (PU) foam production starting from January 3, 2017¹². In China, the regulatory environment is rapidly tightening. The 2021 draft of China's *List of Key Controlled Emerging Pollutants*¹³ proposed a complete phase-out of CH₂Cl₂ as a blowing agent in flexible PU foam by the end of 2025. While this specific mandatory deadline was not retained in the final version, it provides a clear policy signal. Additionally, newly issued mandatory standards (GB 30981.1–2025¹⁴ and GB 30981.2–2025¹⁵) significantly strengthen environmental and health safety requirements for coating products. The *Action Plan for Continuous Improvement of Air Quality*¹⁶ further promotes strict control of construction projects involving paints, adhesives, and cleaning agents with high VOC content, alongside the adoption of low- or zero-VOC alternatives.

Based on these domestic policy trajectories and international precedents, the BPT scenario assumes that the residual usage of high-CH₂Cl₂ formulations in the PU foaming, coating, and adhesive sectors will be progressively phased out over a five-year period from 2026 to 2030. A linear phase-down trajectory is applied to the remaining share of these high-emission applications, which is reflected in the evolution of industry-wide emission factors, reaching complete phase-out (or near-zero limits) by 2030. While actual market transitions may follow non-linear dynamics, this assumption provides a consistent analytical framework for evaluating aggregate mitigation potential.

(2) By-product and Pulp & Paper Sectors

The elemental chlorine bleaching (ECH) process, historically a major source of CHCl_3 emissions in the pulp and paper sector, was classified as an eliminated technology in the 2019 *Guiding Catalogue for Industrial Structure Adjustment*¹⁷. Under the BPT scenario, facilities in China are assumed to progressively optimize processes to meet the best-practice emission levels consistent with the European Commission's BAT reference⁸ by 2035. To reach these targets, emission factors are projected to decrease exponentially, reaching the BAT reference⁸ levels of approximately 0.2 kg/adt for virgin pulp and 3 g/adt for recovered paper pulp by 2035.

For CH_2Cl_2 and CHCl_3 emissions from other inherently high-pollution by-product sources, mitigation is expected to occur synergistically with broader air pollution control efforts. Under the BPT scenario, a continuous regulatory pressure drives a progressive EF reduction of 10% every five years. This trajectory is aligned with China's macro-level targets, such as those outlined in the *Action Plan for Continuous Improvement of Air Quality*¹⁶ and the *Comprehensive Control Plan for Volatile Organic Compounds in Key Industries*¹⁸, which consistently mandate at least 10% reductions in total VOC emissions over successive five-year policy cycles.

We also appreciate the reviewer's suggestion on climate co-benefits of clean heating and coal-to-electricity policies. We note that, while such policies can indeed generate climate and air quality benefits in general, the specific compounds investigated in this study (CH_2Cl_2 and CHCl_3) have relatively low GWPs (11 and 20, respectively) (WMO, 2022), and the contribution of coal use and iron & steel production to their total emissions is minor. Therefore, we respectfully retain our focus of this study on the emission characteristics and sectoral attribution of these species.

Reference:

- WMO. *Scientific Assessment of Ozone Depletion: 2022*. (Geneva, Switzerland, 2022).

8. The authors should add the South China Sea region to Figure 2 to present a complete and accurate national map, aligning with standard cartographic practice.

Response: We sincerely appreciate the reviewer's suggestion on the figures. We have updated **Figure 2** accordingly.

Minor comments:

Line 75, 76, 77, etc.: CH_2Cl_2 emission -> CH_2Cl_2 emissions.

Response: Thank you for pointing this out. We have revised the text in original lines 75, 76, 77, and elsewhere to use “CH₂Cl₂ emissions” instead of “CH₂Cl₂ emission”.

Line 239: “We set a phase-down schedule in the three sectors that the share CH₂Cl₂-free production or product will increase linearly from 20% in 2026 to 100% in 2030.” Is there any reference for the assumption of this linear increase?

Response: We have fundamentally refined our terminology and logic in the revised manuscript and provided the corresponding reference for these. The rationale for the 2026–2030 timeline is anchored in both international regulatory precedents and domestic policy signals, such as China’s *List of Key Controlled Emerging Pollutants*, U.S. EPA’s *Toxic Substances Control Act* (TSCA) and the EU’s *Best Available Techniques (BAT) reference documents*. Please refer to **Line 49-104 (SI)** for details. As this concern aligns with the issues raised in Major comment #7, we kindly refer the reviewer to our response to Major comment #7 for the detailed explanation and the revised text.

Figure 2: The map of the South China Sea is recommended to be added.

Response: Thank you for the suggestion. We have updated **Figure 2** to include the South China Sea region.

Line 196: report -> reported.

Response: Thank you for pointing this out. We have noted the grammatical error; however, upon further revision of this section, we decided to remove the entire sentence to improve the conciseness and flow of the manuscript. As a result, the suggested correction is no longer applicable.

Figure S1: “emisison” -> “emissions”

Response: Thank you for noticing the typo. We have corrected “emisison” to “emissions” in **Figure S1**.

Reviewer #2 (Remarks to the Author):

The authors present some work on current Chinese emissions of CH₂Cl₂ and CHCl₃, based on emission factors of various sources over the course of 25 years (part of which are projections). Given that the increasing global emissions of CH₂Cl₂ in the atmosphere are of growing relevance for stratospheric ozone depletion, this topic is undoubtedly of importance to multiple science communities and of interest to a wide readership.

Response: We thank the reviewer for recognizing the importance of this work. We hope that the revisions and clarifications made in the manuscript further strengthen its value and accessibility to a wide readership.

While this is a well-written manuscript, there are some concerns, the main one being about the traceability of results as well as their uncertainties: The authors have commendably gathered information on activity data and emission factors from quite a wide range of sources. Some of their sources are however inaccessible (e.g., searching for CH₂Cl₂ or dichloromethane at <https://www.stats.gov.cn/sj/> only returns the message "Sorry! Failed to find the information you searched."; other similar examples are, e.g., the China Statistical Yearbook or the China Urban-Rural Construction Statistical Yearbook).

Response: We appreciate the reviewer's concern on the data accessibility. To clarify, all the data used in this study to compile inventories are from literatures or official statistical yearbooks, which are publicly available. First, we would like to clarify the fundamental methodological framework of our inventory. Official statistical sources (such as the NBS database and the China Statistical Yearbook) are utilized exclusively to extract macro-level activity data (e.g., industrial value-added, production volumes, and construction floor space), that are related to emissions of CH₂Cl₂ or CHCl₃. They do not report direct emission quantities or substance-specific chemical inventories. Emissions are subsequently quantified by multiplying these sectoral activity data by compound-specific emission factors. These databases are organized by socioeconomic or industrial activity indicators rather than chemical names; therefore, compound-specific searches may not retrieve the relevant activity data directly.

For example, the process for obtaining **value-added of industry**, used as the allocation factor for CH₂Cl₂ coating emission, is described below:

On the website <https://www.stats.gov.cn/sj/>, users can switch to the English interface. To facilitate easier access and verification of the provincial-level datasets, we have

updated the link in the revised manuscript to point directly to the English-language interface of the National Bureau of Statistics database. Users can navigate via the **Statistical Database** panel to **Annual Data** and select **Regional – Annual by Province**. By switching the display mode from **Region** to **Sequence**, a complete time series of provincial-level data is generated. This ensures the transparency and reproducibility of the activity data used in our emission inventory. Users can navigate **Indicator (left panel) → National Accounts → Gross Regional Product**, where **Value-added of Industry** is available in indicator box (right panel). The final interface for data retrieval is illustrated in **Figure R1**.

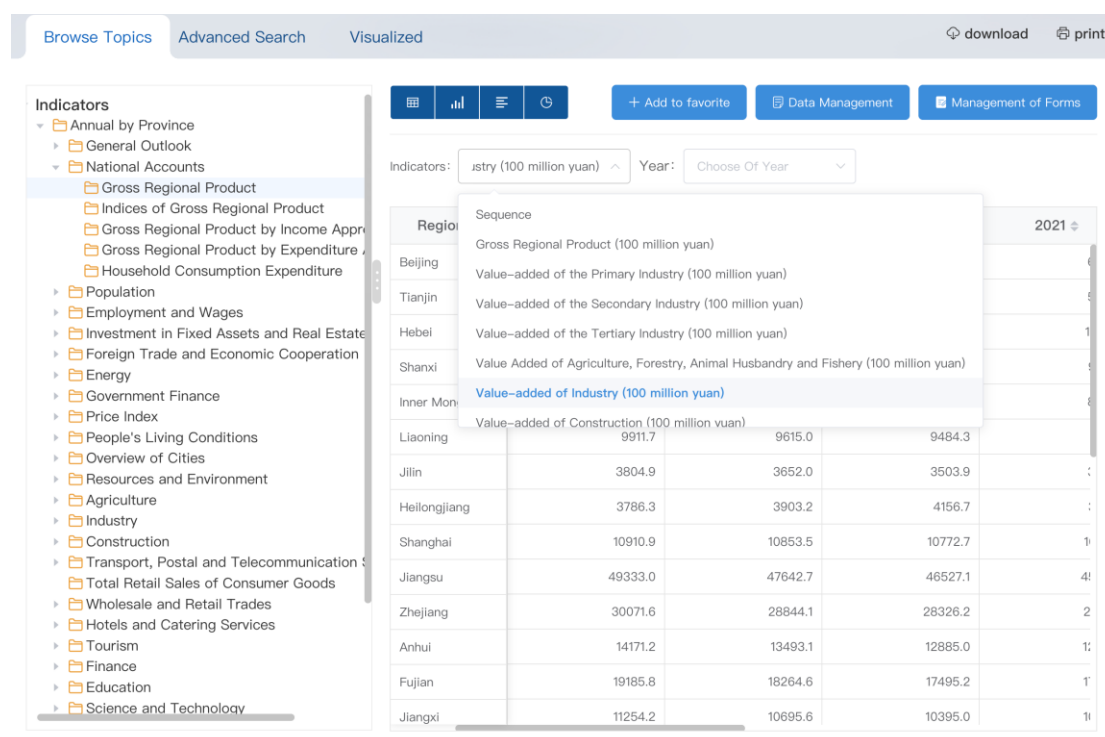


Figure R1. Screenshot of industrial value-added data from the National Bureau of Statistics of China

The activity data in this study were sourced from a series of officially published statistical yearbooks (e.g., China Urban-Rural Construction Statistical Yearbook and China Chemical Industry Yearbook). These are statutory annual publications formally issued by the National Bureau of Statistics of China and relevant government ministries. These yearbooks are standardized public records and are formally publicly available (e.g., from libraries or by purchase). We have attached all the used data as part of the Supplementary Materials.

Other sources are outdated (e.g., IPCC 2000: Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories, Lobert et al., 1999), or it is unclear why only a single number (no range) was chosen for the

emission factor (also Lobert et al., 1999, feedstock use leakage, water treatment, biomass burning, coal combustion,...). As a more general comment, given the continuously evolving nature of industrial processes it is unclear why a single emission factor should even be considered adequate for a) an entire branch of industry, and b) over two decades or more. Even where a range is given, the rationale for this range is not made clear in many cases. Is it, e.g., arbitrarily 10 % for feedstock use leakage/adhesive utilization/coating?

Response: We appreciate reviewer's comment on the emission factor, and we acknowledge that the use of a time-dependent emission factor could better reflect the evolving industry. We have updated the EFs for several high-impact sectors to enhance their representativeness, with the detailed methodology provided in **Text S6 (SI) Line 155-303**:

(1) Flexible PU Foaming sector

CH₂Cl₂ functions exclusively as a physical blowing agent (PBA) in the flexible polyurethane (PU) foam industry, facilitating cellular expansion through phase transition without participating in chemical synthesis. Consequently, the total mass of CH₂Cl₂ incorporated into the formulation is theoretically committed to eventual atmospheric release. Consequently, the entire quantity of CH₂Cl₂ added to the formulation is theoretically destined for eventual emission in the absence of capture measures²⁵. Following *Handbook of Polyurethane Products Manufacturing*²⁶, the baseline EF was parameterized at 0.11 g CH₂Cl₂/g of flexible foam produced.

While end-of-pipe control technologies, such as activated carbon adsorption, may be deployed, their effective mitigation potential is fundamentally constrained by operational requirements. However, it is important to note that foaming workshops necessitate exceptionally high ventilation rates to dilute highly toxic toluene diisocyanate (TDI) vapors. This high-volume dilution results in extremely low CH₂Cl₂ concentrations in the effluent gas, which significantly constrains the capture efficiency of adsorption systems. Previous research suggests that even when exhaust systems are optimized for vapor recovery, the net capture efficiency of CH₂Cl₂ typically ranges between 33% and 46%²⁷. This indicates that for facilities equipped with abatement technologies, actual emissions are approximately 30% to 50% lower than the theoretical values derived from the formulation. Industrial formulations exhibit substantial heterogeneity driven by target foam specifications, such as density and hardness. Notably, CH₂Cl₂ loading rates frequently exceed baseline values in the production of specialized low-density foams. Consequently, the 40% uncertainty parameter is employed as a robust bounding estimate. This interval simultaneously accounts for the potential attenuation of emissions through abatement systems and the inherent

variability of raw material consumption across diverse and non-uniform production lines.

(2) Adhesive and coating industries

To achieve volatile organic compound (VOC) mitigation, the Chinese government has deployed a multifaceted regulatory framework targeting the adhesive and coating industries. Key interventions include the enforcement of mandatory national standards regarding VOC content limits and the introduction of a consumption tax on coatings in 2015^{18,28}. These mandates prioritize upstream source substitution, specifically incentivizing the transition toward water-borne formulations.

The content of halogenated hydrocarbons (CH_2Cl_2 , 1,2-dichloroethane, 1,1,1-trichloroethane, and 1,1,2-trichloroethane) in adhesive products must comply with the regulations set in GB 30982⁹ or GB 33372^{10,11}. Although relevant standards have not tightened the concentration limits for halogenated hydrocarbons (primarily CH_2Cl_2) in adhesives, the market penetration of these high-emission formulations has undergone a sustained decline. This shift has led to a decrease in the overall EF for the adhesive industry, which dropped from approximately 3.8 g/kg in 2010 to 2.8 g/kg in 2022. The calculation methodology is based on the consumption volumes of different adhesive types^{29–32} and their respective halogenated hydrocarbon limits, including GB 18583-2008³³, GB 30982-2014³⁴, and GB 19340-2014³⁵. Under these standards, for solvent-based adhesives, the concentration of halogenated hydrocarbons is restricted to 5 g/kg for neoprene-based types and 50 g/kg for all other varieties, while CH_2Cl_2 levels in SBS (Styrene-Butadiene-Styrene block copolymer) adhesives must not exceed 50 g/kg.

Within the coating sector, mitigation trajectories diverge between sub-sectors. In architectural coatings, where water-borne technologies had already achieved mainstream status by 2010, the potential for further CH_2Cl_2 attenuation is marginal. Conversely, industrial coatings maintain a lower adoption rate of low-VOC alternatives—accounting for less than 20% of the market as of 2019, according to the Ministry of Ecology and Environment¹⁸. Consequently, the aggregate CH_2Cl_2 EF for the coating industry has not exhibited a statistically significant downward trend. To capture the longitudinal impact of these disparate regulatory outcomes, this study adopts annually varying EFs. These factors were derived by integrating the consumption volumes of specific coating categories with their respective water-borne market shares, synthesized from the framework established by Qi et al.³⁶ and applicable mandatory national standards, including GB 18581-2009³⁷, GB 30981-2014³⁸, GB 38469-2019³⁹, GB 18581-2020⁴⁰, GB 18582-2020⁴¹, GB 24409-2020⁴², GB 30981-2020⁴³. Specifically, under these standards, the halogenated hydrocarbon content in solvent-based coatings is capped at 1% for industrial and marine coatings, and 0.1% for

vehicle, architectural, and wood (furniture) coatings.

(3) Pharmaceutical industry

The EF of CH₂Cl₂ from the pharmaceutical industry was calculated based on the material balance using Eq.(S2)²:

$$EF_{pha} = b \times (1 - K) \times 10^3 \times [i \times (1 - \alpha) + (1 - i)] \quad (S2)$$

Here, EF_{pha} is the CH₂Cl₂ emission required to produce a ton of penicillin or cephalosporin, kg/t; b represents the ratio of the solvent mass to the penicillin product or the cephalosporin mass needed for production (7-8); K is the solvent recovery (85%-97%); α is the proportion of waste gas treated in the pharmaceutical industry as a whole 88%-95%; i is the proportion of waste gas treated at an individual plant. The proportion of waste gas undergoing treatment at the individual plant was parameterized at 50% before 2017². Following the implementation of the National Mandatory *Emission Standard of Air Pollutants for the Pharmaceutical Industry* (GB 37823-2019)⁴⁴ in late 2017, this ratio was linearly scaled to reach 100% by 2020. The mean value of EF_{pha} is 0.37 g/g when the proportion of waste gas treated at an individual plant is 50%.

(4) Pulp and paper-making industry

We validated our AOX (Adsorbable Organic Halogens) EFs for the pulp and paper industry using the *Guidelines for Advanced Applicable Technologies for Energy Saving and Emission Reduction in Light Industry*⁴⁵ from the Ministry of Industry and Information Technology of China (MIIT). These guidelines provide specific AOX emission ranges for the two dominant pulping processes in China: wheat straw chemical pulping (representing the majority of domestic virgin pulp) and deinked recovered paper pulping. To ensure consistency across varying production environments, all values are reported in Air-Dry Tonnes (adt), a standard industry measure representing pulp mass at a theoretical 10% moisture content.

To derive a representative national average, we performed a weighted analysis based on the technological distribution and raw material structure of the Chinese industry. According to the MIIT (2012) framework, ‘Advanced Levels’—representing the top-performing facilities—account for approximately 20% of virgin pulp production and 70% of recovered paper pulp production. For virgin pulp, emission intensities range from 0.25–0.72 kg/adt at the advanced level to 1.0–1.2 kg/adt at the general level. For recovered paper pulp, these values are significantly lower, ranging from 0.01–0.15 kg/adt (advanced) to 0.15–0.3 kg/adt (general). By integrating these technological splits with the industry’s raw material structure—where recovered paper pulp consistently

accounted for 70–80% of total production⁴⁶—we determined that the composite AOX emission intensity for the Chinese industry ranges from 0.252 to 0.422 kg/adt for the early 2010s.

Based on the fundamental chemical coupling between liquid-phase AOX and gas-phase CHCl_3 , which are co-generated through identical chlorination and haloform pathways, we adopted the calculated AOX emission range as a scientifically justified proxy for the potential CHCl_3 emission levels. While CHCl_3 is more volatile, the AOX intensity serves as a reliable indicator of the total chlorination capacity of a given facility. This alignment is supported by the fact that the calculated AOX range (0.252–0.422 kg/adt) is fully encapsulated within the CHCl_3 EF (0.053–0.454 kg/adt) from previous literature^{47–49}. This overlap demonstrates that our chosen parameters accurately capture the technological profile of the Chinese paper industry during the study period.

Regarding temporal dynamics, our assumption that the industry-wide average EF converged to the 2012 ‘Advanced Level’ by 2023 is anchored in the sector’s pioneering role in China’s environmental reforms. Specifically, the share of advanced production processes (as categorized in 2012) for virgin and recovered paper pulp increased linearly from 20% and 70% in 2012, respectively, to 100% by 2023. As mandated by the State Council, the pulp and paper industry was among the first to implement the Pollutant Discharge Permitting System, requiring mills to meet technology-based standards as early as 2017⁵⁰. Furthermore, the *Guiding Catalog for Industrial Structure Adjustment* (2019 Edition)¹⁷ officially designated Elemental Chlorine Bleaching (ECH) as an ‘Eliminated Category’, providing a statutory mandate for the decommissioning of chlorine-intensive legacy assets. Combined with the stringent decarbonization and ‘Green Manufacturing’ objectives of the 14th Five-Year Plan (2021–2025)⁵¹, the 2012 advanced benchmarks have effectively transitioned from elective performance targets into the minimum requisite standards for legal operation and market viability by 2023. Consequently, utilizing these parameters provides a robust and conservative framework that reflects the technological modernization of the industry and the substantial environmental dividends accrued from the universal adoption of Elemental Chlorine-Free (ECF) processes. The final calculated EFs are shown in Supplementary Table S3.

(5) Water treatment

For the drinking water treatment sector, the concentration of CHCl_3 in finished tap water typically does not exceed a regulatory or observed upper limit of 100 $\mu\text{g/L}$, with empirical mean values oscillating between 10 and 20 $\mu\text{g/L}$ ^{52,53}. Under a conservative assumption of a 100% volatilization-to-emission efficiency, the atmospheric EF for this sector was parameterized at a range of 1×10^{-2} to 2×10^{-2} g/m³ water treated.

For the wastewater treatment sector, the total EF (EF_{Total}) for $CHCl_3$ was quantified by integrating primary influent volatilization (EF_{pri}) and secondary gaseous release during effluent chlorination (EF_{sec}), as defined in Eqs. (S3)-(S5).

$$EF_{Total} = EF_{pri} + EF_{sec} \quad (S3)$$

$$E_{pri} = C_{inf} \times f_{aeration} \times 10^{-6} \quad (S4)$$

$$E_{sec} = C_{eff,i} \times f_{escape} \times 10^{-6} \quad (S5)$$

The aqueous concentrations of $CHCl_3$ in the influent ($C_{inf,i}$) and effluent ($C_{eff,i}$) were parameterized at 7.0 $\mu\text{g/L}$ and 14.6 $\mu\text{g/L}$, respectively, based on localized measurements in Chinese wastewater treatment plants (Li et al.⁵⁴). To account for phase partitioning during biological treatment, an aeration stripping fraction ($f_{aeration}$) of 0.18 was adopted (Zhu et al.⁵⁵). Furthermore, given the high vapor pressure and biological recalcitrance of $CHCl_3$ in oxygenated surface waters, an ultimate environmental escape fraction (f_{escape}) of 1.0 was applied⁵⁶, assuming complete volatilization within the receiving water boundary based on low concentration in natural water. Based on these parameters, the integrated EF for the wastewater treatment sector was calculated to be $1.6 \times 10^{-2} \text{ g/m}^3$.

(6) Waste incineration

To ensure the reliability of our emission inventory, we utilized localized empirical data from a comprehensive field study of four municipal solid waste incineration plants in North China⁵⁷. To mitigate the impact of facility-specific outliers driven by heterogeneous waste streams, we adopted the mean mass fractions of specific halocarbons within the total VOC (TVOC) profile as representative parameters for the sector. Specifically, CH_2Cl_2 and $CHCl_3$ accounted for an average of 4.58% and 0.73% of the TVOC emissions, respectively. By applying these mean fractions to the observed TVOC emission baseline ($2.43 \pm 2.27 \text{ g/tonne-waste}$), we derived consolidated EFs of $0.111 \pm 0.104 \text{ g/tonne-waste}$ for CH_2Cl_2 and $0.018 \pm 0.017 \text{ g/tonne-waste}$ for $CHCl_3$.

Regarding the use of emission factors from earlier literature, these sources were adopted for specific source categories where more recent, sector-specific measurements are still limited or unavailable. Despite their age, these values remain widely used as baseline references in subsequent studies and inventories, particularly for feedstock use leakage and several diffuse sources. We have retained the process-specific IPCC default EFs from our original inventory, as they remain the most scientifically rigorous way to allocate emissions between production and feedstock use without the risk of double-counting. However, we have significantly strengthened the justification for these

parameters by contextualizing them with recent top-down observational evidence and the current regulatory landscape in China, as shown in **lines 305-316 (SI)**:

For the CH₂Cl₂ and CHCl₃ production sector, this study adopted EFs ranging from 0.5% to 4% according to the IPCC guidelines^{58,59}. Recent inverse modeling and plant-level studies in China by Li et al.⁶⁰ reported aggregate EFs of 4% ± 2% for CH₂Cl₂ and 2% ± 0.6% for CHCl₃. This facility has a production capacity of approximately 400 Gg yr⁻¹, accounting for ~13% of the national production capacity in 2019, supporting its representativeness for the Chinese chloromethane production sector. However, it is critical to note that these observations were conducted at integrated industrial parks where chloromethane production is co-located with downstream facilities, such as HFC-32 and HCFC-22 production lines. Consequently, these observational values represent the cumulative emission intensity of the entire industrial chain—including synthesis, feedstock transfer, and utilization. By maintaining separate IPCC process EFs, our bottom-up model can partition these emissions into their respective sectors while ensuring the total remains consistent with these system-wide observational constraints.

We would like to clarify that a baseline uncertainty of ± 10% was systematically applied to all single-value EFs within our emission inventory model. This parameter was an integral part of our methodology, though not explicitly articulated in the original manuscript. We now explicitly state that while a default ± 10% was used for well-characterized sectors, larger uncertainties of up to ± 40% were assigned to the flexible PU foaming sector as previously explained. This tiered approach follows the recommendations of Velders and Daniel (2014) to capture both inter-sectoral variability and technological evolution and have been widely employed in peer-reviewed literature Chen et al. (2025), Bai et al. (2023), Lickley et al. (2020). We have clarified this rationale in the revised manuscript (**Line 257-261**) and SI (**Line 164-179**).

Line 257-261 (Main text): For emission factors supported by only a single data point, we assigned a normal distribution with a baseline uncertainty of 10% (defined as the standard deviation), following Velders et al.⁴⁷. Specifically, a higher uncertainty of 40% was applied to the flexible PU foam sector to account for the greater variability in its industrial formulations and practices. The rationale for this specific assignment is explained in Text S6.

Importantly, because nearly all non-feedstock CH₂Cl₂ is released to the atmosphere, uncertainties in emission factors primarily affect sector-level allocation rather than the total emissions. As a result, the overall magnitude of CH₂Cl₂ emissions remains well-constrained, and the detailed comparison further strengthens the evidence-based assessment of China's emissions, providing additional confidence in the inventory

results presented in this manuscript.

Reference:

- Velders, G. J. M. & Daniel, J. S. Uncertainty analysis of projections of ozone-depleting substances: mixing ratios, EESC, ODPs, and GWPs. *Atmospheric Chemistry and Physics* 14, 2757–2776 (2014).
- Li, B. et al. Emission factors of ozone-depleting chloromethanes during production processes based on field measurements surrounding a typical chloromethane plant in China. *Journal of Cleaner Production* 414, 137573 (2023).
- Chen, Z. et al. Sustainable management of banked fluorocarbons as a cost-effective climate action. *Environ. Sci. Technol.* 59, 16356–16367 (2025).
- Bai, F. et al. Pathway and cost-benefit analysis to achieve china's zero hydrofluorocarbon emissions. *Environ. Sci. Technol.* 57, 6474–6484 (2023).
- Lickley, M. et al. Quantifying contributions of chlorofluorocarbon banks to emissions and impacts on the ozone layer and climate. *Nat Commun* 11, 1380 (2020).

Most important is however that all of the above, when considered more adequately, would lead to a massive increase of the uncertainties of all emissions derived here. This increase (which is of course also partly due to the uncertain nature of bottom-up estimates) would likely render the main messages of this manuscript void (unless the authors can demonstrate otherwise). Comparisons of such results (for other trace gases) with top-down estimates based on atmospheric observations have demonstrated many differences in the past, and in most cases the bottom-up estimates were revised later on. The fact that the An et al. top-down estimates appear to agree very well with the results of this manuscript over several years gives a little bit of confidence (but only for this period). However, the biggest opportunity to add more credibility to this manuscript would be adding more top-down-based data (which exists for both CH₂Cl₂ and CHCl₃) for a more evidence-based assessment of Chinese emissions.

Response: To provide independent validation, the original Supplementary Information (**Figure S1**) compared our bottom-up CH₂Cl₂ and CHCl₃ emissions with results from previous studies, including both top-down estimates (e.g., An et al., 2023) and other bottom-up inventories. In the revised manuscript, we have expanded this analysis in **Main text line 84-88** as well as **SI Text S1** to provide a more comprehensive assessment of consistency and discrepancies among different studies. The expanded

discussion clarifies the uncertainties in both bottom-up and top-down approaches and demonstrates that, despite these uncertainties, the main conclusions regarding sectoral contributions and temporal evolution remain robust. Specifically, the following text has been added:

Line 84-88 (Main text): The historical CH₂Cl₂ emissions are broadly consistent with the top-down constraints from An et al.¹⁴, but exceed the bottom-up estimates of Feng et al.²⁰. This discrepancy may be explained by our refinement of emission factor parameterization, which addresses the systemic underestimations in previous inventories by incorporating more representative industrial data, as well as our reconstruction of the CH₂Cl₂ production data (refer to Text S1 and Figure S1 for more details).

Line 129-132 (Main text): Our inventory shows a gentle increase in annual total CHCl₃ emissions, whereas top-down estimates exhibit a rapid rise followed by a sharp decline¹³. This discrepancy may be partly attributed to our simplified, time-invariant treatment of natural sources, with further analysis provided in Text S1 and Figure S1.

Line 14-47 (SI): Our historical CH₂Cl₂ emissions are generally consistent with those reported in other studies within the respective uncertainty ranges, particularly with the top-down time series emissions reported by An et al.¹ (Figure S1). The differences between our inventory and the estimates of Feng et al.² mainly arise from the CH₂Cl₂ production data used in the two studies. Feng et al.² adopted originally reported production statistics, whereas we employ reconstructed production data that account for inconsistencies in official records (see Text S5 for details). After subtracting feedstock use for HFC-32, the reconstructed production dataset results in a larger amount of CH₂Cl₂ allocated to non-feedstock applications, thereby establishing a more robust upper-bound constraint for emission estimates. Specifically, in our study, the CH₂Cl₂ content in flexible PU foam formulations and EFs for coating utilization and other relevant industries are more representative of current industrial practices. In contrast, Feng et al.² derived lower EFs by referencing voluntary environmental labeling standards that possess low market penetration among commercial products³. Furthermore, their methodology may have interpreted the lack of explicit halogenated hydrocarbon limits in certain standards as an indication of zero emissions. This assumption potentially leads to more conservative estimates of emission intensities compared to the actual industrial practices observed in this study. The bottom-up estimation framework by An et al.¹ assumes that all CH₂Cl₂ consumption—excluding feedstock utilization for HFC-32 and applications within the pharmaceutical industry—is aggregated into a single, 100% emissive solvent sector. Without a sectoral breakdown, that study potentially overlooks non-emissive pathways, positioning these bottom-up

results as an upper-limit constraint on China's total CH₂Cl₂ emissions.

Our CHCl₃ emissions exhibit a gentle increasing trend, whereas the top-down estimates show a rapid rise prior to 2017, followed by a sharp decline thereafter. We derived annually varying CHCl₃ emission factors for China's pulp and paper industry, specifically accounting for the ongoing phase-out of elemental chlorine (EC) bleaching under national environmental mandates. This temporal refinement is consistent with global industrial trends; for instance, recent studies have documented a multi-fold decline in CHCl₃ emission intensities across the United States, Canada, and Japan over the past two decades as advanced bleaching technologies were universally adopted⁴. Owing to the presence of substantial natural sources of CHCl₃, this study quantifies only emissions from natural land surfaces. These emissions are treated as time-invariant, and therefore interannual variability in natural CHCl₃ emissions is not represented in our analysis. This simplified treatment may contribute to the discrepancies between the bottom-up inventory and top-down estimates, particularly in years when natural emissions are expected to vary in response to meteorological or biogeochemical conditions⁵.

Reference:

- An, M. *et al.* Anthropogenic Chloroform Emissions from China Drive Changes in Global Emissions. *Environ. Sci. Technol.* **57**, 13925–13936 (2023).

Nevertheless, if these concerns as well as the comments/questions below can be addressed appropriately, the manuscript could become suitable for publication in this journal.

Firstly, the English language is quite good for most of the manuscript but might benefit from another iteration.

Response: Thank you for this suggestion. We have carefully revised the manuscript and conducted an additional round of language polishing to further improve clarity, consistency, and readability throughout the text.

69 Referring to Taiwan as a part of China might be commonplace in China but could be perceived as controversial internationally. Perhaps the Editorial Office wants to check whether this is in agreement with journal policy.

Response: We clarify that the treatment of Taiwan in this study is fully consistent with the position of the United Nations and the classification adopted within the United

Nation system. Specifically, Taiwan is considered a region of China in United Nation statistical and reporting frameworks.

84 Calling this an "annual emission inventory" seems inappropriate since part of it is in the future.

Response: We agree that the term “annual emission inventory” may be misleading when applied to results that include future projections. We have revised the text to clarify that the inventory refers to historical annual emissions, while emissions for future years are presented as projections/scenarios.

169 The sudden injection of other Cl-VSLs into the manuscript seems out of place.

Response: We have removed the discussion of other Cl-VSLs and revised the text to maintain a clearer focus on CH₂Cl₂ and CHCl₃.

238-241 What is the rationale for these numbers (especially the BPT scenario)? Text S1 gives no indication as well.

Response: We have comprehensively revised **Text S2** to provide the necessary regulatory and methodological justifications for the BPT scenario. Specifically, the following text has been added:

Line 49-104 (SI): To evaluate future emission trajectories of CH₂Cl₂ and CHCl₃, two scenarios are defined: the Business-As-Usual (BAU) scenario and the Best Practicable Technology (BPT) scenario (as summarized in Table S4). Given that both substances are included in China’s *List of Key Controlled Emerging Pollutants*⁶, accelerated emission mitigation driven by technological and regulatory interventions is anticipated. The BAU scenario projects future emissions based on currently implemented policies with emission factors across all industries remaining at historical levels, whereas the BPT scenario reflects enhanced mitigation informed by stringent international regulatory frameworks—such as the U.S. EPA’s *Toxic Substances Control Act* (TSCA)⁷ and the EU’s *Best Available Techniques (BAT) reference documents*⁸—as well as emerging policy signals in China.

(1) PU Foam, Coatings, and Adhesives Sectors

China has historically implemented a multifaceted regulatory framework targeting volatile organic compounds (VOCs), prioritizing upstream source substitution through

mandatory national standards (e.g., GB 30982⁹, GB 33372^{10,11}). While these policies have catalyzed a transition towards water-borne and low-VOC formulations, the mitigation trajectories and current market penetrations of green alternatives diverge significantly across sub-sectors. The resulting evolution of EFs under historical trends is described in Text S6.

Under the BPT scenario, a full phase-out of the remaining high-emission applications is assumed. Internationally, the U.S. issued the *Final Risk Management Rule for Methylene Chloride*⁷ under TSCA in April 2024 that mandated the phase-out of the vast majority of consumer and industrial uses of CH₂Cl₂, with compliance dates mostly set before 2027. Furthermore, the U.S. EPA's *Significant New Alternatives Policy* (SNAP) effectively prohibited its use in flexible polyurethane (PU) foam production starting from January 3, 2017¹². In China, the regulatory environment is rapidly tightening. The 2021 draft of China's *List of Key Controlled Emerging Pollutants*¹³ proposed a complete phase-out of CH₂Cl₂ as a blowing agent in flexible PU foam by the end of 2025. While this specific mandatory deadline was not retained in the final version, it provides a clear policy signal. Additionally, newly issued mandatory standards (GB 30981.1–2025¹⁴ and GB 30981.2–2025¹⁵) significantly strengthen environmental and health safety requirements for coating products. The *Action Plan for Continuous Improvement of Air Quality*¹⁶ further promotes strict control of construction projects involving paints, adhesives, and cleaning agents with high VOC content, alongside the adoption of low- or zero-VOC alternatives.

Based on these domestic policy trajectories and international precedents, the BPT scenario assumes that the residual usage of high-CH₂Cl₂ formulations in the PU foaming, coating, and adhesive sectors will be progressively phased out over a five-year period from 2026 to 2030. A linear phase-down trajectory is applied to the remaining share of these high-emission applications, which is reflected in the evolution of industry-wide emission factors, reaching complete phase-out (or near-zero limits) by 2030. While actual market transitions may follow non-linear dynamics, this assumption provides a consistent analytical framework for evaluating aggregate mitigation potential.

(2) By-product and Pulp & Paper Sectors

The elemental chlorine bleaching (ECH) process, historically a major source of CHCl₃ emissions in the pulp and paper sector, was classified as an eliminated technology in the 2019 *Guiding Catalogue for Industrial Structure Adjustment*¹⁷. Under the BPT scenario, facilities in China are assumed to progressively optimize processes to meet the best-practice emission levels consistent with the European Commission's BAT reference⁸ by 2035. To reach these targets, emission factors are projected to decrease

exponentially, reaching the BAT reference⁸ levels of approximately 0.2 kg/adt for virgin pulp and 3 g/adt for recovered paper pulp by 2035.

For CH₂Cl₂ and CHCl₃ emissions from other inherently high-pollution by-product sources, mitigation is expected to occur synergistically with broader air pollution control efforts. Under the BPT scenario, a continuous regulatory pressure drives a progressive EF reduction of 10% every five years. This trajectory is aligned with China's macro-level targets, such as those outlined in the *Action Plan for Continuous Improvement of Air Quality*¹⁶ and the *Comprehensive Control Plan for Volatile Organic Compounds in Key Industries*¹⁸, which consistently mandate at least 10% reductions in total VOC emissions over successive five-year policy cycles.

Reviewer #3 (Remarks to the Author):

Zhang et al explore China's contributions to emissions of ozone-depleting chlorine from two short-lived chlorinated chemicals not controlled by the Montreal Protocol, and they project changes through 2035 considering two different scenarios. The results are derived from an activity-based analysis of China's production and use statistics for these chemicals in a variety of applications, and associated emission factors. The authors conclude that emissions of these "uncontrolled" chemicals from China are large relative to some chemicals controlled by the Montreal Protocol, and that those emissions are likely to increase in the future. The analysis is unique and contributes to the state of our understanding, primarily through the quantification of China's large contribution to emissions of these gases and by quantifying benefits associated with potential regulatory actions commensurate with those taken in other countries.

Response: We thank the reviewer for the constructive and positive assessment of our study.

I would find the analysis appropriate for publication after further consideration is given to some key uncertainties associated with this analysis (ODP, IOD); if the authors could provide some additional ground-truth analysis to verify their inventory analysis; and if they are able to successfully address the minor comments provided below.

Estimates of ozone impacts from emissions of these short-lived gases are provided using the Integrated Ozone Depletion Index (IOD) (main text and SI). IOD relies on an accurate understanding of losses in the stratosphere relative to all losses, not the total lifetime, so the lifetime scaling to calculate the IOD for CHCl₃ (given Pyle's analysis of the IOD for CH₂Cl₂) isn't properly considered (re, line 67 of SI). Getting this loss ratio correct is rather important to the conclusions drawn in this paper with respect to ozone impacts, and a discussion is warranted, given that this ratio (strat loss/all loss) is a small number for these short-lived gases and highly uncertain. While Pyle's analysis suggests the ratio is about 8% (strat loss vs total loss), other estimates suggest (SPARC lifetime Report, 2013, table 3.7) that this ratio could be ~3% for chemicals (e.g., CH₃Cl) that have even longer OH lifetimes than CH₂Cl₂ and CHCl₃—suggesting that this ratio for VSLS could be even smaller than 3%. I also appreciate the discussion on lines 158-170 suggesting that the delivery of chlorine from VSLS could be even more efficient than the analysis of mean steady-state lifetimes might suggest. Such uncertainties need to be reflected in values for IOD and

SODP-weighted emissions (CFC-11 eq) estimated for CH₂Cl₂ and CHCl₃ (lines 139-143 and elsewhere).

Response: We sincerely thank the reviewer for this profound and constructive insight regarding the stratospheric loss ratio. We agree that the fraction of stratospheric loss relative to total loss is a fundamental source of uncertainty for VSLs, and the comparison with the SPARC (2013) report is particularly valuable. To address these concerns, we have clarified the methodological distinction between the two metrics used in our study: SODP-weighted emissions (in CFC-11-equivalent) and the Integrated Ozone Depletion (IOD) index.

Regarding SODP-weighted emissions, we utilized region-specific SODPs derived from 3D chemical transport model simulations by Claxton et al. (2019), with specific SODP values for different provinces. This 3D chemical transport model study provides a methodological framework that explicitly accounts for the complex interplay between regional transport, seasonality, and chemical degradation. The use of province-specific SODPs in our study provides a realistic evaluation of the impact on stratospheric ozone. Notably, these SODP values exhibit minimal seasonal variability, as shown in Claxton et al. (2019), and we used the annual average value. We have detailed the calculation in Methods of **Main text, line 323-331**:

We adopted region-specific SODP values for the provinces in Tropical and Temperate Asia based on the data from Claxton et al.⁶. Specifically, for CH₂Cl₂, we used 0.014 for Temperate Asia (Jiangsu, Zhejiang Province, etc.) and 0.019 for Tropical Asia (Guangdong Province, etc.). Corresponding values for CHCl₃ were 0.02 (Temperate Asia) and 0.024 (Tropical Asia). If the province is located at the boundary between the tropical and temperate zones, the SODP for CH₂Cl₂/CHCl₃ emitted shall be calculated on an area-weighted basis. While seasonal SODP values are provided by Claxton et al.⁶, they exhibit minimal intra-annual variability for these regions. Therefore, the use of annualized factors ensures methodological robustness while providing a representative estimate of the aggregate ozone depletion contribution. The SODP of HCFCs, CCl₄, and N₂O are derived from *Scientific Assessment of Ozone Depletion: 2022*²¹.

We have also discussed the underlying uncertainties for the SODP used, in **line 196-202**:

Moreover, the CFC-11-equivalent emissions of Cl-VSLs estimated in this study, derived using a constant, annualized SODP factor for tropical and temperate Asia, do not capture intra-annual variability. Efficient transport pathways, such as the Asian monsoon, can enhance the injection of Cl-VSLs into the stratosphere^{9,11,36,37}, implying that actual SODP values may be higher than those estimated by Claxton et al.⁶. Another

source of uncertainty arises from spatial variability. The SODP values used here were derived at a continental scale and may not adequately capture finer-scale transport processes, such as the substantial variations that occur across different provinces.

Regarding the IOD calculation, we clarify that CH_2Cl_2 and CHCl_3 share highly similar tropospheric degradation pathways and exhibit nearly exactly identical values for both total atmospheric lifetimes and tropospheric lifetimes. We acknowledge the potential uncertainties related to this calculation of IOD, from e.g., from the variation in stratospheric delivery efficiencies. Due to the relative lack of quantitative assessments regarding VSLs lifetimes, we are currently unable to propagate this specific uncertainty into the numerical values of IOD and emissions in CFC-11 equivalents. However, we have expanded the Discussion section to include a comprehensive evaluation of these uncertainties. Specifically, the following text has been added:

Line 178-180 (Main text): However, IOD derivation is critically sensitive to the ratio between a species' total lifetime and its stratospheric lifetime, a parameter that represents the stratospheric loss fraction. For Cl-VSLs, this fraction remains subject to significant uncertainty.

We also acknowledge the potential bias from the lifetimes used by Pyle et al., and we have added the following discussion to **line 118-129 (SI)**:

We would also expect similar τ_{strat} for CH_2Cl_2 and CHCl_3 , from the lifetime calculation ($1/\tau_{\text{atmos}} = 1/\tau_{\text{tropo}} + 1/\tau_{\text{strat}}$), since they exhibit nearly identical values for both τ_{atmos} and τ_{tropo} ²⁰. The ratio of $\tau_{\text{strat}}/\tau_{\text{tropo}}$, which is the rationale to calculate IOD, as recommended in Pyle et al.¹⁹, would then be similar for CH_2Cl_2 and CHCl_3 . Thus, we believe it is appropriate to use CH_2Cl_2 as a proxy for the calculation of IOD for CHCl_3 . Their emission levels in 2010 are set as the baseline for IOD calculation.

However, we noticed that the $\tau_{\text{strat}}/\tau_{\text{tropo}}$ ratio reported for CH_2Cl_2 by Pyle et al.¹⁹ exceeds that of longer-lived species such as CH_3Cl (3%, as documented in the SPARC report²¹), which suggests that the framework of Pyle et al.¹⁹ might partially overestimate the relative stratospheric impact of these Cl-VSLs. This could reflect the high sensitivity of short-lived species to model-specific transport schemes and the potential inclusion of product gas injection. However, we are unable to account for this in this manuscript.

Reference:

- Pyle, J. A., Keeble, J., Abraham, N. L., Chipperfield, M. P. & Griffiths, P. T. Integrated ozone depletion as a metric for ozone recovery. *Nature* 608, 719–723

(2022).

- M. Ko, P. Newman, S. Reimann & S. Strahan (Eds.). SPARC Report on the Lifetimes of Stratospheric Ozone-Depleting Substances, Their Replacements, and Related Species. <http://www.sparc-climate.org/publications/sparc-reports/sparc-report-no6/> (2013).
- Claxton, T., Hossaini, R., Wild, O., Chipperfield, M. P. & Wilson, C. On the regional and seasonal ozone depletion potential of chlorinated very short - lived substances. *Geophys. Res. Lett.* 46, 5489–5498 (2019).
- Hossaini, R. *et al.* The increasing threat to stratospheric ozone from dichloromethane. *Nat Commun* 8, 15962 (2017).
- WMO. Scientific Assessment of Ozone Depletion: 2018. (Geneva, Switzerland, 2018).

Also, it isn't clear what the range quoted for the IOD for CFC11 emissions during 2012-2019 represents.

Response: The quoted range specifically represents the range of excess emissions—defined as the cumulative unreported production and use of CFC-11 during the 2012–2019 period—rather than total global emissions. These unexpected emissions are estimated to be between 120 and 440 Gg (WMO, 2021). Correspondingly, these extra emissions have resulted in a cumulative IOD of roughly 9–34 DU years. Accordingly, we have revised the text to clarify that ‘[For reference, the total global extra emissions of CFC-11 over 2012–2019 due to unreported production, estimated at 120–440 Gg³⁴, caused 9–34 DU years of ozone depletion³³](#)’ (Line 176-178, Main text).

Reference:

- World Meteorological Organization (WMO). Report on the Unexpected Emissions of CFC-11. (WMO, Geneva, Switzerland, 2021).

Inventory analyses of emissions associated with complex and multiple uses and applications, such as that provided in this manuscript, are very useful, but they can provide answers that are quite different from top-down estimates based on atmospheric measurements. Strengthening a reader's confidence in the accuracy of the emission estimates and impacts estimated by the inventory analysis is a key component to strengthening the manuscript. The authors should consider how they might accomplish this independent of simply showing agreement with other

inventory-based analyses. For example, why are no China-specific atmosphere-based estimates of emissions for these VSLs gases are provided in Figure 3? Kim et al (GRL, 2010) published values for 2008 and I'd be surprised if others have not been published for more recent years. Consider that such results are an essential component to establishing and confirming the reliability of the estimates you are providing here through your inventory analysis. Even the total "Asian estimates" provided in Figure 3 might be useful if a careful analysis of China's contribution to those totals over time was possible through some ancillary country-specific data.

Response: We sincerely appreciate the reviewer's emphasis on the importance of cross-validating inventory results with top-down estimates. In this revision, we have systematically refined the emission inventory development process to enhance its methodological robustness and representativeness. Specifically, we have updated the emission factors (EFs) to better align with actual industrial practices in China and, where data were available, incorporated annually varying EFs to capture the dynamic technological shifts within key sectors. By reconstructing the inventory with these refined parameters, we provide a more accurate and reliable foundation for our emission estimates. We respectfully clarify that a comprehensive comparison of our results with several prominent studies (e.g., Kim et al., 2010; Wang et al., 2014; An et al., 2021) was included in **Figure S1** of the Supplemental Information in the initial manuscript. To further consolidate these findings, we have significantly expanded the discussion in the revised **Text S1**, adding the following analysis:

Line 14-47 (SI): Our historical CH₂Cl₂ emissions are generally consistent with those reported in other studies within the respective uncertainty ranges, particularly with the top-down time series emissions reported by An et al.¹ (Figure S1). The differences between our inventory and the estimates of Feng et al.² mainly arise from the CH₂Cl₂ production data used in the two studies. Feng et al.² adopted originally reported production statistics, whereas we employ reconstructed production data that account for inconsistencies in official records (see Text S5 for details). After subtracting feedstock use for HFC-32, the reconstructed production dataset results in a larger amount of CH₂Cl₂ allocated to non-feedstock applications, thereby establishing a more robust upper-bound constraint for emission estimates. Specifically, in our study, the CH₂Cl₂ content in flexible PU foam formulations and EFs for coating utilization and other relevant industries are more representative of current industrial practices. In contrast, Feng et al.² derived lower EFs by referencing voluntary environmental labeling standards that possess low market penetration among commercial products³. Furthermore, their methodology may have interpreted the lack of explicit halogenated hydrocarbon limits in certain standards as an indication of zero emissions. This assumption potentially leads to more conservative estimates of emission intensities

compared to the actual industrial practices observed in this study. The bottom-up estimation framework by An et al.¹ assumes that all CH₂Cl₂ consumption—excluding feedstock utilization for HFC-32 and applications within the pharmaceutical industry—is aggregated into a single, 100% emissive solvent sector. Without a sectoral breakdown, that study potentially overlooks non-emissive pathways, positioning these bottom-up results as an upper-limit constraint on China's total CH₂Cl₂ emissions.

Our CHCl₃ emissions exhibit a gentle increasing trend, whereas the top-down estimates show a rapid rise prior to 2017, followed by a sharp decline thereafter. We derived annually varying CHCl₃ emission factors for China's pulp and paper industry, specifically accounting for the ongoing phase-out of elemental chlorine (EC) bleaching under national environmental mandates. This temporal refinement is consistent with global industrial trends; for instance, recent studies have documented a multi-fold decline in CHCl₃ emission intensities across the United States, Canada, and Japan over the past two decades as advanced bleaching technologies were universally adopted⁴. Owing to the presence of substantial natural sources of CHCl₃, this study quantifies only emissions from natural land surfaces. These emissions are treated as time-invariant, and therefore interannual variability in natural CHCl₃ emissions is not represented in our analysis. This simplified treatment may contribute to the discrepancies between the bottom-up inventory and top-down estimates, particularly in years when natural emissions are expected to vary in response to meteorological or biogeochemical conditions⁵.

We clarify that **Figure 3** is specifically designed to compare the CFC-11-equivalent emissions across different species. We have intentionally excluded previous national-scale estimates from this comparison due to the significant spatial heterogeneity of ODPs for VSLs. Since ODPs are highly sensitive to emission locations, directly converting national aggregate emissions into CFC-11 equivalents could be scientifically misleading. Instead, our study derives these equivalents based on spatially resolved ODPs and emissions, providing a more rigorous and accurate assessment of their aggregate contribution to ozone depletion.

Reference:

- Wang, C. *et al.* Estimating halocarbon emissions using measured ratio relative to tracers in China. *Atmospheric Environment* **89**, 816–826 (2014).
- Kim, J. *et al.* Regional atmospheric emissions determined from measurements at Jeju Island, Korea: Halogenated compounds from China. *Geophysical Research Letters* **37**, (2010).
- An, M. *et al.* Rapid increase in dichloromethane emissions from China inferred through atmospheric observations. *Nat. Commun.* **12**, 7279 (2021).

Details:

Use of significant figures needs to be carefully considered throughout. In particular, quoting projected emissions with 4 or 5 significant figures given uncertainties that are larger than +/-20% is inappropriate.

Response: In the revised manuscript, we have carefully re-evaluated the use of significant figures throughout the text. Specifically, all projected emission values have been adjusted to ensure they are commensurate with their respective uncertainty levels.

Line 18, overstated? To say that these emissions threaten recovery means that recovery might not occur as a result, and that point is not made in the paper. More likely is that these emissions could or will delay recovery, perhaps substantially if they continue? Rephrasing is needed for accuracy.

Response: We have revised the sentence to: [Escalating emissions of chlorinated very short-lived substances \(Cl-VSLs\), which remain unregulated under the Montreal Protocol, could delay the recovery of the global ozone layer \(Line 17-18, Main text\).](#)

Line 24, “would exceed”? Explanation needed

Response: We use “would exceed” because the estimates from 2024 onward are projections in this study rather than historical values. Therefore, the 2024 result represents a projected outcome, and the conditional tense is used to reflect this.

Line 25, international efforts, or domestic efforts in China? Explanation needed

Response: While our inventory specifically focuses on China, the call for 'international efforts' is predicated on both the global distribution of usage patterns and the evolving regulatory landscape of the *Montreal Protocol*. Given that China dominates global feedstock applications, it follows that in other regions of the world with high-emission regions, a larger proportion of CH₂Cl₂ and CHCl₃ consumption is likely allocated to non-feedstock, emissive uses such as solvents. This implies that these regions possess substantial, yet currently untapped, mitigation potential as a larger fraction of their consumption is directly released into the atmosphere. Furthermore, there is an increasing focus within the scientific and policy communities on the need to address the 'unregulated' status of VSLs under the Montreal Protocol. International

coordination is therefore essential to bring these substances under a unified regulatory framework, ensuring that the recovery of the stratospheric ozone layer is not undermined by shifting global emissions. Accordingly, we have revised the final sentence of the abstract to 'This study underscores the need for integrated domestic regulations and international negotiations to address the increasing threat from C1-VSLs to stratospheric ozone recovery' (Line 27-29, Main text).

Line 37, non-zero ODP, what's the point?

Response: We did not intend to introduce or emphasize the concept of “non-zero ODP” in original Line 37. Our statement was meant to highlight the regional variability of ozone depletion potential associated with these compounds, rather than to make a point about the absolute magnitude.

Line 41, citation lacking, what has increased, emission, concentration, both?

Response: Thank you for pointing this out. The text has been changed to: “Their rapid growth in stratospheric chlorine burden is partially counteracting the gains achieved through the phase-out of long-lived ODSs” (Line 46-47, Main text), to avoid ambiguity.

Line 67, report national emissions?

Response: The national emissions referred to in Line 67 are based on atmospheric inversion results. We have revised the text to ‘China is estimated to account for 30-60% of global CH₂Cl₂ and 25-50% of global CHCl₃ emissions, as derived from atmospheric inversion studies^{13,14}’.

Line 86 consider clearer language “emissions reach xxx in the BAU scenario without additional controls.”

Response: We have revised the sentence to clarify that the projected emissions occur under the BAU scenario without additional controls. The revised sentence now reads as follows:

Line 98-102 (Main text): Under the Business-As-Usual (BAU) scenario, CH₂Cl₂ emissions are projected to reach 966 (727–1204) Gg in 2035 in the absence of stringent regulatory interventions. Conversely, the Best Practicable Technology (BPT) scenario, which assumes the gradual phase-out of CH₂Cl₂ use in flexible PU foaming, adhesives,

and coatings, alongside enhanced by-product management (see Methods and Text S2 for details), demonstrated that substantial mitigation is achievable (Figure S2)

Line 97 “from restrictions in those processes”? Also on that line, the wording isn’t clear, is the potential ban being discussed directed toward PU foaming or on use of CH₂Cl₂ in PU foaming?

Response: We have revised the sentence to clarify that the exemption refers specifically to those industrial cleaning processes in which CH₂Cl₂ remains essential. In addition, the wording regarding PU foam has been revised to explicitly indicate that the potential prohibition applies to the use of CH₂Cl₂ in flexible PU foaming processes, rather than to flexible PU foaming itself. The revised sentence now reads as follows (**line 109-115, Main text**):

Notably, under the BPT scenario, industrial cleaning is expected to emerge as the dominant source by 2030, as CH₂Cl₂ remains essential in certain industrial cleaning processes and is therefore likely to be exempt from restrictions²². Furthermore, although the application of CH₂Cl₂ as a blowing agent during flexible PU foam production has not yet been formally prohibited, its inclusion in the *Draft for Comments on the List of Key Controlled Emerging Pollutants (2021 Edition)*²³ by the Ministry of Ecology and Environment of China signals a clear trajectory toward a future regulatory phase-out.

Line 105, future tense used in a way that suggests this reduction is likely to happen. Are there laws in place that will facilitate this? Or is instead are emission reductions possible if these practices were adopted?

Response: We have revised the sentence to clarify that these results are specific to the BPT scenario, which assumes stringent future regulations exceeding baseline expectations, contingent upon technical possibilities. The revised text (now in **Lines 122–125**) reads as follows: Under this scenario, the pulp and paper industry holds the potential to be the principal driver of CHCl₃ emission abatement, contingent upon the synergistic adoption of advanced bleaching technologies and optimized operational practices, alongside enhanced wastewater treatment, and other advanced control measures²⁴. Additionally, we have systematically reviewed other relevant sectors and provided supplementary justifications for future emission reduction pathways. Detailed information can be found in the **SI Text S2**.

Figure 4, it appears that the feedstock process for CH₂Cl₂ is incorrectly indicated as producing HCFC-22 as opposed to HFC-32 (the latter being mentioned in the text, lines 50 and 197). Mention of HCFC-22 on line 37 of the SI is also confusing, as HCFC-22 is not involved with CH₂Cl₂ and HFC-32 production, is it?

Response: Thank you for pointing this out. This was a labeling error. We have corrected Figure 4 accordingly and have also revised the text in the Supplemental Information to consistently refer to HFC-32. The terminology is now consistent throughout the manuscript.

Line 173, unclear on meaning of text: “emissions from these sectors occur simultaneously” as it doesn’t imply immediate benefits. Do you mean emissions occur shortly after use?

Response: Thank you for pointing this out. We agree that the original wording was unclear. Our intention was to emphasize that emissions from these sectors occur rapidly after use, implying that emission reductions would lead to near-immediate benefits. We have revised the text to ‘Emissions from the primary applications of C1-VSLs occur rapidly after use, so mitigation actions can lead to near-immediate reductions in atmospheric releases’ in line 210-212 (Main text).

In paragraph S1 text, some background info is lacking, is it that production reporting for CH₂Cl₂ and CHCl₃ separately was considered less reliable than what you have done? Also lacking are sources or citations for HCFC-22 production amounts in the past that are projected to forward years.

Response: We have provided a more comprehensive explanation and included the specific data sources for HCFC-22 production to further enhance clarity and traceability. The method for CH₂Cl₂ and CHCl₃ production data reconstruction is now shown in Text S5:

Line 139-153 (SI): We recalculate the value of CH₂Cl₂ production and CHCl₃ production²³ based on downstream application data, as the reported values of individual CH₂Cl₂ and CHCl₃ production are not sufficiently reliable. Considering that CHCl₃ is primarily used in HCFC-22 production and, to a lesser extent, in the pharmaceutical industry, we estimate CHCl₃ consumption based on its major downstream applications as well as CHCl₃ import and export. HCFC-22 production has been historically regulated under the Montreal Protocol and is subject to mandatory annual reporting, rendering the corresponding production data relatively reliable. Therefore, CHCl₃

consumption for feedstock use can be inferred from reported HCFC-22 production (from the Chinese Hydrochlorofluorocarbons Production Phase-out Management Plan by the Ministry of Ecology and Environment and ChinaIOL²⁴). By additionally accounting for pharmaceutical uses, which represent approximately 8–10% of total CHCl₃ consumption⁵, the total CHCl₃ production can be estimated. Given the combined production of CH₂Cl₂ and CHCl₃, the remaining fraction after subtracting CHCl₃ is attributed to CH₂Cl₂ production. This method anchors calculations to well-documented downstream applications, improving the reliability and consistency of production estimates.

SI line 33, I'm not clear on the point of "required average annual compound growth rate..."

Response: Thank you for pointing this out. We have removed the extra word and revised the phrase to "required average annual growth rate" in the revised manuscript.

Regarding N₂O and use of the term ODS. In the ozone science community, the term ODS is typically reserved for Montreal-Protocol controlled gases, and N₂O and VSLs are not controlled and therefore not labeled as such. Consider replacing with "chemical influencing ozone concentrations or ozone depletion" or something else.

Response: Thank you for this helpful clarification. In the revised manuscript, we have avoided using ODS for N₂O and VSLs and replaced it with closely related descriptions.

Standard of English usage throughout the manuscript is quite good and typically doesn't limit understanding, though there are many small errors related to verb tense, use of definitive articles, or number agreement between subject and verb that could be addressed.

Response: We thank the reviewer for this positive assessment of the manuscript's English. We have carefully reviewed the manuscript and corrected minor errors related to verb tense, definite articles, and subject–verb agreement throughout to improve clarity and readability.

Response to Editor and Reviewers

We extend our gratitude to the editor and reviewers for their meticulous review and insightful suggestions, which have undoubtedly enhanced the quality of this paper. We sincerely appreciate their valuable contributions. Our response can be found below, with the Lines in the response message referring to the Clear-version of the revised manuscript.

Reviewer #1 (Remarks to the Author):

My previous concernings were well addressed. The following two further comments from the response are provided as below:

1. The authors acknowledge in their response that the time-invariant treatment of natural CHCl₃ sources may explain part of the gap with top-down estimates. This is an important limitation. I recommend explicitly stating it in the main text (e.g., in the Discussion or a dedicated Uncertainty section), rather than only in the response and the Supplementary Information.

Response: We have added text reads as follows in line 125-128 of the main text:

This discrepancy may be partly attributed to our simplified, time-invariant treatment of natural sources, particularly in years when natural emissions are expected to vary in response to meteorological or biogeochemical conditions¹³. Further analysis of the discrepancy is provided in Text S1 and Figure S1.

2. The authors have clarified in the main text that a baseline uncertainty of $\pm 10\%$ was systematically applied to single-value emission factors. To improve reproducibility, it would be helpful to clearly indicate in Supplementary Text S6 which specific sectors adopt this default uncertainty and where it applies.

Response: To ensure clarity, explicit explanatory text has been integrated into both the main text and Supplementary Text S6, accompanied by cross-references to Supplementary Tables S1–S3 for comprehensive emission factor tracking.

Specifically, the relevant text has been updated as follows:

Main text (line 240-246): Following Velders et al.⁴⁴, emission factors supported by only a single data point were assigned a normal distribution with a baseline uncertainty of $\pm 10\%$ (defined as the standard deviation). These configurations are systematically traceable via Supplementary Tables S1–S3, where all single-value parameters are automatically subject to this $\pm 10\%$ uncertainty range. As an exception to this baseline rule, a higher uncertainty of 40% was applied to the flexible PU foam sector to account for the pronounced variability in its industrial formulations and operational practices. The rationale for this specific assignment is explained in Supplementary Text S6.

Supplementary Text S6 (line 157-160): As detailed in the Main Text (Methods), a default baseline uncertainty of $\pm 10\%$ is systematically applied to all single-value emission factor

cataloged in Supplementary Tables S1–S3, while the rationale for specific exceptions (such as the 40% uncertainty for the flexible PU foam sector) is elaborated below.

Reviewer #2 (Remarks to the Author):

I congratulate the authors for improving this manuscript, especially with regard to the transparency of their methodology. Almost all of the points raised in the original review have been addressed appropriately, with the exception of the comment on uncertainties. While the revised manuscript provides much better insight into how uncertainties are being considered, the main message that "findings reveal a sustained upward trend in historical emissions for both substances" is too strong given the large uncertainties shown in Figure 3. This could be improved by adopting a more careful wording, making it clearer to the reader that such trends are found for the best estimate only. Adding uncertainty ranges to the observed trends would also help to elucidate whether these are actually significantly different between 2010 and 2023. Once this rather small issue is addressed, I would recommend the publication the manuscript.

Response: We have revised the text in the Abstract to explicitly clarify that these upward trajectories pertain specifically to our best estimates. Furthermore, we have calculated and embedded the explicit uncertainty ranges for the net emission growth between 2010 and 2023 using standard error propagation. Due to the strict word limit of the Abstract, these detailed incremental uncertainty ranges have been embedded directly into the Results section of the main text.

Revised text in the Abstract (line 16-18):

Our best estimates indicate an upward trend in historical emissions for both substances, with CH₂Cl₂ emissions more than doubling since 2010, and both species are projected to grow substantially by 2035 under the Business-As-Usual (BAU) scenario.

Revised text in the Results section of the Main text:

(line 80-81) Annual CH₂Cl₂ emissions increased from 245 (187-302) Gg yr⁻¹ in 2010 to 572 (453-692) Gg yr⁻¹ in 2023, representing a net growth of 327 (194-460) Gg yr⁻¹ (Figure 1 and S1).

(line 111-112) Total anthropogenic CHCl₃ emissions increased from 60 (42-78) Gg yr⁻¹ in 2010 to 79 (47-111) Gg yr⁻¹ in 2023, with a total increment of 19 (-18-56) Gg yr⁻¹ (Figure 1 and S1).

Reviewer #3 (Remarks to the Author):

The authors have done a thorough job of addressing the review comments. Only a few minor issues remain, see below.

Abstract: lines 25-27, “would exceed” is not very useful without a time indicated, I’d suggest you mention the approximate date or “by the end of that interval” or “by 2035”.

Response: Thank you for the helpful suggestion. To avoid the ambiguity caused by "since 2024," which could be misinterpreted as modifying the HCFC emissions, we have rephrased the sentence to clearly indicate the starting point of this trend.

Revised text in the Abstract (line 20-22):

Crucially, projections indicate that the CFC-11 equivalent emissions of CH₂Cl₂ in China would exceed the country’s aggregate emissions of all hydrochlorofluorocarbons (HCFCs) annually from 2024 onwards under the BAU scenario.

Reconsider the point in the first part of the discussion section. I read the figure to show that total HCFC emissions (from Wu et al., were comparable to CH₂Cl₂ emissions ever since 2015, not just in 2020.

Response: We thank the reviewer for pointing out this distinction. The reviewer is correct that CH₂Cl₂ emissions had already become comparable to the bottom-up HCFC estimates (e.g., Wu et al.) around 2015. However, the specific value cited in this sentence (8.5 ± 1.3 Gg yr⁻¹ CFC-11-equivalent) represents the higher constraints derived from top-down studies, which CH₂Cl₂ emissions did not catch up to until around 2020.

To eliminate the ambiguity caused by the mixed citations in the previous version, we have revised the text to explicitly specify that this comparison is made against the top-down estimates of HCFC and have adjusted the citations accordingly.

The revised text in the Discussion section now reads as follows (line 170-173):

Notably, China’s estimated CH₂Cl₂ emissions in 2020 [7.6 (5.6-9.5) Gg yr⁻¹ CFC-11-equivalent] were already comparable to the national combined emission of HCFC-22, HCFC-141b, and HCFC-142b (8.5 ± 1.3 Gg yr⁻¹ CFC-11-equivalent) from top-down estimates²⁹⁻³¹ and are projected to surpass this aggregate value from 2024 onward under the BAU scenario (Figure 3).

Line 60-61 on the relative contribution of CH₂Cl₂ anthropogenic emissions to total. The 90% contribution was cited from a 2006 report and anthropogenic emissions have increased 2 to 3 fold since potentially suggesting that the relative contribution of this potential natural source is 2 to 3 times smaller now compared to an estimate made then. Also, see the updated discussion in Claxton et al., 2020

Response: We have revised the text to incorporate this dynamic scaling: (Lines 55-57) Historical assessments attributed approximately 90% of global CH₂Cl₂ emissions to anthropogenic activities¹⁷, and recent substantial increases in industrial growth have further diluted the relative contribution of potential natural sources¹².

Apologies for missing the top-down results that appeared in Figure S1. To ensure others don't do the same, I'd highly recommend that you call out figure S1 on line 85 in the Results section, and you could move forward that call-out for CHCl₃ from line 132 to line 130.

Response: We thank the reviewer for this constructive suggestion to enhance the visibility and accessibility of top-down results. We have appropriately adjusted the positioning of the Figure S1 call-outs within the Results section.

Revised text for Figure S1 call-out (for CH₂Cl₂ emission):

(Lines 81-83) The historical CH₂Cl₂ emissions are broadly consistent with the top-down constraints from An et al.¹⁴, but exceed the bottom-up estimates of Feng et al.²⁰ (Figure S1).

Revised text for Figure S1 call-out (for CHCl₃ emission):

(Lines 123-125) Our inventory shows a gentle increase in annual total CHCl₃ emissions, whereas top-down estimates exhibit a rapid rise followed by a sharp decline¹³ (Figure S1).

Consider rewording phrasing suggesting “scenario could achieve...” scenarios don't achieve things, but “In scenario xxx, we estimate that ...” makes clear the point you are making.

Response: We thank the reviewer for pointing out this conceptual inaccuracy and have rephrased the sentence as follows: In the BPT scenario, we estimate that a cumulative CHCl₃ emission abatement of approximately 7% (~1×10² Gg) could be achieved during 2024–2035 compared to the BAU trajectory. (Lines 115-117)

Citations, reconsider citing WMO reports. It is much more appropriate to cite lead authors names for the appropriate chapter. The only time a reference to WMO (2022) or WMO (2021) is appropriate, for example, is if you are referring to the content of the entire report.

Response: We have systematically revised the reference list and text. All citations previously attributing specific data or parameter values to "WMO (2022)" or "WMO (2018)" have been updated to cite the lead authors of the respective chapters.

Examples of these corrected citations include:

James B. Burkholder & Øivind Hodnebrog. Summary of Abundances, Lifetimes, ODPs, REs, GWPs, and GTPs. in *Scientific Assessment of Ozone Depletion: 2022, ANNEX* (Geneva, Switzerland, 2022).

Johannes C. Laube & Susann Tegtmeier. Update on Ozone-Depleting Substances (ODSs) and

The discussion of IOD and VSLS lifetimes in the stratosphere vs troposphere is improved, but is incomplete without a mention of the $\tau_{\text{strat}}/\tau_{\text{trop}}$ value you used. This would allow a straightforward comparison and to the value suggested for CH₃Cl elsewhere—make it easy on the reader so they don't have to dig up other literature to know how the different values you are discussing relate to one another. I'd also suggest you include a call-out to the methods section (or SI) on line 180.

Response: First, we would like to clarify a clerical error in the previous version of the manuscript: the ratio $\tau_{\text{atmos}}/\tau_{\text{strat}}$ was inadvertently mislabeled as $\tau_{\text{strat}}/\tau_{\text{trop}}$ in several instances. This terminology has now been thoroughly corrected throughout the text.

Regarding the reviewer's core suggestions, we have expanded the discussion in **Supplementary Text S3** to explicitly state the values for τ_{tropo} , τ_{strat} , and τ_{atmos} value, alongside their corresponding ratios.

The revised text in **Supplementary Text S3** now reads as follows (line 120-131):

The ratio of $\tau_{\text{atmos}}/\tau_{\text{strat}}$, which serves as the basis for calculating IOD, as recommended in Pyle et al.¹⁹, would then be similar for CH₂Cl₂ and CHCl₃. Thus, we believe it is appropriate to use CH₂Cl₂ as a proxy for the calculation of IOD for CHCl₃. Their emission levels in 2010 are set as the baseline for IOD calculation. Specifically, the values used for τ_{tropo} , τ_{strat} , and τ_{atmos} for CH₂Cl₂ are 180 days²¹, 1–2 years²², and 142 days¹⁹, respectively.

However, we noticed that the $\tau_{\text{atmos}}/\tau_{\text{strat}}$ ratio reported for CH₂Cl₂ (20-39%) by Pyle et al.¹⁹ exceeds that of longer-lived species such as CH₃Cl (3%, as documented in the SPARC report²³). This discrepancy likely reflects the high sensitivity of very short-lived species to model-specific transport schemes and the potential inclusion of product gas injection. However, we are unable to fully account for this source of uncertainty within the simplified IOD framework utilized in our manuscript.